Resonant Energy Transfer in ultracold Rydberg gases

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Resonanter Energietransfer in ultrakalten Rydberg-Gasen

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Introduction

Rydberg atoms are atoms in highly-excited electronic states close to the ionization continuum which display a number of unusual properties. To explain those properties, it is instructive to look at Rydberg atoms in a semi-classical picture: One electron is located on an vastly extended orbit far from the nucleus, maybe comparable to an outer planet in the solar system. Therefore, the negative charge of the electron is separated far from the positive charge of the nucleus, which gives rise to large dipole moments. Furthermore, the outer electron which is only weakly bound to the nucleus is very sensitive to external electric fields, resulting in a large polarizability of the Rydberg atoms. Since the non-excited inner electrons of the atom are on trajectories close to the nucleus, they are hardly resolved from the far out-lying outer electron, which "sees" the inner electrons and the nucleus as a point-like charge. Hence, Rydberg atoms can be treated to a very good approximation as hydrogen-like system, which greatly simplifies the quantum mechanical description and allows to transfer properties from the hydrogen atom such as scaling laws. The energy of a level with principal quantum number $n$ and angular momentum $l$ can be calculated by the simple formula

$$E_{nl} = -\frac{Ry}{(n - \delta_l)^2},$$

where $Ry$ denotes the Rydberg constant. The formula is the same as for the hydrogen atom except for the so-called quantum defect $\delta_l$ that accounts for the small overlap of the outer electron’s wave function with those of the inner electrons [Gal94]. The orbital radius, i.e. the extent of the wave function of the electron, is found to increase as $n^2$, so that it easily reaches dimensions of more than ten thousand Bohr radii for $n \approx 75$, so that a single atom can reach the size of a large molecule. The polarizability of Rydberg atoms even scales as $n^7$, which is not only important in external electric...
fields, but also leads to strong interactions \textit{e.g.} of the van der Waals-type between the atoms. The lifetime of Rydberg atoms is on the order of many microseconds which is very long compared to "normal" excited atoms, so that it is possible to create gases of Rydberg atoms that are stable with respect to spontaneous decay over long timescales.

Considering these exaggerated properties it is not surprising that Rydberg atoms have attracted the interest of physicists for decades. Early experiments demonstrated the sensitivity of Rydberg atoms to electric fields via the Stark effect and field ionization [Zim79]. In beam experiments, interactions between Rydberg atoms were studied via collisions [Saf81, Sto87]. All these experiments were performed in the regime of "hot" Rydberg gases, where the kinetic energy of the atoms is much larger than their interaction energy, so that the interactions can be considered a small perturbation to the thermal motion of the atoms.

Advances in laser cooling techniques in the last decade gave access to the completely new regime of ultracold Rydberg gases, where the situation is reversed: At temperatures around 100 $\mu$K, that are easily reached \textit{e.g.} with magneto-optical atom traps, the atoms can be considered essentially static during typical interaction times, so that the interactions between the atoms fully govern the dynamics of the gas, which includes both the motional and electronic degrees of freedom of the atoms.

A whole series of recent experiments has revealed many interesting phenomena such as the formation of Rydberg molecules [Far03], the conversion of interaction energy to atomic motion [dO03, Li05], which can then lead to the ionization of a Rydberg gas by Rydberg-Rydberg collisions [Mis03], eventually triggering the evolution of cold Rydberg gases into cold plasmas and the reverse [Gal03]. It was also shown that interactions between Rydberg atoms can induce a local suppression of excitation [Sin04b, Ton04, CL05].

A particularly rewarding point is the detection of coherence in the interaction processes between Rydberg atoms. In case of sufficiently strong interactions, one atom interacts with several or even many surrounding atoms, so that an entangled many-body state is created, where the atoms behave in a collective fashion. A detection of such coherent many-body states would put ultracold Rydberg gases in a line with other strongly correlated systems such as Bose-Einstein condensates [Ket99] or atoms in high-finesse resonators [Mes85]. Coherent interactions have become
of particular interest in the context of proposed schemes for quantum information processing using neutral atoms [Bre99, Jak99] which have the advantage of long coherence times due to the weak couplings to the environment. Rydberg atoms combine this long coherence time with strong, long-range interactions, which are crucial for realizing fast quantum gates [Jak00]. All the proposed schemes rely on the control of coherent interactions between Rydberg atoms resp. mesoscopic ensembles of Rydberg atoms [Luk01], so that experiments towards coherent control of Rydberg-Rydberg interactions are an important prerequisite for the realization of quantum computing with Rydberg atoms. A recent feasibility study done by our group [RL05] gives a detailed overview over the different types of interactions in ultracold Rydberg gases and their prospects for quantum information processing.

The most promising candidate are Rydberg gases where the atoms interact via resonant dipole-dipole interactions: Here, two atomic pair states are energetically degenerate, e.g. two atoms in a P-state have exactly the same total energy as if one of the atoms was in an energetically lower-lying S-state and the other one in a different S-state of higher energy than the P-state (P and S denote the angular momenta). In such a situation the atoms acquire a permanent dipole moment, which leads to particularly strong and long-reaching interactions between the atoms. Such strong interactions evoke a redistribution of the internal energies of the atoms in so-called resonant energy transfer processes or “ Förster” processes [För96], where pairs of atoms spontaneously change their electronic states. In the above mentioned example, a pair of atoms initially both in the P-state can transform to a pair where each of the atoms is in one of the two different S-states. The process is resonant, since one of the atoms gains exactly the amount of energy in internal excitation that the other atom loses. Investigations on resonant energy transfer revealed that such strongly interacting systems must indeed be described in terms of many-particle states [And98, Mou98, Aku99, And02].

Resonant energy transfer processes in Rydberg gases follow a well approved principle employed e.g. in biological systems as an elegant means of energy transport, which does not rely on the physical displacement of energy carriers, making it fast and efficient. The basic process is similar: A donor molecule is deexcited to a lower-lying energy level, while, in a correlated event, an acceptor molecule gains exactly the amount of energy lost by the donor, thus transporting the energy from the donor to the acceptor molecule (a detailed overview over resonant energy transfer in
molecular systems is given in [Sch03]). Through cascades of such processes, complex
networks for energy transport can be realized. A prominent example is photosynthesis [Rit02], where the energy of the incoming light is gathered in many so-called
light-harvesting complexes, from which it is transferred via resonant energy transfer
processes to a reaction center where the actual synthesis of the energy carrier
ATP takes place. There have even been speculations that nature may make use of
coherent processes to increase the efficiency of the energy transport in photosyn-
thesis [Orr99]. This brings the focus back to the much simpler Rydberg systems,
which contain the "essence" of the resonant energy transfer and may therefore serve
as a model system in future experiments with which more complex phenomena of
resonant energy transfer can be investigated.

In this thesis I will present simulations and experiments on the dynamics of res-
onant energy transfer processes in randomly distributed clouds of Rydberg atoms.
The results must be seen in the context of other recent achievements of our group:
The coherent excitation of mesoscopic ensembles of Rydberg atoms was demon-
strated by the first-time observation of Rabi flops between ground state and Rydberg
atoms [Dei05b,RL06], so that we can now coherently control the excitation process.
Other experiments focussed on interaction-induced ionization processes [Amt06],
which have to be suppressed in order to preserve coherence in interaction pro-
cesses between Rydberg atoms. The work on the resonant energy transfer pro-
cesses [Wes05b] is thus one further step towards coherent control of Rydberg atoms,
which would open up fascinating perspectives in various fields.

In the first chapter I will introduce resonant energy transfer processes in more
detail and provide the necessary framework for a quantum mechanical description.
Chapter 2 presents a model of resonant energy transfer based on the coherent evolu-
tion of ensembles of up to ten atoms. By performing an average over many different
realizations of such subsystem, resonant energy transfer processes in larger clouds of
Rydberg atoms can be simulated. In the third chapter I will present experiments on
the dynamics of resonant energy transfer and compare the results of the simulations
to the experimental data. A detailed discussion of error sources is included. In the
last chapter, I will discuss possible improvements of the experiment on resonant en-
ergy transfer. Finally, I will conclude with an outlook on future experiments which
illustrate the prospects provided by Rydberg systems.
Chapter 1

Resonant energy transfer between Rydberg atoms

1.1 Interactions between Dipoles

The interaction energy of a two classical dipoles is given by [Jac75] \(^1\)

\[ E = \frac{\vec{\mu}_1 \cdot \vec{\mu}_2}{|\vec{R}|^3} - 3 \frac{\left( \vec{\mu}_1 \cdot \vec{R} \right) \left( \vec{\mu}_2 \cdot \vec{R} \right)}{|\vec{R}|^5}, \] (1.1.1)

where \(\vec{\mu}_1\) and \(\vec{\mu}_2\) denote the dipole moments and \(\vec{R}\) the distance vector between the dipoles. In quantum mechanics the dipole moments is replaced by the dipole matrix element between the states \(|\phi_1\rangle\) and \(|\phi_2\rangle\) which is defined as

\[ \vec{\mu} = \left\langle \phi_1 | \hat{\vec{r}} \right| \phi_2 \rangle = \left\langle \phi_1 | e \hat{\vec{r}} | \phi_2 \rangle, \] (1.1.2)

where \(\hat{\vec{r}}\) denotes the position operator and \(e\) the elementary charge. As \(\hat{\vec{r}}\) has negative parity, \(|\phi_1\rangle\) and \(|\phi_2\rangle\) must have opposite parity in order to have a non-vanishing dipole moment. As pure atom states have a defined parity they can never have a dipole moment (\(|\phi_1\rangle = |\phi_2\rangle\)). Moreover, the Wigner-Eckart-Theorem imposes further restrictions on the angular momenta of the involved states. As \(\hat{\vec{r}}\) is a tensor of rank one the angular momenta of \(|\phi_1\rangle\) and \(|\phi_2\rangle\) must differ by 0, 1 or -1. Since states with equal angular momenta have the same parity, only states whose angular

\(^1\)In this thesis all expressions are given in atomic units, unless otherwise stated.
Chapter 1. Resonant energy transfer between Rydberg atoms

momenta differ by 1 or -1 yield a non-zero dipole moment. Thus, the dipole operator
only connects S- and P-states, P- and D-states, etc. The evaluation of dipole matrix
elements between atomic states is described in Appendix A.

The dipole-dipole interaction can be described in a simple quantum mechanical
picture involving two atoms \( a_1 \) and \( a_2 \) separated by \( \vec{R} \) in the states \( |\psi_{1,i}\rangle \) and \( |\psi_{2,i}\rangle \)
that are connected via non-vanishing dipole matrix elements to the states \( |\psi_{1,f}\rangle \) and
\( |\psi_{2,f}\rangle \). The two relevant atom pair states are then

\[
|\Psi_i\rangle = |\psi_{1,i}\psi_{2,i}\rangle = |\psi_{1,i}\rangle_{a_1} \otimes |\psi_{2,i}\rangle_{a_2} \tag{1.1.3}
\]

and

\[
|\Psi_f\rangle = |\psi_{1,f}\psi_{2,f}\rangle = |\psi_{1,f}\rangle_{a_1} \otimes |\psi_{2,f}\rangle_{a_2} , \tag{1.1.4}
\]

with the dipole-dipole interaction matrix element being

\[
\langle \Psi_f \| \vec{V} \| \Psi_i \rangle = \frac{\langle \psi_{1,f} \| \hat{\vec{\mu}} \| \psi_{1,i} \rangle_{a_1} \cdot \langle \psi_{2,f} \| \hat{\vec{\mu}} \| \psi_{2,i} \rangle_{a_2}}{|\vec{R}|^3} \\
-3 \frac{\left( \langle \psi_{1,f} \| \hat{\vec{\mu}} \| \psi_{2,i} \rangle_{a_1} \cdot \vec{R} \right) \left( \langle \psi_{2,f} \| \hat{\vec{\mu}} \| \psi_{2,i} \rangle_{a_2} \cdot \vec{R} \right)}{|\vec{R}|^5} \tag{1.1.5}
\]

In principle, the distance vector \( \vec{R} \) is a quantum mechanical operator as well. However, as \( \vec{R} \) is almost exclusively determined by the positions of the atomic cores of the two atoms which are very well localized it can be considered a classical vector. It is convenient to work in a spherical basis in order to evaluate (1.1.5) as the dipole matrix elements \( \langle \psi_{1,f} \| \hat{\vec{\mu}} \| \psi_{1,i} \rangle \) and \( \langle \psi_{2,f} \| \hat{\vec{\mu}} \| \psi_{2,i} \rangle \) are then most easily evaluated (see Appendix A). A vector operator \( \vec{A} = (A^x, A^y, A^z) \) in cartesian coordinates can be written in spherical coordinates as

\[
A^+ = \frac{1}{\sqrt{2}} (A^x + iA^y), \quad A^0 = A^z, \quad A^- = \frac{1}{\sqrt{2}} (-A^x + iA^y). \tag{1.1.6}
\]

The scalar product between two vector operators \( \vec{A} \) and \( \vec{B} \) thus becomes

\[
\vec{A} \cdot \vec{B} = -A^+ B^- - A^- B^+ + A^0 B^0 . \tag{1.1.7}
\]
Using the notation
\[ \mu_1^{+, -, 0} = \langle \psi_1, f | \hat{\mu}_1^{+, -, 0} | \psi_1, i \rangle_{a_1}, \]
\[ \mu_2^{+, -, 0} = \langle \psi_2, f | \hat{\mu}_2^{+, -, 0} | \psi_2, i \rangle_{a_2}, \]
the two parts of (1.1.5) can be written as
\[ V_1 = \frac{-\mu_1^+ \mu_2^- - \mu_1^- \mu_2^+ + \mu_1^0 \mu_2^0}{|\vec{R}|^3}, \]
and
\[ V_2 = -3 \left( -\mu_1^+ R^- - \mu_1^- R^+ + \mu_1^0 R^0 \right) \left( -\mu_2^+ R^- - \mu_2^- R^+ + \mu_2^0 R^0 \right) \frac{1}{|\vec{R}|^5}. \]

It is interesting to note that the term \( V_1 \) conserves the sum of the internal angular momenta of atom 1 and 2, while \( V_2 \) can transform internal angular momentum into orbital angular momentum of the combined system of the two atoms which can be understood the following way: Spherical "plus"-operators increase and spherical "minus"-operators decrease the projection \( m_j \) of the angular momentum by one, while the "zero"-component leaves it constant, as can be seen from the Wigner-Eckart-Theorem. In \( V_1 \) only terms such as \( \mu_1^+ \mu_2^- \) occur, where \( \mu_1^+ \) increases \( m_j \) of atom 1 and \( \mu_2^- \) decreases \( m_j \) of atom 2 both by one, thus leaving their sum constant. In \( V_2 \) however, there are terms such as \( \mu_1^+ \mu_2^+ R^+ R^+ \) present which decreases the \( m_j \)-values of both atom 1 and 2 by one. This is compensated by \( R^+ R^+ \) which in principle represents the quantum mechanical operator for the distance between the two atoms, thus increasing the projection of the orbital angular momentum of the combined system by \( 2\hbar \). Since the center of mass motion of the two atoms is in good approximation a classical system (\( \vec{R} \) is assumed to be a classical vector) this influence can be neglected.

Using (1.1.10) and (1.1.11) one can evaluate the dipole-dipole interaction matrix element between any two states of an atom pair.

1.2 Dipole interactions between Rydberg atoms

1.2.1 Resonant vs. off-resonant Dipole interactions

Consider a three-level system consisting of the states P, S and S’ labelled according to their angular momenta. There are non-vanishing dipole matrix elements between
Chapter 1. Resonant energy transfer between Rydberg atoms

P and S and between P and S′ (see section 1.1). The energy of S is $\Delta - \delta/2$ below the energy of P, whereas S′ lies by $\Delta + \delta/2$ above P (Fig. 1.1a). Thus, the atom pair states $|SS′\rangle$ and $|S′S\rangle$ are energetically separated from the state $|PP\rangle$ by $\delta$. Assuming $\Delta \gg \delta$ it is appropriate to reduce the system to these three states, since states such as $|SS\rangle$ or $|SP\rangle$ are energetically far detuned. The reduced basis hence consists of $|PP\rangle$, $|SS′\rangle$ and $|S′S\rangle$, with $|PP\rangle$ interacting with the two other states via dipole-dipole interactions $C_3/R^3$ (the anisotropy of the dipole-dipole interaction shall be contained in the $C_3$-coefficient). Due to the dipole-dipole interactions the energy transfer process

$$P + P \leftrightarrow S + S′$$

(1.2.1)

can occur, i.e. two P atoms transform in an S and an S′ atom and vice versa. Hereby, one atom gains and the other one loses internal energy. In case of vanishing detuning the energy transfer is resonant, as one of the atoms gains exactly the amount of energy the other atom loses (Fig. 1.1b).

The Hamiltonian for a system of two atoms is
1.2. Dipole interactions between Rydberg atoms

\[
H = \begin{pmatrix}
0 & C_3/R^3 & C_3/R^3 \\
C_3/R^3 & \delta & 0 \\
C_3/R^3 & 0 & \delta
\end{pmatrix},
\]

(1.2.2)

where |\(PP\rangle\) is represented in the first row and column. Note that |\(SS'\rangle\) and |\(S'S\rangle\) do not interact as the relevant dipole matrix elements vanish. The (not normalized) eigenstates |1\rangle, |2\rangle and |3\rangle of \(H\) are

\[
|1\rangle = |SS'\rangle - |S'S\rangle
\]

(1.2.3)

\[
|2\rangle = |SS'\rangle + |S'S\rangle - \left(\frac{\delta R^3}{2C_3} + \sqrt{2 + \frac{\delta^2 R^6}{4C_3^2}}\right)|PP\rangle
\]

(1.2.4)

\[
|3\rangle = |SS'\rangle + |S'S\rangle - \left(\frac{\delta R^3}{2C_3} - \sqrt{2 + \frac{\delta^2 R^6}{4C_3^2}}\right)|PP\rangle
\]

(1.2.5)

with the corresponding eigenenergies

\[
E_1 = \delta
\]

(1.2.6)

\[
E_2 = \frac{\delta}{2} + \sqrt{\frac{2C_3^2}{R^6} + \frac{\delta^2}{4}}
\]

(1.2.7)

\[
E_3 = \frac{\delta}{2} - \sqrt{\frac{2C_3^2}{R^6} + \frac{\delta^2}{4}}.
\]

(1.2.8)

Several points are remarkable about this result:

- The "dark" state |1\rangle does not couple via dipole-dipole interactions and the system can hence be reduced to the two states |\(PP\rangle\) and \(1/\sqrt{2} (|SS'\rangle + |S'S\rangle)\). The dipole-dipole interaction between those states is then enhanced by a factor of \(\sqrt{2}\) compared to the interaction between |\(PP\rangle\) and |\(SS'\rangle\).

- For large detunings, i.e. \(\delta \gg C_3/R^3\), the energies \(E_2\) and \(E_3\) can be expanded as

\[
E_2 = \frac{2C_3^2}{R^6}/\delta + \delta
\]

(1.2.9)

\[
E_3 = - \frac{2C_3^2}{R^6}/\delta,
\]

(1.2.10)

so the effective energy shift scales as \(1/R^6\) with the interatomic spacing \(R\). Hence, in the off-resonant case, i.e. for large detunings \(\delta\), the interaction between the atoms is of a van der Waals-type. The interaction is repulsive for one of the states |2\rangle or |3\rangle and attractive for the other one, depending on the sign of \(\delta\).
Section 1.1.3: Resonant energy transfer between Rydberg atoms

$$R = 2 \, \mu m$$
$$R = 4 \, \mu m$$
$$R = 8 \, \mu m$$
$$R = 16 \, \mu m$$
$$R = 32 \, \mu m$$

Figure 1.2: Double-logarithmic plot of the interaction-induced energy shift $|E_3|$ vs. the detuning $\delta$ for five interatomic spacings $R$. The numbers were calculated for $|P⟩ = |32P_{3/2}⟩$, $|S⟩ = |32S_{1/2}⟩$ and $|S'⟩ = |33S'_{1/2}⟩$, which yields a $C_3$-coefficient of $943 \times 966$ a.u. The anisotropic part $V_2$ of the interaction (1.1.5) was set to zero. The four upper curves scale as $1/R^3$ for $\delta = 10$ kHz, while the four lower curves scale as $1/R^6$ for $\delta = 100$ MHz. In between, the transition from resonant dipole interactions to van der Waals-interactions occurs. It is thus possible that strong resonant dipole interactions occur between closely spaced atom pairs, while further separated atom pairs experience weak van der Waals-interactions. In the regime of resonant dipole interactions the energy shift is widely independent of the detuning, while in the van der Waals-regime the energy shift decreases as $1/\delta$, as can be seen from the expansions of (1.2.8).

- On or near resonance, i.e. for $\delta \ll C_3/R^3$, the situation is different: $E_2$ and $E_3$ can be expanded as

$$E_2 = \frac{\delta}{2} + \sqrt{2} \frac{C_3}{R^3}$$
$$E_3 = \frac{\delta}{2} - \sqrt{2} \frac{C_3}{R^3}$$

(1.2.11) (1.2.12)
so the energy shift scales as $1/R^3$ with the spacing $R$ between the atoms. A strong mixing of the states $|PP\rangle$ and $1/\sqrt{2} (|SS'\rangle + |S'S\rangle)$ occurs, so that the resulting single-atom states are superpositions of P with S and S' and thus do have a permanent dipole moment, unlike the pure atom states P, S or S'. Therefore, on resonance or near resonance the interaction is a far-reaching interaction between permanent dipoles which we call "resonant dipole interaction". A system initially in $|PP\rangle$ performs oscillations between its initial state and $1/\sqrt{2} (|SS'\rangle + |S'S\rangle)$ with a frequency proportional to their interaction strength.

- The modulus of $E_3$ represents the interaction-induced energy shift as $|E_3| = 0$ if no interaction is present ($C_3/R^3 = 0$). Note that $|E_3| \approx \frac{2C_3^2/\delta}{R^6}$ for large $\delta$ and $|E_3| \approx \sqrt{2} \frac{C_3}{R^3}$ for vanishing $\delta$. Fig. 1.2 shows this energy shift in dependence of the detuning $\delta$ for typical interatomic spacings. The transition from the regime of resonant dipole interactions where neighboring curves are separated by a factor of $2^3$ to the Van-der-Waals regime where this factor becomes $2^6$ is well visible. It is evident from Fig. 1.2 that one cannot rigorously discriminate between the two regimes since closely spaced atom pairs can still interact via resonant dipole interactions, while further separated atom pairs already experience interactions of van der Waals-type.

In a cloud of atoms dipole-dipole interactions exist between any two atoms. In chapter 2 we will investigate the dynamics of the production of $S$ atoms in a cloud of randomly distributed atoms initially all in $P$ through energy transfer (1.2.1) on resonance.

1.2.2 Tunability of the interaction

In the previous section, it was shown, that the effective energy shift caused by dipole-dipole interactions can vary over orders of magnitude depending on the detuning $\delta$ between the pair states. This detuning is usually a fixed property of the involved atomic states, and it is more than unlikely to find such energetic separations between states that the resonance condition is accidentally fulfilled. The separation of neighboring levels e.g. for Rydberg atoms around $n = 30$ is typically on the order of 50-100 GHz, compared to a detuning of few Megahertz necessary to achieve the
resonance condition for most interatomic spacings (Fig. 1.2). However, there exist series of Rydberg pair states, where the resonance condition is at least not strongly violated. One example are the states $|nPnP\rangle$ and $|nS(n+1)S\rangle$ for $n \approx 30$, where the detuning is on the order of 1 GHz.

One can now exploit the large polarizability of the Rydberg atoms, which opens up the unique possibility to tune the atomic energy levels in the required range via the Stark effect. With moderate electric fields of a few V/cm, energy adjustments of hundreds of Megahertz can be realized. This way, it is indeed possible to achieve the resonance condition. The Stark effect leads to an admixture of other states, e.g. a $P$-state acquires admixtures of $S$ and $D$. The question arises in how far the treatment of the resonant energy transfer given in the previous section is still valid, if the resonance condition $\delta = 0$ is achieved through admixture of other, not considered levels. This point is treated in detail in the doctoral thesis of Kilian Singer [Sin04a], so we restrict ourselves to the main statement: If the energy shift induced by the dipole-dipole interactions is much smaller than the energetic difference to the energy levels that are admixed by the Stark effect, the manifestation of the Stark effect in (1.2.2) is indeed only a modified detuning $\delta$. In the case of $S = |32S_{1/2}\rangle$, $S' = |33S_{1/2}\rangle$ and $P = |32P_{3/2}\rangle$ shown in Fig. 1.2, the typical interaction-induced energy shifts are below 100 MHz, while the closest energy levels that can be admixed via the Stark effect are more than 70 GHz away, so it is indeed justified to restrict the treatment to $PP$, $SS'$ and $S'S$. In this case, the pair states $|32S_{1/2}33S_{1/2}\rangle$ and $|33S_{1/2}32S_{1/2}\rangle$ are energetically separated by approx. 1 GHz from $|32P_{3/2}32P_{3/2}\rangle$ for zero electric field, but are on resonance at an electric field of 11.5 V/cm.

1.2.3 Excitation exchange processes

In the system consisting of the states $P$, $S$ and $S'$ there also exist dipole-dipole couplings between the states $|PS\rangle$ and $|SP\rangle$ resp. $|PS'\rangle$ and $|S'P\rangle$ (Fig. 1.3). These lead to the excitation exchange processes

$$P + S \leftrightarrow S + P$$

(1.2.13)

and

$$P + S' \leftrightarrow S' + P,$$

(1.2.14)
1.2. Dipole interactions between Rydberg atoms

Figure 1.3: System of three energy levels P, S and S’. The atom pair states $|PS'\rangle$, $|PS\rangle$ (black circles, a and b), $|S'P\rangle$ and $|SP\rangle$ (gray circles, a and b) are depicted schematically. In both cases a) and b) the depicted states are energetically degenerate.

which are always resonant (Fig. 1.3), independent of a potential detuning $\delta$ between the states $|PP\rangle$ and $|SS'\rangle$. The interaction is thus a far-reaching resonant dipole interaction for all $\delta$, scaling as $1/R^3$ with the interatomic spacing $R$.

It is important to note that the excitation exchange processes (1.2.13) and (1.2.14) can only occur when there are both $P$ and $S$ resp. $P$ and $S'$ atoms present and that do not change the number of $P$, $S$ and $S'$ atoms. However, they can lead to a migration of $S$ and $S'$ excitation through the cloud of $P$ atoms, which has to be taken into account when modelling the dynamics of the energy transfer process (1.2.1). While excitation exchange processes do not produce $S$ or $S'$ atoms themselves they can nevertheless affect the energy transfer in the following way as has been pointed out by [Mou98, And98]: As depicted in detail in Fig. 1.4 excitation exchange processes can lead away $S$ and $S'$ excitation, hence enabling strongly interacting atom pairs to produce multiple $S$ and $S'$ states. This is particularly important in the off-resonant case, where the interaction between all but the closest atom pairs is strongly suppressed (Fig. 1.2) so that only few accidentally closely-lying atom pairs undergo energy transfer processes and produce $S$ and $S'$ states. Excitation exchange processes, however, are always resonant, so at least for similar $C_3$-coefficients their interaction strength may easily exceed the one of the energy
Figure 1.4: Schematic description of how excitation exchange processes can enhance the production of $S$ and $S'$ atoms in a system of $P$ atoms (a) off resonance according to [Mou98, And98]: A energy transfer process (1.2.1) occurs between atoms 1 and 2 that are closely spaced by accident and therefore have a high probability to transform into $S$ and $S'$ (b) despite the detuning between $PP$ and $SS'$. Through excitation exchange processes the $S$ and $S'$ excitations are transferred to the outer atoms 3 and 4 (c+d), leaving the initial pair in $PP$ and thus free to undergo another energy transfer process (1.2.1) (e).

transfer process by orders of magnitude in the off-resonant case. Therefore, they provide an effective mechanism to lead excitation away from strongly interacting pairs, thus enhancing the production of $S$ and $S'$ atoms.
Chapter 2

Model of resonant energy transfer in a cold Rydberg gas

2.1 Previous work

Resonant energy transfer processes were first studied in collision experiments [Saf81, Sto87], where two atoms shortly interact via dipole-dipole interactions during a characteristic collision time determined by the velocity of the interaction partners. These experiments can be well described in a two-atom picture, since the short interaction time effectively prevents interactions between more than two atoms. However, such a collision picture of resonant energy transfer breaks down once the velocity of the atoms decreases towards the point where the atoms can be considered essentially static during typical timescales required for dipole-dipole interactions. In this case, interactions between atoms are only determined by the constant interatomic spacings and one can expect that an atom might be involved in interactions with several interaction partners. Signatures for many-body effects were first detected by [And98] and [Mou98] who investigated the dependence of resonant energy transfer processes on the applied electric field, \( \delta \) (sec. 1.2.1). They found the resonance to be much broader than expected which they could qualitatively explain by excitation exchange processes that are described in section 1.2.3. This is where the many-body aspect enters the description of “frozen” Rydberg gases since it is compulsory to consider systems of more than two atoms atoms to explain the enhanced signal off resonance: one closely spaced atom pair undergoes
energy transfer processes despite the detuning from the resonance (see sec. 1.2.1) and thus forms a reaction center, from where the products migrate to surrounding "spectator" atoms through excitation exchange processes. [Mou98] described those surrounding atoms in terms of a band structure of states which absorbs the products of energy transfer processes from the few closely spaced pairs in a diffusion process. The band structure in systems determined by $1/R^3$-interactions was then investigated in detail by [Aku99]. In an elegant experiment [And02] used a variant of the Ramsey interference method to directly demonstrate manifestations of excitation exchange processes. By diagonalizing the full Hamiltonian for a system of four atoms (including dipole-dipole interactions between all atoms) on and off resonance, [Low98] analyzed the energy level structure of such a subsystem, which is maybe best characterized as a precursor to a band structure. However, he restricted himself to one particular constellation of four atoms he considered typical for the system, which does not yet account for fluctuations in the positions of the atoms. Furthermore, [Low98] measured the time development of the S population produced in a resonant energy transfer $P + P \rightarrow S + S'$ in dependence of the Rydberg density, but did not exploit his theoretical framework to model it.

### 2.2 Description of model

The task of the model is to describe the dynamics of the population of S or $S'$-atoms that are produced in a resonant energy transfer $P + P \rightarrow S + S'$ in a randomly distributed cloud of Rydberg atoms. It is based on the many-body picture of [And98] and [Mou98] and extends the work of [Low98]. Unlike in most previous works a quantitative description is attempted, so that the anisotropy of the dipole-dipole interaction has to be taken into account.

Depending on the geometry and the power of the excitation lasers the number of excited Rydberg atoms is typically in the range between a couple of dozen and many thousand. It is of course impossible to calculate the full dynamics of such a many-body system, as the calculational effort increases strongly the more atoms are considered. Therefore we restrict ourselves to randomly selected subensembles of the atom cloud, for which the evolution can be calculated in a reasonable amount of time. The results are subsequently averaged over a large number of such subsystems.
The model is evaluated for the resonant energy transfer process

$$32P_{\frac{3}{2}, m_j = \frac{3}{2}} + 32P_{\frac{3}{2}, m_j = \frac{3}{2}} \leftrightarrow 32S_{\frac{1}{2}} + 33S_{\frac{1}{2}},$$

where half of the atoms are assumed to be initially in $32P_{\frac{3}{2}, m_j = +\frac{3}{2}}$ and half in $32P_{\frac{3}{2}, m_j = -\frac{3}{2}}$.

### 2.2.1 Coherent evolution of a subensemble of atoms

A basis vector $|\Psi\rangle$ of a many-body state containing N atoms at the positions $\vec{r}_1$, $\vec{r}_2$, ..., $\vec{r}_N$ can be written as a product of pure atom states

$$|\Psi\rangle = |\psi_1 \psi_2 \psi_3 ... \psi_N\rangle,$$

where $\psi_i$ denotes the state of the atom at the position $\vec{r}_i$ which can be either P, S or S’. The N-atom basis therefore in principle contains $3^N$ states. However, as all atoms are initially in the P state and the resonant energy transfer only produces S and S’ atoms as pairs, only states with equal numbers of S and S’ atoms can be populated. This considerably reduces the number of states, e.g. from 6561 to 1107 states for eight atoms or from 59049 to 8953 states for ten atoms. A complete basis set can thus be generated by taking the initial state $|PPP...PP\rangle$, plus all permutations.
Chapter 2. Model of resonant energy transfer in a cold Rydberg gas

of the state $|SS'PP... PP\rangle$, plus all permutations of the state $|SS'SS'... PP\rangle$, etc. The calculations were performed for systems consisting of four, six, eight and ten atoms.

The next step is to generate the corresponding Hamiltonian. The resonant energy transfer process (1.2.1) is possible between each two atoms in the P state and between an S and an S’ atom. Furthermore, each P atom interacts with each S atom via the excitation exchange process (1.2.13) and with each S’ atom via (1.2.14). Fig. 2.1 shows all possible interactions for a state with four P atoms and two SS’ pairs. The Hamiltonian contains non-zero entries between each two basis states that are connected via one of the processes (1.2.1), (1.2.13) or (1.2.14). With the dipole operators $\hat{\mu}_i$ and $\hat{\mu}_j$ acting on the atoms i and j at the positions $\vec{r}_i$ and $\vec{r}_j$, the interaction $\hat{H}_{ij}$ between those two atoms is

$$\hat{H}_{ij} = \frac{\hat{\mu}_i \cdot \hat{\mu}_j}{|\vec{r}_i - \vec{r}_j|^3} - 3 \left( \hat{\mu}_i \cdot (\vec{r}_i - \vec{r}_j) \right) \left( \hat{\mu}_j \cdot (\vec{r}_i - \vec{r}_j) \right) \frac{1}{|\vec{r}_i - \vec{r}_j|^5}.$$  \hfill (2.2.3)

The total Hamiltonian $\hat{H}$ can then be written as

$$\hat{H} = \sum_{i,j, i \neq j} \hat{H}_{ij},$$  \hfill (2.2.4)

where $\hat{H}_{ij}$ acts only on atoms i and j. It is important that matrix elements for basis states connected by more than one of the processes (1.2.1), (1.2.13) or (1.2.14) are zero. This can be seen with the following example: The interaction matrix element for states connected by two processes, e.g. $|PPSS'\rangle$ and $|SS'PP\rangle$, is then

$$\left\langle PPSS' \right| \hat{H}_{12} + \hat{H}_{13} + \ldots \left| SS'PP \right\rangle = \left\langle PP \right| \hat{H}_{12} \left| SS' \right\rangle \left\langle SS' \right| PP \rangle + \ldots = 0.$$  \hfill (2.2.5)

It is important to note that each of the atoms preserves the sign of its $m_j$-value during resonant energy transfer processes: An atom initially in $32P_{3/2, m_j = +3/2}$ can e.g. be transformed to $32S_{1/2, m_j = +1/2}$ as part of a resonant energy transfer process. For a next resonant energy transfer or excitation exchange process, selection rules would allow for transitions both back to $32P_{3/2, m_j = +3/2}$ and to new states $32P_{3/2, m_j = +1/2}$ and $32P_{3/2, m_j = -1/2}$. However, as a static electric field is required in experiments to achieve the resonance condition, the two latter states are detuned.
from $32P_{3/2, m_j=+3/2}$ due to the Stark effect, thus violating the resonance condition, so that they are not populated for energetic reasons. An atom initially in $32P_{3/2, m_j=+3/2}$ is therefore restricted to its initial state and $32S_{1/2, m_j=+1/2}$ (likewise atoms with $m_j = -3/2$), so that each atom can be assigned either a "plus"- or a "minus"-sign, that is preserved during the interaction process. This greatly simplifies the description of the system, as each P-state can only transform to a single S-state and vice versa (actually, this is the reason why there is no need to explicitly include $m_j$-values in the above basis set). Nevertheless, it must be known for each atom, whether it is "plus" or "minus", as the $m_j$-values determine the spherical component of the dipole matrix elements (sec. 1.1) that contribute to (2.2.3). The matrix element between two many-body states therefore depends on the combination of the signs of the $m_j$-values of the two interacting atoms.

Knowing the positions $\vec{r}_i$ and $\vec{r}_j$ of the two interacting atoms and the dipole matrix elements $\vec{\mu}_i$ and $\vec{\mu}_j$ for the states of the respective atom, the interaction matrix elements can be evaluated, which is described in detail in section 1.1. The Hamiltonian is automatically generated in a program written in Mathemetica [WRI03], that compares each two states of the basis set and evaluates the interaction matrix element between them. We then numerically solve the time-dependent Schrödinger equation in Matlab [Inc02] with the initial condition

$$|\Psi(t = 0)\rangle = |PPPP...PP\rangle,$$

yielding the time development of the probability amplitudes of all basis states. To obtain the probability for a specific atom to be in the S-state one has to add up the probability amplitudes of all states where this atom is in S. An example is given in Fig. 2.2 where the time development of a system of eight randomly placed atoms in two dimensions is shown. The resonant energy transfer first occurs between closely spaced atoms\(^1\), which initially perform fast oscillations between the states $|P\rangle$ and $1/\sqrt{2} (|SS'\rangle + |S'S\rangle)$. The oscillations are damped by the couplings to the outerlying spectator atoms whose built-up of S population is much slower. Note that the probability to be in S is always equal to the probability to be in $S'$ since the process $P + P \leftrightarrow S + S'$ is indistinguishable from $P + P \leftrightarrow S' + S$, so the maximum probability to be in S is 50% for each atom.

\(^1\)Note that the anisotropy of the dipole interaction is deactivated in this example due to the orientation of the quantization axis. See Fig.2.2.
Figure 2.2: Time development of a random atom ensemble in two dimensions. For the atoms 1-4 the probability to be in S is given in the diagrams below. The quantization axis was chosen perpendicular to the atom plane and all atoms were assumed to be initially in the $m_j = +3/2$ state, so in this special case the dipole interactions are not anisotropic and only depend on the interatomic spacings. The size of the atom ensemble is on the order of 50 $\mu$m. At $t=0$ the electric field is switched to resonance. One can clearly see fast oscillations between the closely spaced atom pairs 3/7 and 4/8, that are damped by the "spectators" 1, 2, 5 and 6, which perform oscillations on longer time scales (see text).
It is then easy to calculate the total number of atoms in the S state which is the sum over the S-probabilities of all atoms. This already flattens out parts of the oscillation and gives the curve a characteristic envelope as can be seen in Fig. 2.3 for the particular system shown in Fig. 2.2.

Fig. 2.2 and 2.3 are meant to illustrate the processes that govern the internal dynamics of a Rydberg atom cloud. The dynamics of each system considered depends strongly on the positions and relative orientations of the atoms. Nevertheless, it is already sufficient to sum over the coherent evolutions of only eight randomly placed atoms to partly wash out the oscillations predicted for each atom and to yield an envelope which features an initial rise followed by a relatively constant value with only minor long-term oscillations. As this effect increases with the number of frequencies involved it is to be expected that the curves will become smoother the more atoms are considered, despite of the coherent nature of the underlying processes.

2.2.2 Average over the realizations of the system

To model the time development of the S population for a large cloud of Rydberg atoms we average over a number of randomly chosen subsystems. For this we randomly place a thousand atoms in a box whose side length $a$ is chosen as

$$a = \sqrt[3]{n/1000^{-1}},$$

where $n$ denotes the atomic density. With this we ensure that the average density in the box corresponds to the desired density, but that local density fluctuations can nevertheless occur as the atoms are randomly placed. Furthermore, we dice out for each of the atoms whether it is in the $m_j = +3/2$ or in the $m_j = -3/2$ state, with the probability being equal for each state. From the center of the box, the smallest possible sphere that contains four, six, eight or ten atoms is selected to determine a subsystem of atoms for which the coherent evolution can be calculated according to section 2.2.1. Although the choice of a compact spherical ensemble may be questionable due to the anisotropy of the dipole interactions (atoms do not necessarily interact most strongly with their next neighbor) this effect is still taken into account as long as the subsystem is chosen large enough. The procedure is repeated for a number of realizations of the system, i.e from 1000 realizations for subsystems of ten atoms to 4000 realizations if subsystems of four atoms are
Figure 2.3: Total number of S atoms produced in the particular system of eight atoms described in Fig. 2.2. At $t=0$ the electric field is switched to resonance. The maximum value of approximately three S atoms corresponds to a fraction of 37.5% of the atoms that are in the S-state. The inset shows the shapes of the S-amplitudes for the atoms 1 to 8 that yield the above curve when summed up. This summation process considerably reduces the amplitude of the oscillations, that are visible for the single atoms, relative to the maximum signal.

considered. The time development of the S population is then averaged over these subsystems.

There are two possible interpretations that justify this procedure from a physical point of view: First, if one decomposes the atom cloud in a number of non-interacting subsystems, all interactions between the atoms of different subsystems are clearly neglected. However, the errors made hereby decrease the larger one chooses the subsystems considered. They have to be sufficiently large that edge effects originating from clipping the interactions with neighboring subsystems do not dominate the dynamics.

Second, in the experiment one has to average the signal over a number of experi-
mental cycles in order to improve the signal-to-noise ratio. In each cycle a new cloud of cold atoms with random atom positions is produced. This means that experimental data are also obtained as averages over various realizations of the system, just as it is done in the model. This is particularly important if one goes to very low atom numbers (smaller than 50) where one might indeed expect to see remnants of oscillations in the population of S atoms if it was possible to follow the time development of one particular realization. This is of course impossible as the measurement of the S population at a certain interaction time is destructive (chapter 3). Thus, even at low atom numbers one would experimentally obtain an average over various realizations of the system. To overcome this problem the atoms would have to be placed at defined positions, so that the same system can be repeatedly measured.

2.3 Results of the model

2.3.1 Dependence on number of atoms in subensemble

Fig. 2.4 shows the modelled development of the $S$ population for a density of $10^8$ cm$^{-3}$ depending on the number of atoms contained in the subensembles. All models initially predict a steep increase, which can be attributed to strongly interacting pairs, and eventually approach a value of approximately 0.25, which corresponds to a 50% probability for each atom to be in $P$ and respectively 25% to be in $S$ or $S'$. The $S$ population at a certain interaction time increases with the atom number since the probability to find strongly interacting atom pairs in the subsystem is enhanced if more atoms are considered. However, there is hardly any difference in the shape of the curves which is a crucial criterion for the comparison to experimental data. Moreover, the differences between the curves progressively fade out when more atoms are taken into account. One would indeed expect such a convergence behavior as the limit of averaging over increasingly larger subsystems is to evaluate the full quantum mechanical dynamics of the whole cloud of atoms. While this is impossible due to the insufficient computational resources, the curves in fig. 2.4 suggest that the essential dynamics is already contained in subsystems of six to ten atoms. For these, one can assume edge effects, i.e. atoms near the edge of the subsystem only have potential interaction partners on one side, to have only minor influence, so
Chapter 2. Model of resonant energy transfer in a cold Rydberg gas

Figure 2.4: Comparison of the modelled time development of the $S$ population obtained as average over subensembles with different atom numbers at a Rydberg density of $10^8$ cm$^{-3}$.

that the full dynamics of the cloud can indeed be reproduced by averaging over such non-interacting subsystems. In order to analyze further and also to compare to experimental data, we chose the six-atom model which is a good compromise between computation time and accuracy.

2.3.2 Dependence on the Rydberg density

From the model we find a strong dependence of the resonant energy transfer on the Rydberg density, as one can see in Fig. 2.5. The interaction strength scales as $1/R^3$ with the interatomic spacing $R$ and therefore the average interaction strength should be proportional to the Rydberg density $n$. A good measure for the average interaction strength is the initial transfer rate $\gamma$, which corresponds to the initial slope of the curves in Fig. 2.5. The proportionality between Rydberg density $n$ and initial transfer rate $\gamma$ is shown in table 2.1. One can compare the transfer rate $\gamma$ obtained from Fig. 2.5 to some sort of average transfer rate $\gamma^* = \mu\mu' n$ that is easily
2.3. Results of the model

2.3.3 Role of migration of excitation

In section 1.2.3 we described how excitation exchange processes \( S + P \leftrightarrow P + S \) and \( S' + P \leftrightarrow P + S' \) can enhance the production of \( S \) atoms in the off-resonant case, which could also be confirmed experimentally [And98, Mou98, And02]. On resonance however, the interaction strengths of the resonant energy transfer process \( P + P \leftrightarrow S + S' \) and the excitation exchange processes are approximately equal, so that the picture that can be successfully applied to the off-resonant case (sec. 1.2.3) is no longer valid. The role of excitation exchange processes on resonance is thus not clear.
Table 2.1: Comparison of the initial transfer rates of the curves depicted in Fig. 2.5. The proportionality between \( n \) and \( \gamma \) can be inferred from the third column. We define the calculated transfer rate \( \gamma^* \) as the scalar part \( V_1 \) of the interaction strength for an atom pair separated by \( R_{av} = \sqrt[3]{n^{-1}} \), i.e. \( \gamma^* = \mu \mu' / R_{av}^3 = \mu \mu' n \). The differences between \( \gamma \) and \( \gamma^* \) are due to the anisotropic part of the interaction and the random positions of the atoms.

<table>
<thead>
<tr>
<th>density ( n ) (cm(^{-3}))</th>
<th>rate ( \gamma ) ((\mu s^{-1}))</th>
<th>rate ( n/\gamma ) (10(^8) (\mu s) cm(^{-3}))</th>
<th>calc. rate ( \gamma^* ) ((\mu s^{-1}))</th>
<th>( \gamma / \gamma^* )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 3 \times 10^6 )</td>
<td>0.0112</td>
<td>2.67</td>
<td>0.0056</td>
<td>2.01</td>
</tr>
<tr>
<td>( 1 \times 10^7 )</td>
<td>0.0423</td>
<td>2.36</td>
<td>0.0185</td>
<td>2.28</td>
</tr>
<tr>
<td>( 3 \times 10^7 )</td>
<td>0.119</td>
<td>2.52</td>
<td>0.056</td>
<td>2.14</td>
</tr>
<tr>
<td>( 1 \times 10^8 )</td>
<td>0.381</td>
<td>2.63</td>
<td>0.185</td>
<td>2.05</td>
</tr>
<tr>
<td>( 3 \times 10^8 )</td>
<td>1.201</td>
<td>2.50</td>
<td>0.556</td>
<td>2.16</td>
</tr>
</tbody>
</table>

While all results shown above were obtained with excitation exchange processes included, we evaluated the \( S \) population with the six-atom model with deactivated excitation exchange processes, i.e. we only included interactions in the Hamiltonian that arise from resonant energy transfer processes. We found no difference to the curves where excitation exchange processes were allowed for a range of Rydberg densities, so excitation exchange processes do not enhance the production of \( S \) atoms if the process \( P + P \leftrightarrow S + S' \) is on resonance. Excitation exchange processes certainly do occur, but on average have no influence on the production of \( S \) atoms.

2.4 Comparison to atom pair model

The most simple way of describing the resonant energy transfer is a pair model that decomposes the cloud of atoms into non-interacting pairs each of which is allowed to undergo the process \( P + P \leftrightarrow S + S' \). Such a model attempts a description of the resonant energy transfer without including many-body effects. It is thus an important question whether such a two atom model is capable of reproducing experimental data.
As has been discussed in section 1.2.1 each of the atom pairs will feature Rabi-like oscillations between the states $|PP\rangle$ and $1/\sqrt{2}(|SS'\rangle + |S'S\rangle)$ with a frequency determined by the respective interaction strength. To obtain the fraction of $S$ atoms that are produced in a certain interaction time one again has to perform a Monte-Carlo average over the different orientations and interatomic spacings. In the models with four to ten atoms this has been done by choosing subensembles as spatially closed units of atoms around the center of a randomly generated atom cloud. For a two-atom model this procedure must be modified: In principle, each two atoms in the cloud can be considered a pair that can undergo a resonant energy transfer process. Therefore, it has to be decided which two atoms form a pair. For one randomly selected atom the fastest resonant energy transfer (i.e. the one with the highest oscillation frequency) will occur with the atom that interacts most strongly, making this the dominant process for this atom pair. Note that the atom interacting most strongly does not necessarily have to be the closest atom due to the anisotropy of the interaction. Selecting subsystems (i.e. atom pairs in the case of the two atom model) according to the position of the atoms, as it was done for the models with
four to ten atoms, is thus not justified. For the many-atom models the situation is different since due to the $1/R^3$ dependence of the interaction strength the atom interacting most strongly will most likely be at least one of its neighboring atoms, which has a high probability to also be contained in the subensemble if this is only chosen sufficiently large.

For the two-atom model we therefore randomly place 1000 atoms at the desired average density in a box, then take the atom closest to the center and select the atom interacting most strongly as its interaction partner. We subsequently calculate the time development of the $S$ population given by the time-dependent Schrödinger equation for this atom pair and finally average over 1000-5000 realizations of the atom cloud. Note that there is a difference between selecting only one atom pair out of the atom cloud and then averaging over many random realizations of the cloud, as we do it, and decomposing one random atom cloud in most strongly interacting atom pairs, that are averaged over. However, we checked that there is no visual difference in the results of the simulation between the two methods, so the first method was chosen for simplicity reasons.

A comparison of the results of the six and the two atom model for three different Rydberg densities is shown in Fig. 2.6. The most striking difference between the two models is the overshoot visible for all densities, which can be understood the following way: The curves are obtained as averages over oscillations with different frequencies. The range of different frequencies provided through the randomness of the position of the atoms is not sufficient for the oscillations to fully average out over the first period, thus leading to the overshoot over the steady state value of 0.25. One can also see in Fig. 2.6 that the curves for the six and the two atom model start with the same initial transfer rate. To understand this it is instructive to look at Fig. 2.2: The atoms three and four which each have a strong interaction partner start with Rabi-like oscillations just as in the two atom case. Moreover, the weakly interacting atoms one and two initially give no significant contribution to the $S$ population, which is similar to the two atom case where the small oscillation frequency of such pairs prevents a built-up of $S$ population for small interaction times. Thus, pair processes dominate for small interaction times, while for longer interaction times, many-body effects such as the damping of the amplitude of the oscillations for atom three and four through interaction with neighboring atoms (Fig. 2.2) take over in the many-body atom model, leading to significant differences in
the shape of the curves.

\section*{2.5 Role of atomic motion}

The model described in the previous sections assumed the atoms to be fixed in space. Initially, this is a very good approximation as the atoms are prepared at temperatures of below 100 \(\mu\)K. The thermal velocity of the atoms is thus around 0.1 to 0.2 \(\mu\text{m}/\mu\text{s}\), while the average spacing between the atoms is more than 10 \(\mu\text{m}\) for densities of up to \(5 \times 10^8\) \(\text{cm}^{-3}\). Thus, the displacement of the atoms due to their thermal velocity can be neglected for the time scales considered.

However, the atoms experience accelerating forces due to the electronic potentials which depend on the interatomic spacings. Reversely, any displacement of the atoms changes the electronic potentials and thus influences the dynamics of the resonant energy transfer.

As an ultimate consequence, atoms that are accelerated towards each other on an attractive \(C_3/R^3\) potential may collide. This is known to evoke a redistribution of the excitation to other Rydberg states or Penning ionization of one of the atoms [Li05, WF04]. Such atoms no longer participate in resonant energy transfer processes which reduces the number of detectable \(S\) atoms. Moreover, ions produced in collisions locally change the electric field and shift some of the atoms out of resonance, hence blocking the resonant energy transfer. This makes it necessary to assess the influence of atomic motion on the resonant energy transfer.

\subsection*{2.5.1 Forces on atoms due to electronic potentials}

A system of \(N\) atoms at the positions \(\vec{r}_1, \vec{r}_2, \ldots, \vec{r}_N\) with Hamiltonian \(H\) is supposed to be in the state \(|\Psi(\vec{r}_1, \ldots, \vec{r}_N, t)\rangle\). The force exerted on the \(i\)-th atom is minus the gradient with respect to \(r_i\) of the energy of the system, \emph{i.e.}

\[
\vec{F}_i(\vec{r}_1, \ldots, \vec{r}_N, t) = -\nabla_i \langle \Psi(\vec{r}_1, \ldots, \vec{r}_N, t) \mid H \mid \Psi(\vec{r}_1, \ldots, \vec{r}_N, t) \rangle.
\] (2.5.1)

The \(k\) eigenstates of the system are \(|\Phi_1(\vec{r}_1, \ldots, \vec{r}_N)\rangle, \ldots, |\Phi_k(\vec{r}_1, \ldots, \vec{r}_N)\rangle\) with the corresponding eigenenergies \(E_1(\vec{r}_1, \ldots, \vec{r}_N), \ldots, E_k(\vec{r}_1, \ldots, \vec{r}_N)\). Hence, the force on the \(i\)-th atom can be written as
\[ \vec{F}_i(r_1, \ldots, r_N, t) = -\sum_{j=1}^{k} \vec{\nabla}_i \left( |\langle \Phi_j(r_1, \ldots, r_N)|\Psi(r_1, \ldots, r_N, t)\rangle|^2 \cdot E_j(r_1, \ldots, r_N) \right) . \] (2.5.2)

If two atoms on resonance are considered, the eigenstates and eigenenergies are (see sec. 1.2.1)

\[ |\Phi_1\rangle = \frac{1}{\sqrt{2}} (|SS\rangle - |S'S\rangle), \quad E_1 = 0 \] (2.5.3)

\[ |\Phi_2\rangle = \frac{1}{\sqrt{2}} \left( \frac{1}{\sqrt{2}} (|SS\rangle + |S'S\rangle) - |PP\rangle \right), \quad E_2 = +\sqrt{2} \frac{C_3}{|R_1 - R_2|^3} \] (2.5.4)

\[ |\Phi_3\rangle = \frac{1}{\sqrt{2}} \left( \frac{1}{\sqrt{2}} (|SS\rangle + |S'S\rangle) + |PP\rangle \right), \quad E_3 = -\sqrt{2} \frac{C_3}{|R_1 - R_2|^3} \] (2.5.5)

If the initial state is \(|PP\rangle\) we obtain

\[ |\Psi(t)\rangle = \frac{1}{\sqrt{2}} \left( -e^{-i\frac{E_2}{\hbar}t} |\Phi_2\rangle + e^{-i\frac{E_3}{\hbar}t} |\Phi_3\rangle \right) , \] (2.5.6)

so the admixtures of \(|\Phi_2\rangle\) and \(|\Phi_3\rangle\) are equal. Thus, as \(E_2 = -E_3\) the force on the atoms vanishes in this case. We must emphasize that the effect of a vanishing force must be attributed to the somewhat artificial situation of isolated atom pairs, where the admixture of the two eigenstates is equal independent of the interaction strength and thus of the interatomic separation. For systems with more atoms there generally is a resulting force, which then induces motion of the atoms. From (2.5.1) and (2.5.5) we estimate that the potential between an atom pair cannot be much stronger than approx. \(C_3/R^3\).

As a measure for the time scale of the atomic motion we take the time \(\tau_{coll}\) that a pair of atoms initially separated by \(R_0\) in an attractive \(C_3/R^3\) potential needs to collide. The collision time can be calculated analytically with the following approach: The sum of potential and kinetic energy is constant for all times.

\[ E_{tot} = -\frac{C_3}{R^3} = \frac{1}{2} \frac{m_{Rb}}{2} \left( \frac{d}{dt} R(t) \right)^2 - \frac{C_3}{R(t)^3} , \] (2.5.7)

where \(m_{Rb}/2\) denotes the reduced mass of two Rubidium atoms. This can be transformed and integrated.
2.5. Role of atomic motion

\[ \tau_{\text{coll}} = \int_0^{\tau_{\text{coll}}} dt = \int_{R_0}^{R_0^0} \frac{1}{\sqrt{\frac{4C_3}{m_{\text{Rb}}} \left( \frac{1}{R^3} - \frac{1}{R_0^3} \right)}} dR \]  

\[ = \frac{\sqrt{\pi}}{2 \Gamma\left(\frac{2}{3}\right)} \sqrt{\frac{m_{\text{Rb}} R_0^5}{C_3}} \approx 0.37 \sqrt{\frac{m_{\text{Rb}} R_0^5}{C_3}} \]

On the other side, the rise time of the \( S \) population \( \tau_{\text{rise}} \) is on the order of the inverse interaction strength

\[ \tau_{\text{rise}} \approx \frac{R_0^3}{C_3} \]  

as has been shown in section 2.3.2. We expect the motion of the atoms to have negligible influence on the resonant energy transfer if the collision time is much larger than the rise time of the \( S \) population, \( i.e. \)

\[ \frac{\tau_{\text{coll}}}{\tau_{\text{rise}}} = 0.37 \sqrt{\frac{m_{\text{Rb}} C_3}{R_0^3}} \gg 1. \]  

It is remarkable that (2.5.10) has a very weak dependence on the atomic density \( n \). With the average interatomic spacing \( R_0 \) being the inverse of the third root of \( n \) one obtains \( \tau_{\text{coll}}/\tau_{\text{rise}} \propto \sqrt[3]{n} \). It therefore mostly depends on the \( C_3 \) coefficient of the interaction and the respective atomic mass which are fixed parameters of the system if the motion of the atoms has influence on the resonant energy transfer. For the process \( 32P + 32P \leftrightarrow 32S + 33S \) in \( ^{87}\text{Rb} \) (\( C_3 \approx 3 \times 10^5 \text{ au}, m_{\text{Rb}} \approx 1.6 \times 10^5 \text{ au} \)) and typical atomic densities between \( 10^6 \text{ cm}^{-3} \) and \( 10^9 \text{ cm}^{-3} \) we obtain \( \tau_{\text{coll}} > 50 \tau_{\text{rise}} \), so it is not expected the motion of the atoms to have a significant influence on the initial rise of the \( S \) population. It is to be emphasized that the above estimations were made for average interatomic spacings of the system. Atoms that accidentally deviate strongly from this average can indeed experience a strong influence of the atomic motion and/or collisions. However, this will only apply to a small fraction of the atoms.

2.5.2 Atom pair model for moving atoms

We investigated the most simple system, namely a resonant energy transfer process occurring between an atom pair, which experiences an accelerating force due to an attractive \( C_3/R^3 \) potential. To do so we included the motion of the atoms in the
two atom model described in section 2.4. While we have seen in the last paragraph that the force vanishes if only pairs of atoms are considered, this will nevertheless give an idea of the importance of the motion for the resonant energy transfer.

We neglect the anisotropic part $V_2$ of the interaction (1.1.5) and solve the time-dependent Schrödinger equation with a time-dependent interaction strength $V_1(t) = \mu \mu' / R(t)^3$, where $R(t)$ obeys Newton’s Law

$$\frac{m_{Rb}}{2} \frac{d^2}{dt^2} R(t) = -\frac{d}{dR} \left( -V_1(R) \right). \quad (2.5.11)$$

The second minus sign on the right side is because we require the potential to be attractive. This approach is only valid as long as the electronic wave functions of the two atoms are well separated. The orbital radius of the relevant states is on the order of $1600 \, \text{a}_0 \, \text{[Gal94]}$, so we introduced a collision radius of $5500 \, \text{a}_0$ and terminated the calculation for interatomic separations smaller than this. Once the two atoms approach within the collision radius, a state-changing or ionizing collision becomes likely and the pair does no longer participate in the resonant energy transfer process. When performing the same Monte-Carlo average as in section 2.2.2, we can take the fraction of atom pairs that have collided until a certain interaction time as a crude measure for the importance of collision processes and thus ionization in the system. Although the collision radius was assigned rather arbitrarily its exact value is not that important: At typical interatomic spacings of $>5 \, \mu\text{m}$ the atoms will start on the relatively flat part of the potential and then ”roll” down in the potential well with rapidly increasing velocity. They will spend only a small fraction of the time they take to collide at separations below $1 \, \mu\text{m}$, so the choice of the collision radius is not critical.

Fig. 2.7 shows the evolution of the $S$ population for two atom pairs with different atomic masses that undergo a resonant energy transfer process while being accelerated towards each other. The electronic state is the same for both elements, so we assumed the $C_3$ coefficients to be equal, and both pairs are initially separated by the same spacing. For $^{87}\text{Rb}$ little effect of the interaction-induced atomic motion is visible, while for the much lighter $^{23}\text{Na}$ the oscillations become faster which corresponds to an enhancement of the interaction as the atoms approach each other. Thus, a simple way to reduce unwanted effects like collisions that arise from atomic motion is to use heavier atoms such as cesium.

When we perform the same Monte-Carlo average over the interatomic spacings
Figure 2.7: Time development of the resonant energy transfer (interaction strength $C_3/R^3$) for a pair of $^{23}\text{Na}$ and $^{87}\text{Rb}$ atoms which are accelerated towards each other due to an attractive $C_3/R^3$ potential, with $C_3 = 3 \times 10^5$ au. The interatomic spacing was chosen to be 5.4 $\mu$m which leads to a collision of the sodium atoms after approx. 10 $\mu$s. The oscillation period is more and more reduced for sodium, as the interaction strength increases with the decreasing interatomic spacing. For rubidium, little effect is visible.
Table 2.2: Fraction of rubidium atom pairs that collide within a certain interaction time \( \Delta t \) if one assumes purely attractive interaction potentials. \( C_3 = 3 \times 10^5 \) au.

<table>
<thead>
<tr>
<th>density ( n ) (cm(^{-3}))</th>
<th>( \Delta t &lt; 1 )µs</th>
<th>( \Delta t &lt; 2 )µs</th>
<th>( \Delta t &lt; 5 )µs</th>
<th>( \Delta t &lt; 10 )µs</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 1 \times 10^7 )</td>
<td>0%</td>
<td>0%</td>
<td>0%</td>
<td>&lt;1%</td>
</tr>
<tr>
<td>( 3 \times 10^7 )</td>
<td>0%</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
</tr>
<tr>
<td>( 1 \times 10^8 )</td>
<td>&lt;1%</td>
<td>&lt;1%</td>
<td>1%</td>
<td>3%</td>
</tr>
<tr>
<td>( 3 \times 10^8 )</td>
<td>&lt;1%</td>
<td>1%</td>
<td>4%</td>
<td>8%</td>
</tr>
<tr>
<td>( 1 \times 10^9 )</td>
<td>1%</td>
<td>4%</td>
<td>10%</td>
<td>23%</td>
</tr>
</tbody>
</table>

as in section 2.4 we see that the initial rise of the \( S \) population remains unchanged if motion is included in the model for Rydberg densities between \( 10^6 \) cm\(^{-3}\) and \( 10^9 \) cm\(^{-3}\). As can be seen in Fig. 2.7 the motion of the atoms has negligible effect on the first couple of oscillation periods, which exclusively determine the initial rise. However, for densities of more than \( 10^8 \) cm\(^{-3}\) a non-negligible fraction of the atom pairs collide, which leads to a steady decrease of the \( S \) population. Fig. 2.8 shows a comparison of the models with and without motion for a density of \( 3 \times 10^8 \) cm\(^{-3}\). The decrease is correlated to the fraction of atom pairs that are lost due to collisions, as in the steady state roughly 25% of the atoms will be in \( S \). Table 2.2 gives an overview over the fraction of atom pairs that collide at different densities if one assumes purely attractive potentials between the atoms. The real potentials are not purely attractive as discussed above, so the numbers of table 2.2 must be understood as upper limits. Nevertheless, one can clearly infer from table 2.2 that the influence of the atomic motion on the resonant energy transfer is negligible on a microsecond time scale for densities below about \( 10^8 \) cm\(^{-3}\).

2.5.3 Influence of ions on resonant energy transfer

If \( \text{Rb}^+ \)-ions are produced they remain in the Rydberg cloud for microseconds due to their heavy mass. They locally change the electric field and can thus shift some of the Rydberg atoms out of the resonance. If an ion is produced, its electric field "freezes" all resonant energy transfer processes in a certain volume around it (which
Figure 2.8: Comparison of the time development of the $S$ population at a Rydberg density of $3 \times 10^8 \text{ cm}^{-3}$ for the two atom model described in the text with and without motion of the atoms included (left axis, lines). The initial rise is unchanged, while the curve where motion was allowed subsequently decreases due to atoms lost in collisions. The loss of $S$ atoms is roughly one forth of the collided atom pairs (green pillars, right axis), as in the steady state 25% of the atoms are in $S$. Note that the absolute numbers of the $S$ population are not comparable to Fig. 2.4, Fig. 2.6 and Fig. 2.5 as the anisotropic part of the interaction is not included here.
Table 2.3: Average number of Rydberg atoms, that are shifted out of resonance by an ion. The numbers are obtained using the probability functions of Appendix B.

<table>
<thead>
<tr>
<th>width of resonance</th>
<th>Rydberg density/ cm$^{-3}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$1 \times 10^7$</td>
</tr>
<tr>
<td>0.5 V/cm</td>
<td>$&lt; 0.1$</td>
</tr>
<tr>
<td>0.2 V/cm</td>
<td>$&lt; 0.1$</td>
</tr>
<tr>
<td>0.05 V/cm</td>
<td>0.2</td>
</tr>
</tbody>
</table>

population to be smaller, while an offset of P atoms, that cannot undergo resonant energy transfer processes, is created. If the width of the resonance of the energy transfer is known (which depends on factors such as excitation exchange processes, sec. 1.2.3), the volume in which resonant energy transfer processes are inhibited can easily be calculated. Using the probability functions defined in Appendix B, the average number of Rydberg atoms that are shifted out of resonance by an ion can be calculated, which is shown in table 2.3. For small Rydberg densities or broad resonances, ion densities comparable to the Rydberg density are necessary to evoke a significant influence on the resonant energy transfer. For higher Rydberg densities or narrow resonances however, each ion inhibits the resonant energy transfer for many surrounding Rydberg atoms, so that few ions are sufficient to significantly reduce the number of Rydberg atoms available for resonant energy transfer processes.

2.6 Spontaneous decay of the atoms

The lifetimes of the involved states $32P$, $33S$ and $33S$ are $\tau_{32P} = 30\mu s$ and $\tau_{32S} \approx \tau_{33S} = 20\mu s^2$, which introduces decoherence into the system on a microsecond time scale. While it is not possible to include an incoherent process such as the spontaneous decay in a coherent description of the resonant energy transfer processes, we have to account for the finite lifetimes to be able to compare the results of the simulations to experimental data. We therefore included the finite lifetimes in a phenomenological way by multiplying the results of the model with an exponential

\footnote{The lifetimes include the spontaneous decay and decay induced by black body radiation [Gal94].}
2.6. Spontaneous decay of the atoms

decay factor $e^{-t/\tau_{\text{eff}}}$. As the atoms are in superpositions of $^{32}\text{P}$, $^{32}\text{S}$ and $^{33}\text{S}$, the effective lifetime must involve contributions from both $\tau_{32\text{P}}$ and $\tau_{32\text{S}}$ resp. $\tau_{33\text{S}}$. For simplicity reasons, we chose $\tau_{\text{eff}}^{-1} = \frac{1}{2}(\tau_{32\text{P}}^{-1} + \tau_{32\text{S}}^{-1})$, i.e. the effective decay rate $\tau_{\text{eff}}^{-1}$ is the average of the decay rates of $^{32}\text{P}$ and $^{32}\text{S}/^{33}\text{S}$, which yields $\tau_{\text{eff}} = 24\mu\text{s}$. This corresponds to the case that the atoms have equal probability to be $^{32}\text{P}$ or $^{32}\text{S}/^{33}\text{S}$, which is true on average once the populations have reached a steady state. We hereby may make errors for the smaller densities where it takes times comparable to the lifetimes to reach the steady state.

The treatment of the spontaneous decay does not consider possible modifications of the dynamics of the resonant energy transfer processes through the spontaneous decay, which effectively leads to a reduction in the density of the Rydberg atoms available for resonant energy transfer processes. The errors made hereby are expected to be negligible if the population transfer is fast compared to the lifetimes, i.e. the populations already approach a steady state in periods much smaller than the lifetimes. However, we cannot exclude effects of the finite lifetimes on the resonant energy transfer processes for the smaller densities, where this is not necessarily the case.
Chapter 3

Experimental observation of resonant energy transfer

In this chapter, I will present measurements on the time development of the resonant energy transfer

\[
32P_{3/2,|m_j|=3/2} + 32P_{3/2,|m_j|=3/2} \leftrightarrow 32S_{1/2} + 33S_{1/2}. \tag{3.0.1}
\]

First, I will give a brief description of the Rydberg experiment, followed by simulation on the detection efficiency of the Rydberg atoms. I will then present the experiment on the resonant energy transfer and compare the model of chapter 2 to the results. Various error sources will be discussed.

3.1 Preparation of a cold Rydberg gas

A detailed description of the experimental setup used to create a cold Rydberg gas is given in the doctoral thesis of Kilian Singer [Sin04a]. After a short overview of the general setup I will therefore concentrate on the parts of the experiment that are crucial for the investigation of resonant energy transfer processes. Furthermore, I will present simulations on the detection scheme for Rydberg atoms that were performed during my thesis.
Figure 3.1: Photo of the MOT-chamber taken during the setup of the experiment. (1) field grids with holders used for field ionization of the Rydberg atoms. (2) Coils generating the magnetic quadrupole field for the MOT. (3) Compensation coils producing magnetic offset fields. (4) Micro channel plate detector (MCP). (5) Schematic drawing of the trapped atom cloud. Figure adapted from [Sin04a].

3.1.1 Experimental setup

A magneto-optical trap (MOT) is used to trap and cool a cloud of Rubidium atoms, that are delivered from dispensers located inside the vacuum chamber. The magnetic field gradient needed for the trapping scheme is produced by a pair of coils in Anti-Helmholtz configuration (Fig. 3.1). Trapping laser beams are shone in from all six directions and exert a radiation pressure on the atoms directed towards the center of the trap (Fig. 3.2 b). The trapping light is generated by a commercial diode laser (Toptica, TA 100) and frequency-stabilized via atomic spectroscopy. The MOT runs on the transition between the hyperfine level $F = 2$ of the $^{87}$Rb ground state $|5S_{1/2}\rangle$ and the $F = 3$ level of the first excited state $|5P_{3/2}\rangle$ (Fig. 3.2 a). A grating stabilized diode laser (designed and built in our group [Sin04a]) is used to repump atoms from the $F = 1$ level of the ground state which is "dark" for the MOT light to the "bright" $F = 2$ state. For this purpose, the atoms are excited
from $|5S_{1/2}, F = 1\rangle$ to $|5P_{3/2}, F = 2\rangle$ from where they spontaneously decay within nanoseconds back both to the desired $F = 2$ level of the ground state and the $F = 1$ level, thus remaining in the repumping cycle until they finally decay into $F = 2$. In our MOT we typically trap between $10^7$ and $10^8$ atoms at peak densities around $10^{10}$ cm$^{-3}$ and temperatures around 100 µK, with a loading time of roughly five seconds. The diameter of the trapped atom cloud, which is determined by the magnetic field gradient and by the detuning of the trapping light from the atomic resonance, is on the order of 1 mm.

The cold Rubidium atoms are excited to the Rydberg states via the two-photon excitation scheme depicted in Fig. 3.2 a. The first photon with a wavelength of approx. 780 nm that excites the atoms to the intermediate state $|5P_{3/2}, F = 2\rangle$ is provided by a commercial high-power diode laser with a line width <1 MHz (Toptica, DLX 110). The second photon with approx. 480 nm is generated by a cw laser system consisting of a frequency-doubled semiconductor laser (Toptica, TA-SHG 110) and excites the atoms to the Rydberg states. The frequency of this laser can be

Figure 3.2: a) Trapping and excitation scheme for $^{87}$Rb. See text. b) Schematic setup for the preparation of an ultra-cold Rydberg gas from a magneto-optical trap. See text. Figure adapted from [Sin04a].
tuned by more than 5000 GHz, covering the range from Rydberg levels with principal quantum number of 30 to above the ionization threshold. The frequency of this laser is locked to the modes of an ultra-stable reference cavity, thus providing a light source with a line width $<2$ MHz which is sufficiently narrow to spectroscopically resolve single Rydberg levels. All laser beams can be independently switched by accousto-optical modulators (AOM) with switching times around 100 ns. For the experiments performed on resonant energy transfer processes, the two excitation lasers were aligned in a counterpropagating configuration. The red laser had a waist of 1100 $\mu$m, thus illuminating the whole atom cloud in the MOT. The blue laser was focussed to a waist of $(37 \pm 2) \mu$m with a Raleigh range of about 1 mm, which defines the excitation volume in which a couple of ten thousand ground state atoms are present.

The atom cloud is imaged on a photodiode, so that the fluorescence of the cloud (i.e. photons from the trapping lasers scattered by the atoms during the trapping process) can be monitored. As only atoms in the launch state $|5P_{3/2}, F = 2\rangle$ scatter photons (the few transitions from $F = 1$ due to the repumping laser can be neglected), the fluorescence is a measure for the number of atoms in the MOT and, as the size of the cloud is fixed, for the atomic density. As the atoms on average scatter photons with the same rate, the fluorescence signal is proportional to the launch state density.

Metal grids with high optical transmission (denoted field plates in Fig. 3.2) are placed on either side of the atom cloud and can be used to apply defined electric fields to the atom cloud. They are also used to ionize the Rydberg atoms, which is part of the state-selective detection scheme. Depending on the state Rydberg atoms ionize at electric fields between a few and a few hundred V/cm. The created Rb$^+$-ions are then accelerated towards a Micro-Channel-Plate detector (MCP, Fig. 3.2) where they are detected. As the ionization energy is different for each Rydberg state, one can discriminate between different Rydberg states contained in the cloud by applying a sufficiently slow electric field ramp. Rydberg states ionize the later the more deeply they are bound, so that the ions originating from strongly bound states will arrive later at the MCP than ions from weakly bound states. Through the field ionization spectra, it is also possible to detect Rb$^+$-ions that have formed through collision processes in the Rydberg cloud. Such ions are already present at the start of the field ramp and will thus instantly be accelerated towards the MCP.
Therefore, they are recorded shortly after the start of the field ramp as a peak in the field ionization spectrum. This peak is clearly separated from all peaks that originate from field ionization of Rydberg atoms.

The dynamics of the ion clouds which can affect the detection efficiency of the MCP is investigated in detail in section 3.2.

### 3.1.2 Excitation of P states and selected $m_j$ states

As the atoms undergo the final excitation step to the Rydberg states from the intermediate state $|5P_{3/2}\rangle$, a transition to $P$-states is forbidden due to selection rules for dipole transitions. For Rydberg atoms this problem can be overcome by exciting in a static electric field. A moderate field of a few V/cm, which can be applied via the metal grids in the vacuum chamber (Fig. 3.2) is sufficient to induce a permanent electric dipole moment, which the electric field of the laser light can couple to. Quantum mechanically, the static electric field leads to a small admixture of $S$- and $D$-character to the $P$-state due to the Stark effect, so that the dipole matrix element with the $|5P\rangle$-state does not completely vanish. This state is then adiabatically transferred into a pure $P$-state when the electric field is switched back to zero. The excitation rates reached with this method are considerably lower compared to excitation of $S$- and $D$-states.

A static electric field also allows to spectroscopically select certain $m_j$-sublevels, which are degenerate and thus not selectively addressable at zero field. The Stark effect leads to a splitting of levels with different $|m_j|$, which is larger than the line width of the excitation laser already at moderate fields. However, it is not possible to separate levels with $+m_j$ and $-m_j$ with this method.

### 3.1.3 Control of Rydberg density

If a constant excitation rate from the launch state $|5S_{1/2}, F = 2\rangle$ to the Rydberg states is assumed (i.e. a rate depending only on the laser power, but not on the density), the Rydberg density can be controlled by varying the density of the launch state. This can be done through optical pumping into the "dark" state $|5S_{1/2}, F = 1\rangle$ [Sin04b]. For this, the repumping laser is switched off for a variable time before turning on the excitation lasers, so that atoms that are accidentally redistributed to the "dark" state are no longer transferred back to the MOT cycle.
Such atoms are not resonant with the excitation lasers, so they are not excited to Rydberg states. Furthermore, they experience no trapping force and thus leave the trapped cloud after some time. Either process is sufficient to reduce the launch state density. The amount of density reduction is determined by the rate of the redistribution to the dark state and the time the repumping laser is switched off. This must be on the order of milliseconds in order to achieve a considerable reduction of the density. The redistribution rate is not exactly known, so the reduction of the launch state density is determined by fluorescence measurements. As the fluorescence signal is directly proportional to the launch state density one can determine the ratio between the reduced density and the maximum density that is achieved when the repumping laser is left on. Note that the density distribution in the MOT is not homogeneous, but has the shape of a Gaussian, so that the fluorescence signal must be understood as an average over this density distribution.

The determination of absolute densities of Rydberg atoms is unfortunately a non-trivial task. Although the absolute density of atoms in the launch state can be measured with a good accuracy via absorption pictures [Sin04a], the excitation rate to the Rydberg levels is not exactly known as a number of factors contribute to this excitation rate:

- For the limit of low laser power, the excitation rate for a single atom from the intermediate state $|5P_{3/2}\rangle$ to the Rydberg levels is proportional to the intensity of the blue excitation laser at the site of the atom [Met02]. However, the intensity distribution of the beam is a Gaussian, so that there is a distribution of excitation rates across the beam profile, which then overlaps with the distribution of ground state atoms in the MOT, so that one can at most determine an average excitation rate and thus an average density. This is unsatisfactory for the investigation of density-dependent Rydberg-Rydberg interactions.

Note that the same argument in principle also holds for the first excitation step, but as the red laser is broader than the atom cloud, only the central, relatively flat pat of the gaussian intensity distribution can excite atoms, so that the variation of the excitation rate is negligible in this case. The blue laser has to be focussed this strongly because the transition strength from the intermediate state to the Rydberg states is very weak, thus requiring strong laser intensities to achieve a sufficient excitation rate.
• The excitation process can be simulated using the optical Bloch equations for a three-level system [Dei05b]. They account for the line width of the lasers and effects such as the spontaneous decay of the intermediate state $|5P_{3/2}\rangle$ or power saturation of the transition, which strongly influence the excitation process. However, the results of the optical Bloch equations critically depend on factors such as the Rabi frequencies of the lower and upper transition, which are often not known to an accuracy that would allow a reliable determination of the excitation rate.

• While the two above mentioned factors make it hard to determine the excitation rate, they conserve the density ratio of the launch state densities when the atoms are transferred to a Rydberg state, since neither of them has a dependency on the launch state density itself. However, we cannot rule out the possibility of an interaction-induced excitation blockade due to interactions between Rydberg atoms [Luk01]. Such interactions always depend on the interatomic separations in the gas and thus on the launch state density. In the blockade regime, less atoms are excited to Rydberg states than to be expected from the excitation rate. Although the density ratio of the launch states is known, the ratio of the Rydberg densities is different as the blockade effect is more pronounced at higher densities. At the extreme case of a very strong blockade, the Rydberg densities are approximately equal for all launch state densities. The real density ratio of the Rydberg densities lies somewhere between this extreme case and the ratio of the launch state densities, which is the case if no blockade occurs. While certain aspects of the excitation blockade have been investigated in detail [Sin04b, Ton04, CL05], a complete picture is still missing.

3.2 Detection efficiency for Rydberg atoms

The above mentioned problems could be solved with a detection scheme for the Rydberg atoms that can be gauged to deliver absolute atom numbers. In the following, I will attempt a characterization of error sources that make this task difficult.

After the ionization of the Rydberg atoms the created Rb$^+$-ions are accelerated towards the MCP detector. During the time of flight each ion experiences repulsive
Coulomb forces by the charge of neighboring ions which is known to evoke a Coulomb explosion of the ion cloud. This can strongly influence the impact area of the ion cloud on the MCP and thus the number of ions impacting per area which is the crucial property for saturation effects of the MCP. A saturation of the MCP in turn affects the efficiency that the Rydberg atoms are detected with. A simulation of the dynamics of the ion cloud was thus performed to obtain estimations of the detection efficiency.

The Rydberg atoms are excited in a cylindrical volume with a radius of $37 \mu m$ (waist of the blue excitation laser) and a height of $1 \text{ mm}$ (diameter of the MOT). In our case, this cylinder lies parallel to the metal grids. For detection, a positive voltage $U_{\text{ramp}}$ that is ramped up from zero is applied to the left metal grid (see Fig. 3.3), while the central grid remains grounded. At a defined voltage $U_{\text{ion}}(nl)$ of the ramp which is characteristic for each state, the Rydberg atoms are field-ionized into an electron and a Rb$^+$-ion. The electrons are attracted by the positively charged left metal grid and leave the excitation volume within roughly a nanosecond, thus having no effect on the much heavier ions whose dynamics is rather on the time scale of few microseconds. If the field ramp is sufficiently slow, the ions to a good approximation only experience the ionizing voltage $U_{\text{ion}}(nl)$ by which they are accelerated towards the central grid. They penetrate the grid and enter region II (Fig. 3.3) which can be set to a defined electric field by a third metal grid at a potential $U_{\text{II}}$ placed directly in front of the MCP. The ions hit the detector which is at a high negative voltage almost instantly after penetrating this final grid.

We performed a simulation in C of the dynamics of the ion cloud by numerically solving Newton’s equation for the trajectory of each ion [Wes05a]. The simulations were performed for the parameters chosen in the experiment. The electric field in region I was assumed to be on the order of $300 \text{ V/cm}$ which roughly corresponds to the ionizing field for Rydberg states around $n = 32$ [Gal94]. We chose region II to be field-free so that the ion cloud experienced no external electric force here. The electric fields are homogeneous to a good approximation as the metal grids producing the fields are much larger than the ion cloud. The simulations were performed over the range of 40 to 10000 ions contained in the cylindrical excitation volume (sec. 3.1.1). This roughly corresponds to Rydberg densities between $10^7 \text{ cm}^{-3}$ and $2 \times 10^9 \text{ cm}^{-3}$.

Each ion is assigned a random starting position according to a three-dimensional
3.2. Detection efficiency for Rydberg atoms

Figure 3.3: Schematic drawing of the parts of the setup relevant for ion detection. Depicted are the cloud of Rydberg atoms (red), three metal grids which can be set to defined potentials and the Micro-Channel-Plate detector (MCP). The atom cloud is not drawn to scale. See text.

gaussian distribution with a width of 0.5 mm (the radius of the atom cloud in the MOT) parallel to the metal grids and 37 µm (the waist of the blue excitation laser) in the two remaining directions, which is accomplished by the function gsl_ran_gaussian from the Gnu Scientific Library [Gal]. Although the real distribution of the Rydberg atoms can deviate from a Gaussian in case of considerable power saturation (sec. 3.1.3), the assumed initial distribution is still a good approximation due to the dimensions of the excitation laser and the MOT (sec. 3.1.1).

In each integration step the total force acting on each atom has to be evaluated, which is the sum of the external electric force and the Coulomb forces due to all other atoms. The external electric force is \( \vec{F}_{el} = e \vec{E}_I \) if the ion is in region I and zero if it is in region II. Here, \( e \) denotes the elementary charge and \( \vec{E}_I \) the electric field in region I, in our case 300 V/cm pointing towards the MCP. The Coulomb force on the k-th ion can be written as

\[
\vec{F}_{Coul,k} = \sum_{i \neq k} \frac{e^2}{4\pi \varepsilon_0} \frac{\vec{r}_k - \vec{r}_i}{|\vec{r}_k - \vec{r}_i|^3},
\]

where \( e \) denotes the elementary charge, \( \varepsilon_0 \) the vacuum permittivity and \( \vec{r}_i \) the position vector of the i-th ion. The sum has to be performed over all ions. The determination of the Coulomb forces is the most time-consuming part of the simulation, as the calculational effort grows quadratically with the number of ions. For clouds of more than 10000 ions the program therefore becomes unpractical. The numerical
integration of Newton’s equation is performed using a fourth-order Runge-Kutta integration scheme [Lam73]. For simplicity reasons, we set \( \mathbf{r} = (\mathbf{r}_1, ..., \mathbf{r}_N) \) and the respective expressions \( \mathbf{v} = (\mathbf{v}_1, ..., \mathbf{v}_N) \) and \( \mathbf{F} = (\mathbf{F}_1, ..., \mathbf{F}_N) \) for the velocities and forces. The positions \( \mathbf{r}(t) \) and velocities \( \mathbf{v}(t) \) of the ions are propagated to \( \mathbf{r}(t + \Delta t) \) and \( \mathbf{v}(t + \Delta t) \) by the following scheme:

\[
\begin{align*}
k_1 &= \Delta t \frac{1}{m_{Rb}} \mathbf{F}(x_n); & l_1 &= \Delta t \mathbf{v}_n;
k_2 &= \Delta t \frac{1}{m_{Rb}} \mathbf{F}(x_n + \frac{l_1}{2}); & l_2 &= \Delta t (\mathbf{v}_n + \frac{k_1}{2})
k_3 &= \Delta t \frac{1}{m_{Rb}} \mathbf{F}(x_n + \frac{l_2}{2}); & l_3 &= \Delta t (\mathbf{v}_n + \frac{k_2}{2})
k_4 &= \Delta t \frac{1}{m_{Rb}} \mathbf{F}(x_n + l_3); & l_4 &= \Delta t (\mathbf{v}_n + \frac{k_3}{2})
\end{align*}
\]

\[
x_{n+1} = x_n + \frac{1}{6} (k_1 + 2k_2 + 2k_3 + k_4);
x_{n+1} = v_n + \frac{1}{6} (l_1 + 2l_2 + 2l_3 + l_4)
\]

We cannot explicitly specify the errors made by the numerical integration, but we checked that no change in the final shape and dimensions of the ion cloud was visible when increasing the number of integration steps. As we are finally interested in “global” properties such as the the impact density and the extension of the ion cloud, stronger errors made for single ions which cannot be ruled out are tolerable.

For simplicity reasons, we did not set a termination condition, if an ion reached the position of the MCP, but terminated the simulation for all ions when the center of the cloud reaches the detector. The coordinates of the ions in the plane parallel to the MCP were then taken as impact coordinates, which is justified due to the insignificant extension of the ion cloud perpendicular to the MCP. Fig. 3.4 shows the results of the simulation for 100, 1000 and 10000 ions compared to the initial distribution. One can clearly see the expansion of the cloud due to the Coulomb repulsion between the ions which is the larger the more ions are “packed” in the excitation volume.

Each channel of the MCP has a certain dead time, i.e. a time after the impact of an ion that the respective channel is insensitive to further ions. This dead time is considerably longer (around 1 ms) than the the impact time of the ion cloud, so that only one ion per channel can be detected when an ion cloud reaches the MCP. For
3.2. Detection efficiency for Rydberg atoms

Figure 3.4: Impact coordinates on the MCP for clouds of 100, 1000 and 10000 ions obtained from the simulation. Each of the blue rhombuses corresponds to the impact of one ion. For comparison, the approximate size and shape of the initial distribution of Rydberg atoms which the ions are created from is shown. Note that in all cases the ion clouds are far smaller than the MCP which has a diameter of 18 mm, so that all ions finally impact on the detector.

small impact densities, this has negligible influence due to the statistical distribution of the atoms, but once the impact densities is in the range of the channel density of the MCP a considerable number of ions is left undetected. The MCPs (Topag, MCP-MA-25/2) used in the experiment feature micro-channels with diameters of 10 µm in a hexagonal configuration, with the distance between neighboring channels being 12 µm. In order to obtain estimations for the number of not detected ions from the simulation we partition the impact plane in areas of $12 \times 12 \, \mu\text{m}^2$ and count how many of these areas are hit by ions. The ratio of this number to the total number of ions in the cloud is a measure for the detection efficiency. The real losses are
Table 3.1: Estimates of the detection efficiency of the MCP obtained from the simulation. The numbers must be understood as lower limits (see text). The ion number is the number used for the simulation, which approximately corresponds to the given Rydberg densities if the density is assumed to be constant over the excitation volume.

<table>
<thead>
<tr>
<th>Rydberg density</th>
<th>ion number</th>
<th>detection efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1 \times 10^7$ cm$^{-3}$</td>
<td>40</td>
<td>100%</td>
</tr>
<tr>
<td>$2.5 \times 10^7$ cm$^{-3}$</td>
<td>100</td>
<td>95%</td>
</tr>
<tr>
<td>$1 \times 10^8$ cm$^{-3}$</td>
<td>400</td>
<td>92%</td>
</tr>
<tr>
<td>$2.5 \times 10^8$ cm$^{-3}$</td>
<td>1000</td>
<td>88%</td>
</tr>
<tr>
<td>$1 \times 10^9$ cm$^{-3}$</td>
<td>4000</td>
<td>75%</td>
</tr>
<tr>
<td>$2.5 \times 10^9$ cm$^{-3}$</td>
<td>10000</td>
<td>66%</td>
</tr>
</tbody>
</table>

overestimated, as the hexagonal pattern of the channels is not taken into account, but simulated values must be understood as crude estimates anyway.

The obtained detection efficiencies are presented in table 3.1. It is clear that the saturation effects must be taken into account for Rydberg densities exceeding approx. $10^8$ cm$^{-3}$. On the other hand, the detector saturation does not follow a pure saturation function, which can be attributed to the Coulomb expansion of the ion cloud. As can be seen in Fig. 3.4 an increase in the number of ions in the excitation volume also leads to an increase in the size of the ion cloud when reaching the detector, which reduces the impact density and thus saturation effects. As the ion cloud is still considerably smaller than the sensitive area of the detector, one could exploit this effect by giving the ion cloud more time to expand. This can be done by applying a positive voltage $U_{II}$ to the metal grid in front of the MCP (Fig. 3.3). As long as $U_{II}$ is smaller than $\frac{1}{2} U_{ion}(nl)$ the ion cloud is decelerated once it enters region II, but still reaches the detector.

So far, we restricted ourselves to the case where only one Rydberg state is detected, i.e. only one ion cloud impacts on the MCP. However, if several Rydberg states which ionize shortly after each other are present in the excitation volume, it is possible that saturation of the detector evoked by one of the produced ion clouds has influence on the detection efficiency for consecutive clouds. This issue will be
3.3 Investigating resonant energy transfer

In the following, we will describe experiments investigating the dynamics and density dependence of the resonant energy transfer process

\[ 32P_{\frac{3}{2}}|m_j|=\frac{3}{2} + 32P_{\frac{3}{2}}|m_j|=\frac{3}{2} \rightarrow 32S_{\frac{1}{2}} + 33S_{\frac{1}{2}}. \]  

(3.3.1)

The resonance condition for this process is fulfilled for a static electric field of \( E_{\text{res}} = 11.5 \) V/cm (see sec. 1.2.2), so that we can control the process via the electric field. As small residual electric fields are present in our setup, we do the fine adjustments of the electric field by optimizing on a maximum population of 32S atoms.

One experimental cycle which has a repetition rate of approx. 15 Hz consists of the following timing sequence (see Fig. 3.5):

- At the beginning, the MOT is in a steady-state. The repumping laser is

![Figure 3.5: Timing scheme for the excitation lasers and the electric field. A short excitation pulse excites a gas of Rydberg atoms. The electric field is switched to the resonance condition for a variable time \( \Delta t \), after which it is ramped up for state-selective field ionization. See text for details.](image-url)
switched off for a variable time between zero and 9 ms, leading to a decrease in the launch state density, which is used to control the Rydberg density (see sec. 3.1.3).

- At \( t=0 \) (Fig. 3.5) both the trapping lasers and the magnetic field of the MOT are switched off. Although the cloud is now free to expand, the atoms can be considered essentially static for the period of many microseconds, as the gas is cooled to a temperature around 100 \( \mu \text{K} \).

- Both excitation lasers are switched on for 100 ns (\( P_{780\text{nm}} = 750 \mu\text{W}, P_{480\text{nm}} = 30 \text{ mW} \)). As the excitation of \( ^3\text{P} \) is forbidden at zero field due to dipole selection rules, a static electric field of \( E_{\text{exc}}=15 \text{ V/cm} \) is applied during excitation. This also allows to spectroscopically select the \( ^3P_{\frac{3}{2}} \) states with \( |m_j| = \frac{3}{2} \). The excitation time is chosen as short as the finite switching times of the acousto-optical modulators allow in order to prevent the production of ions during excitation. After excitation, a gas of Rydberg atoms in \( ^3P_{\frac{3}{2}}, |m_j| = \frac{3}{2} \) has been created.

- 100 ns later, the electric field is switched to \( E_{\text{res}}=11.48 \text{ V/cm} \) for a variable interaction time \( \Delta t \), which can be chosen in the range between 80 ns and 10 \( \mu\text{s} \). During this time, resonant energy transfer processes occur and populations of \( 32\text{S} \) and \( 33\text{S} \) atoms are accumulated.

- The electric field is switched back to \( E_{\text{exc}} \) for 500 \( \mu\text{s} \), which instantly stops resonant energy transfer processes. Excitation exchange processes (sec. 1.2.3) remain on resonance and thus do occur, but do not change the the accumulated populations of \( 32\text{S} \) and \( 33\text{S} \).

- The electric field is subsequently ramped up for state-selective field ionization of the Rydberg atoms. The Rydberg states in question ionize at an electric field of approx. 300 V/cm. The created \( \text{Rb}^+ \)-ions are detected with the MCP. Note that the \( E_{\text{exc}} \) is deliberately chosen above \( E_{\text{res}} \) as the field ramp otherwise has to be be swept over the resonance condition.

- The trapping lasers and the magnetic field are switched on again, so that the MOT can reload and reach its steady-state before the next cycle starts. During this time a high voltage is maintained at the metal grids to remove
any ions from the cloud that are unintentionally produced. This is necessary because the reloading time is considerably longer than the time of the actual experiment, so that a population if ions, though produced at a small rate, can accumulate if it is not steadily removed.

The experiment was performed for three different densities: To achieve a high density, the repumping laser was left on all time. For a medium resp. low density the repumping laser was turned off for 5 ms resp. 9 ms before excitation to the Rydberg states. From measurements of the fluorescence, the ratios between the three launch state densities could determined to 1 : 0.2 : 0.06 (see sec 3.1.3).

The field ionization spectra obtained from the MCP are amplified by a Timing Filter Amplifier and then electronically averaged over 100-200 experimental cycles in order to increase the signal-to-noise ratio. Field ionization spectra on and off resonance are shown in Fig. 3.6. To determine populations of the Rydberg states, the signal has to be integrated. It is clear from the spectra that it is difficult to determine an accurate population for 33S as its signal appears on a underground of 32P. Therefore, it is not possible to select an integration window, in which only signal from the 32P-state is contained. For the same reason, the 32P population, which does not only appear in the central large peak, but also in adjacent areas, can only be determined with a large error. However, the 32S peak is clearly separated, thus providing accurate and reliable information on the resonant energy transfer. For evaluation purposes and to compare with the model of resonant energy transfer (chapter 2), we will thus focus on the 32S signal, and use the information on 32P only as a cross-check. The 33S signal is not used for evaluation.

3.4 Experimental results and comparison to the model

The results of the measurements of the resonant energy transfer are shown in Fig. 3.7. For zero interaction time, only the initially excited 32P atoms are detected\(^1\).

\(^1\)For the high density, some 32S atoms are already detected for the smallest possible interaction time of 80 ns. As the sum of two times the 32S population and the 32P population remains approximately constant, we can infer the data point for $\Delta t = 0$ to be around 9.
Figure 3.6: Field ionization spectra obtained as raw data from the MCP (amplified by a Timing Filter Amplifier). The orange trace is taken off resonance, while the black trace shows the spectrum on resonance for an interaction time of $\Delta t = 5 \mu s$. The central $32P$-peak is cut off in both cases. One can clearly see, how population of $32S$ and $33S$ appears in the resonant case. The position of the peaks for the respective states can be determined by exciting exclusively this state. The population of the states can be determined by setting integration windows over the respective areas. The $32P$ population was determined as the area below the central large peak. A part of it is not recorded, as the peaks to the left and right of the central peak also contain signal of $32P$.

During the first few microseconds the number of $32P$ atoms drops, while a population of $33S$ atoms builds up. The redistribution process is faster at higher Rydberg densities, as to be expected from the density-dependence of the dipole-dipole interactions. The sum of two times the $32S$ population and the $32P$ population is approximately constant. For interaction times of more than $5 \mu s$, the $32S$ signal begins to decrease again due to the spontaneous decay of the atoms. The lifetime of $32S$ and $32P$ including decay induced by black-body radiation is $\tau_{32S} = 20 \mu s$ and $\tau_{32P} = 30 \mu s$.

The excitation time is chosen as short as possible to prevent a production of Rb$^+$-ions during excitation. However, at the high density, a small number of ions,
which is probably produced in interaction-induced collisions (see sec. 2.5), is already detected at the smallest measured interaction time of 80 ns, which increases with longer interaction times. From the MCP signals, we estimate that less than 10% of the Rydberg atoms are ionized for the high density at interaction times of 10 $\mu$s. For interaction times smaller than 1 $\mu$s, during which the 32S population builds up for this density, we estimate that less than 5% of the Rydberg atoms are ionized. For the smaller densities ionization, the ion signal is too small to yield reliable results.

### 3.4.1 MCP signal and Rydberg density

The ratios between the absolute MCP signals of the 32P population for zero interaction time should reflect the ratios of the Rydberg densities. From Fig. 3.7, one can determine the ratios between high (signal taken as 9, which is the sum of 32P and two times 32S at $t = 80$ ns), medium and low density to be approx. $1 : 0.56 : 0.37$, which is in contrast to the ratios of the respective launch state densities which was determined to $1 : 0.2 : 0.06$. A reliable determination the Rydberg densities is a crucial point when comparing the experimental data to the model described in chapter 2. We will therefore discuss possible explanations for the deviations before comparing to the model:

1. As discussed in section 3.1.3 we have to consider the possibility of an interaction-induced excitation blockade for higher launch state densities, which could explain the deviations. In this case the ratio of the Rydberg densities would not correspond to the ratio of the launch state densities. If one relies both on the determination of the launch state density ratios from the fluorescence measurements and the determination of the Rydberg density ratios from the MCP signals, one can infer a massive suppression of excitation by a factor of six for the high compared to the low density. An excitation blockade in a gas of Rydberg atoms in a single state can be induced by van der Waals-interactions or dipole-dipole interactions between permanent dipoles. In our case, we can rule out van der Waals-interactions, since no excitation blockade was observed for $n = 62$ [Sin04b] and the van der Waals-coefficient scales as $n^{11}$ with the principal quantum number $n$. Interactions between permanent dipoles indeed occur in our case, as the excitation to 32P is performed in a static electric field (sec. 3.1.2), which polarizes the atom and thus induces a permanent dipole
Figure 3.7: Measured time development of the populations of $^{32}$S and $^{32}$P atoms for different Rydberg densities. The amplification is equal for all densities, so that the absolute numbers of the MCP signal are comparable. Exact error bars cannot be given, as not all sources of systematic errors can be quantified. However, the statistical uncertainties are reflected in the spread of the points. The population of $^{32}$P cannot be determined as accurately as the $^{32}$S population, since the $^{32}$P peak overlaps with other states in the field ionization spectrum (see Fig. 3.6 and text).
moment. Calculations in second-order perturbation theory yield a substantial dipole moment of approx. 120 au for an atom in $|32P_{3/2}\rangle$ at an electric field of 15 V/cm. If the interaction energy between a pair of atoms is so much larger than the line width of the excitation laser, that even off-resonant excitation can be ruled out, only one of the atoms can be excited. According to (1.1.1) the upper limit for the interaction energy of two permanent dipole moments $\mu$ separated by $R$ is $\Delta E = 4\mu^2/R^3$. For a line width of 2 MHz, atom pairs separated by less than 3 $\mu$m thus are blockaded. In order to achieve a density reduction by a factor of six, one atom on average would have to blockade five surrounding atoms. It is important that the density of the intermediate state $5P_{3/2}$ is relevant for blockade effects, as the excitation to the interacting Rydberg levels starts from this state. If we assume a maximum launch state density of $10^{10}$ cm$^{-3}$, which is known from absorption pictures [Sin04a], the maximum possible density for the intermediate state is $5 \times 10^9$ cm$^{-3}$ if the excitation is assumed to be incoherent. This corresponds to the high density case, while the respective densities for the medium and low density are $10^9$ cm$^{-3}$ and $3 \times 10^8$ cm$^{-3}$. Table 3.2 shows estimates for the amount of blockade that is to be expected for the relevant range of densities if unity excitation probability without blockade is assumed. For realistic excitation rates the effects of the blockade would be smaller. Therefore and since the interaction-induced energy shift $\Delta E$ is overestimated, the numbers must be understood as upper limits. Nevertheless, they make clear that excitation blockade can only be relevant for the high density, where the real Rydberg density can be up to a factor of two smaller than to be expected from the ratio of the launch state densities. For the medium density and the low density the effects are below 10% and thus negligible. The strong deviation of the MCP signal ratios from the launch state density ratios in any case cannot be explained by a strong interaction-induced excitation blockade.

2. A different explanation for the deviation of the MCP signals would be a strong saturation of the detector for higher densities. This means that the ratio of the Rydberg densities corresponds to the ratio of the launch state densities, but that for higher densities a large part of the ions is not detected. Once again, a discrepancy of a factor of six has to be explained. From the simulations on
Table 3.2: Probabilities for an atom excited to a Rydberg state to blockade the excitation of surrounding atoms. To obtain these numbers, we considered each atom within the radius of 3 \( \mu \text{m} \) of the excited atom to be blockaded. The numbers correspond to the case that each atom without blockade, has unity probability to be excited. For smaller excitation rates the blockade effects are less pronounced. See Appendix B.

<table>
<thead>
<tr>
<th>density of 5P_{3/2} atoms</th>
<th>probability for an excited atom to blockade</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>&gt; 1 atom</td>
</tr>
<tr>
<td>5 \times 10^9 \text{ cm}^{-3}</td>
<td>43%</td>
</tr>
<tr>
<td>1 \times 10^9 \text{ cm}^{-3}</td>
<td>11%</td>
</tr>
<tr>
<td>5 \times 10^8 \text{ cm}^{-3}</td>
<td>5%</td>
</tr>
</tbody>
</table>

the detection efficiency in section 3.2 we know that the detection efficiency for a single cloud of ions is still better than 50\% in the relevant range of Rydberg densities. The important point is that the simulations were performed for a single ion cloud. However, as one can see from the field ionization spectra in Fig. 3.6 several states corresponding to several ions clouds are detected shortly after each other. As the dead time of the channels of the MCP is much longer than the time window in which the different ion clouds arrive, the detection efficiency may drastically decrease for the later ion clouds as the earlier ion clouds already saturate a large fraction of the available channels. Furthermore, the argument that the Coulomb expansion of the ion cloud partly reduces the MCP saturation does not hold if the ions arrive in several clouds, as the clouds more or less impact on the same area of the detector. We performed the simulations on the detection efficiency described in sec. 3.2 for two and three ion clouds, which consecutively impact on the MCP. All channels that have already been hit by at least one ion of the preceding clouds are considered to be saturated and cannot detect any further ions. For clouds of 1000 ions each, the detection efficiency which is almost 90\% for the first cloud drops to 60\% for the second and 45\% for the third cloud. While this simulation is not meant to exactly describe the situations that lead to the spectra of Fig.
3.4. Experimental results and comparison to the model

3.4.2 Comparison to model

In order to compare the model of chapter 2 to the experimental data the modelled curves (with the finite lifetime of the atoms factored in, see sec. 2.6) must be scaled differently in height for each density. This scaling factor reflects both the different number of Rydberg atoms due to the different densities and the saturation of the MCP. We performed a least-square analysis to determine the best scaling factors for a range of densities for both the six- and the two-atom model. With \( D_{\Delta t_i} \) denoting the experimentally determined 32S-population for the interaction time \( \Delta t_i \), s the scaling factor and \( M_n(\Delta t_i) \) the 32S-population modelled for the Rydberg density \( n \) at an interaction time \( \Delta t_i \), the least square error \( \Pi_n \) is defined as

\[
\Pi_n = \sum_i \left( D_{\Delta t_i} - s \cdot M_n(\Delta t_i) \right)^2 ,
\]  

(3.4.1)

where the sum is performed over all experimental data points. The best scaling factor can be determined by minimizing \( \Pi_n \), and this minimal least square error
then quantifies the amount of agreement for the respective density $n$. It also helps to reveal differences between the many-body and the pair model in the capacity to describe the experimental data. The experimental data points are not equally spaced with respect to the interaction time, which leads to fitted model curves that visually do not agree well with the experimental data. Interaction times below $1 \mu s$ are overrepresented in such fits as the measured data points are concentrated in this region (Fig. 3.7), so that the fitted curves agree well in this region while they do not adequately describe the rest of the interaction time. To overcome this problem, we added points by linearly interpolating between measured data points to obtain a constant point density over the entire range of interaction times.

Fig. 3.8 shows the results for the two- and the six-atom model for a range of densities. The six-atom model fits best for $n = 1.75 \times 10^8 \text{ cm}^{-3}$ for the high density and $n = 5 \times 10^7 \text{ cm}^{-3}$ for the medium density. For the low density, a range of densities between approx. $n = 1.5 \times 10^7 \text{ cm}^{-3}$ and $n = 3 \times 10^7 \text{ cm}^{-3}$ describes the experimental data best. The agreement of the six-atom model for those optimum densities is substantially better than for any of the modelled densities in the two-atom model. At least for the medium and the high density, the fit is strongly influenced by the data points at 5 and 10 $\mu s$, which are responsible for half of the points considered in the fits. The procedure of adding data points is justified for interaction times below $5 \mu s$ (see Fig. 3.7), so we repeated the least-square analysis this for interaction time of less than $5 \mu s$, which yields no substantial differences compared to Fig. 3.8: The least square errors are still consistently larger for the two-atom model than for the six-atom model. The six-atom model still agrees best for $n = 1.75 \times 10^8 \text{ cm}^{-3}$ for the high density and for $n = 5 \times 10^7 \text{ cm}^{-3}$ for the medium density. For the low density, the minimum of the least square errors is between $n = 1.5 \times 10^7 \text{ cm}^{-3}$ and $n = 5 \times 10^7 \text{ cm}^{-3}$, which is also in agreement with Fig. 3.8.

A comparison of the six-atom model to the experimental data is shown in Fig. 3.9. The densities of the modelled curves are selected as those three densities at the ratio $1 : 0.2 : 0.06$ that feature the smallest least square errors (see Fig. 3.8). The shown curves are simulations for $n = 2.5 \times 10^8 \text{ cm}^{-3}$ for the high density, $n = 5 \times 10^7 \text{ cm}^{-3}$ for the medium and $n = 1.5 \times 10^7 \text{ cm}^{-3}$ for the low density. The modelled and measured $32\text{P}$ populations are also shown, although the $32\text{P}$ population cannot be determined to the same accuracy as the $32\text{S}$ population (sec. 3.3), so that it is not clear how significant the deviations from the modelled curves are.
3.4. Experimental results and comparison to the model

Figure 3.8: Least square errors for the six- and the two-atom model for the three densities investigated in the experiment. Each of the squares and rhombuses corresponds to one density for which the 32S population was modelled. The best agreement with the experimental data at a fixed density ratio of 1 : 0.2 : 0.06 (which is the ratio of the Rydberg densities in the experiment) is for $n = 1.5 \times 10^7$ cm$^{-3}$ for the low density, $n = 5 \times 10^7$ cm$^{-3}$ for the medium density and $n = 2.5 \times 10^8$ cm$^{-3}$ for the high density, which all lie within close range of the minima of the least square errors.
Figure 3.9: Comparison of the six-atom model to the measured time development of the $^{32}$S and $^{32}$P populations. The solid lines are simulations for the specified densities. For presentation purposes, the experimental data are scaled in height to fit the modelled curves for the $^{32}$S population. The three densities feature a ratio of 1 : 0.2 : 0.06. Furthermore, they are all within close range of the minimum of the least square errors (see Fig. 3.8).
The influence of the Rb$^+$-ions formed through collisions on the resonant energy transfer process remains to be assessed. From the measurement, we estimated that less than 5% of the Rydberg atoms are ionized for the high density at interaction times of less than 1 $\mu$s, which is roughly the rise time of the 32S population. From independent measurements performed in our group, the width of the resonance can be determined to around 0.2 V/cm, so that we can estimate from table 2.3 that each ion on average inhibits the resonant energy transfer for one Rydberg atom, which reduces the effective Rydberg density by roughly 5%. This can be neglected compared to other error sources.

The main results can be summarized as follows:

- The six-atom model can very well reproduce the shape of the measured curves even under the constraint of the fixed density ratio. A model based on coherent many-body processes is thus in satisfactory agreement with the experiment.

- The maximum Rydberg density of $2.5 \times 10^8$ cm$^{-3}$ is a reasonable value considering the distribution of densities in the excitation volume with a peak launch state density of $10^{10}$ cm$^{-3}$. This corresponds to a peak excitation probability on the order of 5% which is reasonable considering that the transition to 32P is dipole-forbidden for zero field. It is very well in the range of values obtained when the excitation process is simulated solving the optical Bloch equations [Dei05b]. This simulation can only deliver a range of excitation probabilities, as relevant parameters such as the involved Rabi frequencies are not accurately known.

- The six-atom model is consistently superior to the two-atom model in reproducing the experiment. The important point herby is that a model based on many-body interactions is better suited than a model that only involves interactions between pairs of atom. While not being a rigorous proof it suggests that the dynamics of the resonant energy transfer process is governed by many-body effects.

- We have managed to quantitatively reproduce the experimental data with a treatment based on the coherent evolution of small subsystems of atoms, combined with a Monte-Carlo scheme to account for the variety of random
positions of the atoms. This gives us confidence that the developed method can be successfully applied to model similar systems.
Chapter 4

Conclusion and Outlook

In this chapter, I will first discuss possible improvements on the experiment on the resonant energy transfer. I then present experiments to further explore dipole-dipole interactions between Rydberg atoms. Simulations underline the experimental possibilities.

4.1 Improvements of the experiment on resonant energy transfer

For the investigation of resonant energy transfer processes it is crucial to prepare a system with a well-defined Rydberg density. Therefore, it is desirable to implement several improvements in the experiment described in the previous chapter.

- Due to the Gaussian distribution of launch state densities in the MOT, a whole range of Rydberg densities between zero and the peak density is found in the excitation volume, so that a comparison to the model of chapter 2 which is based on a constant density is only possible in terms of an average density. While we still find a good agreement of the model with the experimental data, it is desirable to achieve a constant launch state density over the whole excitation volume to sharpen the comparison. This can be done by restricting the excitation volume to a small region in the center of the MOT, where the launch state density is more or less constant. The easiest way is to cross the beams of the blue and the red excitation laser under an angle in the center of
the MOT. For this purpose, the red laser has to be attenuated and focussed to a waist much smaller than the diameter of the trapped atom cloud. Only atoms that can interact with photons from both the red and the blue beam can be excited to Rydberg states. The size of the excitation volume can be controlled by adjusting the waists of the excitation lasers at the crossing point. At small Rydberg densities, only few Rydberg atoms may be present in the small excitation volume, which makes their detection difficult. The range in which the Rydberg density can be varied is therefore smaller than in the case of counter-propagating beams. Another disadvantage of the crossed configuration is that the polarizations of the two excitation beams relative to each other are not well defined, which is important for some applications (see below).

- While a constant launch state density can be achieved with the above techniques, the problem of spatially different excitation probabilities due to the Gaussian beam profiles remains. The easiest way to achieve a constant intensity profile is to filter the Gaussian beam with a pinhole, which considerably reduces the power in the resulting beam. For this reason, it is not suitable at least for the blue beam, as all available laser power is required for adequate excitation rates to the Rydberg states. However, a method to transform gaussian beams into a constant intensity profile using holographic techniques has recently been implemented in the experiment [Dei05a]. Very little laser power is lost with this method. The size of the "flat-top" profile, i.e. the diameter over which the intensity is constant before rapidly dropping to zero, is on the order of 50 µm, which makes the adjustment critical, as the "flat-top" regions of both the red and the blue laser must be precisely overlapped. At a Rydberg density of $10^8$ cm$^{-3}$, only around ten Rydberg atoms are present in the excitation volume formed by such beams, which practically prevents the possibility to work with even smaller densities.

In order to investigate the density dependence of the resonant energy transfer it is desirable to achieve Rydberg densities higher than $2.5 \times 10^8$ cm$^{-3}$. The problem in the current experiment is the small excitation probability of only a few percent to the dipole-forbidden 32P state. The electric field applied to allow for excitation to the P-state at all furthermore induces a permanent electric dipole moment, so that an interaction-induced excitation blockade cannot be excluded for high excitation
4.1. Improvements of the experiment on resonant energy transfer

probabilities (sec. 3.4.1). Both problems would be overcome by a direct excitation of 32P atoms at zero field, which is not possible in the present two-photon excitation scheme. However, a three-photon excitation scheme could be implemented using microwaves. Here, the ground atoms are transferred to the Rydberg state $|31D_{5/2}\rangle$, which is dipole-allowed, with the present two-photon excitation scheme. A third photon provided by a microwave source then stimulates the atoms down to $|32P_{3/2}\rangle$, which is energetically approx. 75 GHz below the D-level. With incoherent excitation, an increase in the 32P-density of one order of magnitude compared to the present status can be achieved. Using coherent excitation, the excitation probability can be increased towards the launch state density. A coherent two-photon excitation to Rydberg levels has recently been demonstrated in our experiment [RL06]. Due to the long lifetime of the Rydberg levels, a coherent population transfer from a Rydberg D-level to a P-level can be achieved by applying a microwave $\pi$-pulse after the atoms have been excited to the D-state. Other advanced excitation schemes such as Rapid Adiabatic Passage, which has been realized for the two-photon excitation in our experiment [Dei05b], but has also been shown for multi-photon schemes [Mae06], can increase the Rydberg density to values close to the launch state density as well. Rapid Adiabatic Passage is also very robust against interaction-induced blockade of excitation, so that it is an ideal scheme to achieve high Rydberg densities. For a further increase of the Rydberg density it is necessary to increase the launch state density, so that other trapping schemes have to be implemented. With a dipole trap [Chu86] or a dark-spot MOT [Ket93] the launch state density can be further increased by one to two orders of magnitude. At such densities however, the energy shift induced by resonant dipole-dipole interactions for $n \approx 30$ is on the order of 1 GHz and population transfers, e.g. from 32P to 32S, are expected to occur within nanoseconds. The present equipment of the experiment is not suitable to resolve interactions on such timescales, e.g. the electric fields cannot be switched with the necessary accuracy. Improvements have to be made here before being able to explore interactions at very high densities.

In the current experiment, we were able to spectroscopically select certain $m_j$-sublevels due to the applied electric field (sec. 3.1.2), which is favorable for both modelling and controlling the interactions between the atoms, as the number of different interaction strengths present in the system increases with the number of $m_j$-sublevels. Through excitation in an electric field, we were able to restrict the
system to $\left|32P_{3/2}, m_j = +3/2\right>$ and $\left|32P_{3/2}, m_j = -3/2\right>$ in the current experiment. For a three-photon excitation scheme using microwaves, it is indeed possible to excite only one $m_j$-sublevel: The ground state atoms are first optically pumped into $\left|5S_{1/2}, F = 2, m_F = +2\right>$ through $\sigma^+$-polarized pump light from the red laser. $\sigma^+$-polarized light from both the red and the blue excitation lasers then excites the atoms exclusively into $\left|31D_{5/2}, m_j = +5/2\right>$. From this level, the only allowed transition when shining in resonant microwaves is to $\left|32P_{3/2}, m_j = +3/2\right>$. Except for the microwave setup, the scheme has already been implemented in our experiment [Dei05b]. A crucial point is that both the red and the blue excitation beam must be $\sigma^+$-polarized with respect to the same axis, which is not possible in the crossed configuration necessary to select a small excitation volume with a constant launch state density. However, it is possible to shine in both excitation lasers from the same direction and select the excitation volume with a focused repumping beam (sec. 3.1.1) perpendicular to the excitation lasers. Only atoms in the small tube lighted by the repumping beam can be excited at all, so that only atoms in the region where the repumping beam overlaps with the excitation lasers can be excited. The details of the setup are given in [Dei05b].

### 4.2 Resonant energy transfer between mesoscopic ensembles

A next step towards coherent control over Rydberg-Rydberg interactions could be to demonstrate interactions between distinct mesoscopic systems of Rydberg atoms, which is of great interest for quantum information processing [Luk01]. Such an experiment is perfectly within the scope of our current setup. The basic idea is to create two distinct clouds of Rydberg atoms in different states, e.g. one in $32S$ and the other one in $33S$. By applying an electric field, the resonant energy transfer process

$$32S + 33S \rightarrow 32P + 32P$$  (4.2.1)

is switched to resonance. In neither of the single clouds $32P$ atoms can be produced, so that only interactions between atoms of both clouds can lead to a build-up of a P population. This means that the detection of P atoms is already a sufficient proof for interactions between the two mesoscopic ensembles, as long as the system is
properly prepared. We can use the setup where the two excitation lasers are crossed under a right angle described in the previous section to create the two distinct atom clouds. The repumping laser must be overlapped with the blue beam, while the red beam perpendicularly cuts out the excitation volume. First, a cloud of atoms with an approximate diameter of below 20 \( \mu \text{m} \) is excited to the Rydberg state \( 32S \). The excitation lasers have to be strongly focussed to achieve such a small volume. By using beam shaping techniques as described in the previous section, it is even possible to sharply define the edges of the cloud. Using a two-photon microwave transition, the atoms are transferred to the \( 33S \) state, which is energetically approx. 260 GHz above. The position of the red excitation laser is then translated along the the path of the blue excitation laser to select a second excitation volume, in which the atoms are then again excited to the \( 32S \) state by shortly switching on the red laser. The two clouds must not be separated further than approx. 50 \( \mu \text{m} \) to achieve a sufficient interaction strength, requiring precise control over the beam positions. During excitation of the second cloud the blue laser, which is resonant with the transition from \( 5P \) to \( 32S \), again lights the first cloud and stimulates all atoms that have not been transferred to \( 33S \) by the microwave pulse down to the \( 5P \)-level, from where they almost instantly decay to the ground state. It is important that all \( 32S \) atoms are removed from the first cloud, as otherwise resonant energy transfer processes between \( 32S \) and \( 33S \) could occur within this cloud. The electric field is then switched to resonance and the \( 32P \) population after a certain interaction time is detected. It should be possible to see an increase in the \( 32P \) population when the spacing between the two clouds is decreased. While such an experiment imposes high requirements on the accuracy of both the beam positions and the timing scheme, it would be an impressive demonstration that the interaction strength between two ensembles of atoms can be controlled.

4.3 Structured systems of Rydberg atoms

Rydberg atoms offer the unique possibility to control strong interatomic interactions. As we have seen resonant dipole-dipole interactions can be switched on and off by applying static electric fields. However, true control of the interactions requires control over the positions of the atoms, since the strength of the interaction critically depends on the interatomic spacings and also on the orientation of the atoms relative
to the electric field. So far, the atoms are randomly distributed in the atom cloud and their positions are different in every experimental cycle. This restricts the investigation of the resonant energy transfer to average properties of the whole cloud such as the populations of certain states at a certain interaction time. While this can give hints of the coherent nature of the underlying processes, it is necessary to explore reproducible systems, that are both sufficiently small and sufficiently structured, to obtain direct evidence of the quantum phenomena, that govern the dynamics of the Rydberg interactions.

Today, dipole traps and optical lattices offer a convenient way to create regularly structured, reproducible systems and thus control the positions of atoms. Moreover, the typical spacings and dimensions of such systems are very well suited to investigate interactions between Rydberg atoms, \textit{i.e.} the spacing between the nodes of a standing light wave produced by a CO$_2$-laser, in which Rb atoms can be trapped, is on the order of 5 $\mu$m, which corresponds to the typical interatomic spacing in a Rydberg gas at a density of approx. $10^9$ cm$^{-3}$. Furthermore, Mott-Insulator states of a Bose-Einstein condensate [Gre02] have opened up the possibility to precisely control the number of atoms trapped in each potential well formed by the optical lattice. Thus, it is possible to produce systems with a single atom per lattice site, which, when excited to Rydberg states, amounts to a reproducible system of regularly spaced interacting atoms. Together with spatially resolved detection of Rydberg atoms, such ensembles can provide entirely new possibilities to furnish direct evidence for coherent many-body phenomena in Rydberg systems.

We illustrate this by considering a linear chain of eight atoms at interatomic spacings of 10 $\mu$m (Fig. 4.1). The first atom in the chain is excited to the Rydberg state $\ket{32S_{1/2}}$, while the other atoms are in $\ket{32P_{3/2}}$. Excitation exchange processes (sec. 1.2.3) lead to a propagation of the S excitation along the chain, with a remarkable probability of more than 80 \% to find the last atom of the chain in the S-state after approx. 2.5 $\mu$s (Fig. 4.1). The direction of propagation is then reversed, while an increasing delocalization of the S-state occurs. After approx. 5 $\mu$s, the S-excitation has returned to the first atom with more than 60\% probability. While many experimental obstacles have to be overcome before realizing such an experiment, it could in principle be conducted the following way: Atoms are trapped in the nodes of a standing wave provided by a CO$_2$-laser, with a filling factor of one atom per site, thus creating the atom chain. The creation of a chain
4.3. Structured systems of Rydberg atoms

Figure 4.1: Propagation of a $|32S_{1/2}, m_j = +1/2\rangle$-state through a linear chain of atoms in $|32P_{3/2}, m_j = +3/2\rangle$ via excitation exchange processes calculated for eight atoms with an interatomic spacing of 10 $\mu$m. Interactions between each two atoms are taken into account. While the distribution of the S-state across the chain is shown in the upper part (each of the circles represents an atom), the time development of the probability to be in the S-state for each of the atoms is displayed below.
of cesium atoms with a defined atom number has already been demonstrated in an experiment [Sch04]. A larger number of atoms in the chain is excited to an S-state by excitation laser beams that are partially overlapped with the trapping beam. A microwave pulse then transfers those atoms to the P-state. Subsequently, strongly focused excitation beams perpendicular to the chain excite only atoms at one (or a few) sites to an S-state. The S-excitation(s) are then free to propagate along the chain, which can be measured through spatially resolved detection of the S-states after different interaction times. The detection scheme does not necessarily have to resolve single sites of the chain, as long as the region of the initial excitation can be clearly distinguished from regions where S-excitation can only appear when propagated along the chain.

The propagation of excitation along a linear chain of atoms could be a first approach to modelling simple energy transport phenomena with Rydberg atoms, as discussed in the introduction. It is clear that in the above example energy (namely the energetic difference between the P- and the S-state) is transported within $2.5 \, \mu s$ over a distance of $70 \, \mu m$ from one to the other end of the chain, with an efficiency of more than 80%. For the implementation of models for more complex systems, the capacities to precisely prepare and control single neutral atoms must be greatly improved.

### 4.4 Coherent excitation of a strongly interacting Rydberg gas

In all experiments discussed so far, the Rydberg systems were prepared in a certain initial state and then evolved exclusively due to the interatomic interactions. Another promising approach to investigating coherent properties in Rydberg systems is to study the interplay between Rydberg-Rydberg interactions and a light-field that drives coherent Rabi cycles between ground and Rydberg state. Here, both the interactions of each atom with the light and and interactions between the atoms determine the evolution of the system. Coherent excitation schemes have been successfully implemented in our experiment [Dei05b], so that the necessary tools are provided to investigate impacts of interatomic dipole-dipole interactions on the coherent excitation.
One of the effects that are to be expected is the so-called dipole blockade [], an interaction-induced blockade of excitation which was already discussed briefly in section 3.4.1. Such a blockade of excitation occurs when the interaction energy between a pair of atoms is so large that the excitation laser is too far detuned from the resonance of the pair to excite both atoms. If an ensemble of $N$ atoms is coherently excited with a Rabi frequency $\Omega_R$ in the regime of total blockade, i.e. only one of the $N$ atoms can be excited, this ensemble is predicted to perform oscillations between zero and one with a modified Rabi frequency $\sqrt{N}\Omega_R$ [Luk01]. Using the methods developed in chapter 2 we can simulate the dynamics of such a system and reproduce effects like the dipole blockade. Moreover, this allows us to explore intermediate regimes where the system is not completely blockaded and make predictions for the coherent excitation of such systems. We consider a system where the atoms can be in the ground state $g$ and in the Rydberg states $Ry$, $Ry'$ and $Ry''$. The ground state is coupled to $Ry$ via the Rabi frequency $\Omega_R$, while dipole-dipole interactions $V_{dd}$ exist between pairs of Rydberg atoms with interatomic spacing $R$ so that the energy transfer process

$$Ry + Ry \leftrightarrow Ry' + Ry''$$ (4.4.1)

is possible. This means that both $Ry - Ry'$ and $Ry - Ry''$ have non-vanishing

\[
\begin{aligned}
|Ry' Ry''\rangle, |Ry'' Ry'\rangle & \quad V_{dd} \delta \\
|Ry Ry\rangle & \quad 2\Delta \\
\end{aligned}
\]

\[
\begin{aligned}
\Omega_R \\
\Delta \\
\Omega_R \\
\end{aligned}
\]

|g Ry\rangle, \langle Ry g|}

\[
|g g\rangle
\]

**Figure 4.2:** Scheme of energy levels and couplings for pair states. The scheme is not drawn to scale. See text.
dipole moments $\mu$ and $\mu'$, with $V_{dd} = \mu\mu'/R^3$. For simplicity reasons, we do not consider the anisotropic part of the dipole-dipole interaction and assume $\mu = \mu'$. The couplings and energetic separations of the pair states are schematically shown in Fig. 4.2. The energetic separation between the pair states $|\text{Ry Ry}\rangle$ and $|\text{Ry' Ry''}\rangle$ is $\delta$, so that the energy transfer process is resonant for $\delta = 0$. The energetic detuning of the laser light from the atomic resonance $g \rightarrow \text{Ry}$ is denoted $\Delta$. Just as in the model presented in chapter 2, it is now possible to generate the Hamiltonian for ensembles of atoms involving two or more atoms and numerically solve the time-dependent Schrödinger equation, yielding the probabilities for the possible states of the system from which the populations of the different atomic states can be evaluated. This can then be averaged over random realizations of the system for a given atomic density with the Monte-Carlo scheme described in detail in section 2.2.2. Fig. 4.5 shows the time development of the entire Rydberg population, i.e. atoms in the states $\text{Ry, Ry'}$ and $\text{Ry''}$, simulated with a model involving four atoms, for different interaction strengths and detunings $\delta$ at an atomic density of $n = 10^{10}$ cm$^{-3}$, which corresponds to the density of ground state atoms achieved with the current setup. The excitation laser was assumed to be exactly on resonance ($\Delta = 0$), with a Rabi frequency $\Omega_R = 10$ MHz. For a large detuning of $\delta = 1$ GHz and small interactions strengths, the population oscillates between zero and four, i.e. each of the four atoms performs independent Rabi oscillations. With increasing interaction strength, the oscillations loose contrast and eventually become almost completely damped. Furthermore, the maximum population decreases, because closely spaced atom pairs experience a blockade excitation. For a detuning of $\delta = 100$ kHz, which is much smaller than the average interaction strength and thus not distinguishable from the resonance case $\delta = 0$, even the smallest interaction is sufficient to damp the oscillations. Higher interaction strengths further decrease the maximum population, until for $\mu = 4000$ au, a total excitation blockade is reached. The excitation is then evenly distributed among the four atoms (which one can check by looking at the excitation probabilities for each atom) and performs oscillations with a modified Rabi frequency $\sqrt{4}\Omega_R$. However, this is the point where the limitation of a model with four atoms is reached, so that it completely fails to describe the dynamics of a cloud of e.g. a hundred atoms. It then describes the average population of distinct ensembles of four atoms, as it can in principle be achieved by a Mott-Insulator state with a filling factor of four in an optical lattice. For smaller interaction strengths, it
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\[ \delta = 100\text{kHz} \]

\[ \delta = 1\text{GHz} \]

\[ \mu = 500\text{au} \]

\[ \mu = 1000\text{au} \]

\[ \mu = 2000\text{au} \]

\[ \mu = 4000\text{au} \]

**Figure 4.3:** Time development of the population of Rydberg atoms (atoms in the states \( Ry, Ry', \) and \( Ry'' \)) simulated with a model involving interactions between four atoms. The atomic density is \( n = 10^{10} \text{ cm}^{-3} \), the excitation laser is on resonance \( (\Delta = 0) \) and the Rabi frequency is \( \Omega_R = 10 \text{ MHz} \) for all graphs. There is a factor of four in interaction strength between neighboring rows of diagrams, as the interactions strength is proportional to \( \mu^2 \). The graphs are averages over 30 realizations of the system, which is sufficient to reproduce the main features of the curves very well (we checked this by comparison with selected curves obtained as averages over more realizations). See text.
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Figure 4.4: Reduced level scheme for the excitation of strongly dipole-coupled pairs of Rydberg atoms. The part of the Hamiltonian made up of the pair states $|\text{Ry Ry}\rangle$, $|\text{Ry'} \text{Ry}''\rangle$ and $|\text{Ry}'' \text{Ry'}\rangle$ which are coupled by the dipole-dipole interactions can be diagonalized. One obtains the two uppermost states as eigenstates which are detuned by $\pm \sqrt{2} \mu^2 / R^3$ depending on the distance $R$ between the two atoms, plus a state $1/\sqrt{2} (|\text{Ry} \text{Ry}'\rangle - |\text{Ry}'' \text{Ry}'\rangle)$, which does not couple to the light field. Likewise, only the symmetric combination of $|\text{g Ry}\rangle$ and $|\text{Ry g}\rangle$ has to be taken into account.

is justified to clip some of the interactions between the atoms and partition the cloud in subensembles of four, as the interaction on average is only sufficiently strong to reach some neighboring atoms during the typical interaction time. For very large interactions as in the case of a total blockade however, more extended subsystems have to be considered to properly model the dynamics of a larger cloud of atoms, as interactions between far separated atoms have to be taken into account as well.

Experiments on coherent excitation of a Rydberg gas on a resonance are under way in our group [RL06]. Like the measurements on resonant energy transfer processes described in chapter 3, such experiments suggest that coherent processes govern the dynamics of a strongly interacting Rydberg gas. A proof of this coherence could be the observation of oscillations of Rydberg states that can only be populated through interaction processes, in our case Ry' and Ry''. Due to the great range of
interaction strengths present in a randomly distributed gas, such oscillations are usually averaged out and are thus not observable. However, by coherently "driving" the system with the right frequency, it may be possible to see remnants of such oscillations. The idea is explained in Fig. 4.4, where the relevant atom pair states on resonance ($\delta = 0$) are depicted: The ground state $|gg\rangle$ only couples to the symmetric state $1/\sqrt{2} (|gRy\rangle + |Ryg\rangle)$ with a Rabi frequency modified by a factor of $\sqrt{2}$. As described in detail in section 1.2.1, the part of the Hamiltonian made up of the pair states $|RyRy\rangle$, $|Ry'Ry''\rangle$ and $|Ry''Ry'\rangle$ which are coupled by the dipole-dipole interactions can be diagonalized, yielding the eigenstates depicted in Fig. 4.4. The light field couples to two of them, which are detuned from the atomic resonance by $\pm \sqrt{2} \mu^2 / R^3$ (Fig. 4.4). The frequency of the excitation laser can now be detuned from the atomic resonance frequency in such a way, that it is approximately resonant with one of the eigenstates, which is then preferably populated. The system then performs oscillations between the ground and the selected eigenstate, with the exact shape and frequency depending e.g. on the detuning $\Delta$ from the intermediate state (Fig. 4.4). Such effective two-level systems are treated in detail in the diploma thesis of J. Deiglmayr [Dei05a]. As the states $Ry'$ and $Ry''$, which do not directly couple to the light field, are contained in the selected eigenstate, one would observe oscillations of the population of these states. The problem is now that the effective energy shift due to the dipole-dipole interactions depends on the spacing $R$ between the two interacting atoms, so that the excitation laser is only resonant with pairs at the correct spacing. However, the laser can be tuned to be resonant with pairs at the spacing which is most likely for the respective atomic density. The hope is that the variation of interatomic spacings $R$ is not sufficient to completely wash out the oscillations. This can be simulated using the techniques described above, particularly with the model involving four atoms, which also accounts for many-body effects that might further reduce the contrast of possible oscillations. Fig. 4.5 shows the simulated populations of atoms in the state $Ry'$ which can only be populated through energy transfer processes for two different interaction strengths. Although averaged over 200 random realizations of the system, a first oscillation period is clearly visible, with a contrast of approx. 50%. This can only be interpreted in the

---

$^{1}$The most likely interatomic spacing at a density of $n = 10^{10} \text{ cm}^{-3}$ according to the next neighbor probability function (Appendix B) is approximately $2.75 \mu \text{m}$, so that the most likely interaction strength and thus energy shift can be easily calculated.
Figure 4.5: Simulation of the number of atoms in the state \(Ry'\), which can only be populated through energy transfer processes, with an atom involving four atoms. The maximum number, \(i.e.\) 100\% of the atoms, is 4. The graphs were obtained on resonance (\(\delta = 0\)) for a density of \(n = 10^{10}\) cm\(^{-3}\). The dipole matrix elements of \(\mu\) were 500 au and 1000 au, so that the most likely interaction strengths are 10.4 MHz and 41.6 MHz. The detunings \(\Delta\) of the excitation laser were chosen by approx. a factor of \(\sqrt{2}\) smaller than the most likely interaction strengths, so that is is resonant to excite pairs at the most likely spacing. See text.

way, that atoms are transferred to \(Ry'\) and then back to a different state through energy transfer processes, which is a clear signature for coherence in the process. If one succeeds to observe such remnants of oscillations in an experiment, this can be seen as a first direct evidence for coherence in interactions between Rydberg atoms.
Appendix A

Calculation of dipole matrix elements

In order to evaluate the dipole matrix element between two atomic states characterized by their quantum numbers \( n, j \) and \( m_j \) it can be decomposed in a radial and an angular part. The radial part can be calculated as

\[
\mu_{\text{rad}} = \int_0^\infty r^2 R_{n_1j_1}(r) r R_{n_2j_2}(r) \, dr
\]

where \( R_{nj} \) is the radial wave function of the respective state. For Rydberg atoms the evaluation of (A.0.1) is relatively simple as hydrogen-like wave functions that are analytically available can be used [Zim79, Mar94].

The angular part reflects the geometry of the system, \textit{i.e.} the orientation of the quantization axis of the system. In order to evaluate the dipole-dipole interaction matrix elements in section 2.2.1 the angular dipole matrix element between \( P_{3/2, m_j=+3/2} \) and \( S_{1/2, m_j=+1/2} \) has to be evaluated. As the position operator acts only on the orbital and not on the spin-part of the wave function it is in this special case convenient to work in a basis, where spin and orbital angular momentum are not coupled. \( \vec{r} \) denotes the oriented unit vector \( \vec{r}/|\vec{r}| \).

\[
\left\langle P_{3/2, m_j=+3/2} \left| \vec{r} \right| S_{1/2, m_j=+1/2} \right\rangle = \\
\left\langle s = 1/2, m_s = +1/2 \left| l = 1, m_l = +1 \left| \vec{r} \right| l = 0, m_l = 0 \right\rangle \right| s = 1/2, m_s = +1/2 \right\rangle = \\
\left\langle l = 1, m_l = +1 \left| \vec{r} \right| l = 0, m_l = 0 \right\rangle
\]

(A.0.2)
To evaluate this further the position operator can be written in the spherical basis $r_+, r_0, r_-$, where only $r_+$ has a non-zero matrix element. This can again be explained by the Wigner-Eckart-Theorem as the Clebsch-Gordon coefficients for the $r_0$ and $r_-$ matrix elements vanish. (A.0.2) can be evaluated following [Tho94] which leads to

$$
\langle P_{3/2, m_j=+3/2} | \hat{r}_+ | S_{1/2, m_j=+1/2}\rangle = \langle l = 1, m_l = +1 | \hat{r}_+ | l = 0, m_l = 0 \rangle = 1/\sqrt{3}. 
$$

(A.0.3)

Thus the entire dipole matrix element can be evaluated. A similar procedure yields the other relevant matrix elements

$$
\langle S_{1/2, m_j=+1/2} | \hat{r}_- | P_{3/2, m_j=+3/2}\rangle = -1/\sqrt{3} \quad \text{(A.0.4)}
$$

$$
\langle S_{1/2, m_j=-1/2} | \hat{r}_+ | P_{3/2, m_j=-3/2}\rangle = -1/\sqrt{3} \quad \text{(A.0.5)}
$$

$$
\langle P_{3/2, m_j=-3/2} | \hat{r}_- | S_{1/2, m_j=-1/2}\rangle = 1/\sqrt{3}. \quad \text{(A.0.6)}
$$
Appendix B

Next neighbor probability functions

Consider a ball with radius $R$ and volume $V = \frac{4}{3}\pi R^3$ around one designated particle, in which particles at a homogeneous average density $n$ are present. The number $N$ of particles in the ball therefore is $N = nV$.

For every single particle, the probability density at a distance $r$ from the designated particle in the origin is

$$p_{\text{single}}(r) = \frac{1}{V} \cdot 4\pi r^2.$$ (B.0.1)

The probability to be found within a distance smaller than $r$ from the designated particle is

$$P_{\text{single}}[0, r] = \frac{1}{V} \cdot \frac{4}{3}\pi r^3,$$ (B.0.2)

which is the integral over $p_{\text{single}}$ from 0 to $r$. Similarly, the probability to be found at a distance greater than $r$ from the designated particle in the origin is

$$P_{\text{single}}[r, R] = 1 - \frac{1}{V} \cdot \frac{4}{3}\pi r^3.$$ (B.0.3)

For $N$ particles, the probability density $p_{\text{nn}}(r)$ to find the closest particle to the designated particle (i.e. the next neighbor) at distance $r$ is then the probability to find one particle exactly at $r$ and all other particles at distances greater than $r$, multiplied by a factor $N$ on one due to combinatorics:

$$p_{\text{nn}}(r) = \binom{N}{1} \frac{1}{V} 4\pi r^2 \times \left(1 - \frac{1}{V} \frac{4}{3}\pi r^3\right)^{N-1}$$ (B.0.4)
This expression is normalized, i.e. \( \int_0^R \! p_{nn}(r) \, dr = 1 \), and independent of \( R \) as long as \( R \) is chosen sufficiently large.

Similarly, the probability \( P^m(r) \) to find exactly \( m \) particles within a distance \( r \) of the designated particle is the probability to find \( m \) particle at distances smaller than \( r \) and \( N-m \) particles at distances greater than \( r \), multiplied again by a combinatorial factor:

\[
P^m(r) = \binom{N}{m} \left( \frac{1}{V} \cdot \frac{4}{3} \pi r^3 \right)^m \times \left( 1 - \frac{1}{V} \cdot \frac{4}{3} \pi r^3 \right)^{N-m} \tag{B.0.5}
\]

The probability \( P^{>m}(r) \) to find more than \( m \) particles within a distance \( r \) of the designated particles is then

\[
P^{>m}(r) = \sum_{i=m}^{N} P^i(r), \tag{B.0.6}
\]

while the average number \( N_{av}(r) \) of particles within \( r \) of the designated particle can be calculated to

\[
N_{av}(r) = \sum_{i=1}^{N} \, i \cdot P^i(r). \tag{B.0.7}
\]

All the above probability functions are independent of \( R \) and thus \( N \), as long as \( R \) is chosen sufficiently large compared to \( r \), i.e. in the limit of large \( N \).
Bibliography


