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TUTORIAL

An experimental and theoretical guide to strongly interacting Rydberg gases

Robert Löw1, Hendrik Weimer2,3, Johannes Nipper1, Jonathan B Balewski1, Björn Butscher1, Hans Peter Büchler4 and Tilman Pfau1

1 Physikalisches Institut, Universität Stuttgart, 70569 Stuttgart, Germany
2 Physics Department, Harvard University, 17 Oxford Street, Cambridge, MA 02138, USA
3 ITAMP, Harvard-Smithsonian Center for Astrophysics, 60 Garden Street, Cambridge, MA 02138, USA
4 Institut für Theoretische Physik III, Universität Stuttgart, 70569 Stuttgart, Germany

E-mail: r.loew@physik.uni-stuttgart.de

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Abstract

We review experimental and theoretical tools to excite, study and understand strongly interacting Rydberg gases. The focus lies on the excitation of dense ultracold atomic samples close to, or within quantum degeneracy, high-lying Rydberg states. The major part is dedicated to highly excited S-states of rubidium, which feature an isotropic van der Waals potential. Nevertheless, the setup and the methods presented are also applicable to other atomic species used in the field of laser cooling and atom trapping.

(Some figures may appear in colour only in the online journal)

Nomenclature

Here, we list the nomenclature of some important quantities to avoid misunderstanding due to varying notations in other publications.

\( n \) Density of ground state atoms
\( n_r \) Density of Rydberg atoms
\( \Omega_f \) Coupling strength of the first excitation step (here the 5S–5P transition in rubidium at 780 nm)
\( \Omega_b \) Coupling strength of the second excitation step (here the 5P–\( n \)S,\( n \)D transition in rubidium at 480 nm)
\( \Omega_R \) Effective Rabi frequency for the two photon excitation
\( \Omega \) Collective Rabi frequency
\( r_b \) Blockade radius
\( \alpha \) Rescaled coupling strength \( \Omega_R \)
\( \delta \) Detuning with respect to the lower transition, here 5S–5P
\( \Delta \) Detuning with respect to the two photon transition, here 5S–\( n \)S
\( \eta \) Rescaled detuning \( \Delta \)

1. Introduction

The field of Rydberg atoms has been revolutionized by the combination with ultracold atomic gases. The availability of commercial high-power diode laser systems allows for efficient excitation into electronically highly excited states with excellent frequency resolution. Here, the major motivation behind many experiments is to make use of the strong interaction mechanism among Rydberg atoms, which become especially apparent at large atomic densities as found in ultracold gases. On the other hand, the precise spectroscopy of the interaction energies is supported by the availability of trapped atoms, the almost negligible Doppler effect given by small temperatures and the possibility of preparing all atoms in a specific quantum state with high fidelity. Combining this control over internal and external degrees of freedom for atoms, molecules and ions with switchable long-range interactions establishes an ideal basis for applications in quantum simulation, quantum computation and quantum optics.

Generally, there exist two pathways to prepare a controllable ensemble of strongly interacting particles. One
way is to start with a system of weakly interacting particles as in the case of quantum degenerate gases and then try to implement strong interactions among them, e.g., with Feshbach resonances [1], optical lattices [2], high-finesse resonators [3], or by extreme cooling in order to make the initially weak interaction energy the dominant scale [4]. The second approach utilizes already strongly interacting particles as polar molecules [5] and ions, where the latter already constitutes the best working quantum computer [6, 7] and universal quantum simulator [8] in the world. The drawback of the latter approach is that the presence of interactions requires the particles to be restricted to specific geometries.

At this point, Rydberg states might be an ideal add-on to both approaches, since they are switchable and exhibit interaction strengths comparable to ionic systems [9].

The major interaction mechanisms between two highly excited atoms at large distances is typically given by the van der Waals interaction and for smaller distances by the dipole–dipole interaction [9]. The effects of both interaction types have been observed in laser cooled clouds [10–18] and also in optically and magnetically trapped clouds [19–21]. A prominent effect of the strong interaction mechanism is an excitation blockade in which the energy of a Rydberg state is shifted out of resonance with respect to the exciting laser by a neighbouring Rydberg atom. This excitation blockade is typically referred to as the dipole blockade, without distinguishing explicitly between real dipole–dipole interactions $\sim 1/r^3$ and its off-resonant counterpart, i.e. the van der Waals interaction $\sim 1/r^6$.

The most drastic consequence of the dipole blockade is the occurrence of a quantum phase transition to a crystalline phase of Rydberg excitations [22–24]. The parameters controlling the transition can be tuned with external laser fields. While this Rydberg crystal has so far eluded direct observation, critical fluctuations of this quantum phase transition have already been reported [25] in a non-adiabatic manner. The realization of the crystalline phase, which is the ground state of the system, would require adiabaticity during preparation [26–29].

The blockade effect is also used in a more involved approach with exactly two atoms located in a certain distance establishing the prototype of a quantum gate [30, 31]. Since the loading of individual sites is still probabilistic [32], the scalability of this approach is limited as long as there is no deterministic loading scheme available [33, 34]. An alternative way to achieve a large quantum register with indistinguishably resolved sites is a Mott-insulator state with single-site resolution [35, 36] or a switchable local electric field, e.g., produced by an electron beam [37] that allows an addressing via the spatially dependent Stark shift.

Although the first proposal to use the interaction between two spatially resolved Rydberg atoms [38] seemed quite involved in the year 2000, it triggered a large theoretical effort and several experimental groups started to implement these ideas. There exist elaborate proposals for the realization of a universal quantum simulator [39–41] and various ways to implement a quantum computer [9, 38, 42–44]. Some of these schemes rely on collective states, where one Rydberg excitation is shared by several atoms [42, 45, 46]. In this tutorial, we will focus on the many-particle character of the collective states in an ensemble of ultracold atoms with extensions well larger than the blockade radius. A detailed understanding of the excitation dynamics in these systems into a strongly correlated quantum system, its microscopic description, the limitations of the effective models used, the sources of decoherence and dephasing and finally of the applicable experimental techniques will not only be helpful for the implementation of various quantum simulation and quantum computation protocols but also for other tasks involving interacting Rydberg atoms. One could be the idea of combining interacting Rydberg atoms with quantum degenerate gases [47–50] to render new interaction mechanisms. Besides the effects of the strong interaction on the material wavefunctions, there also exists a strong backaction on the radiation field, which can produce and also analyse various kinds of non-classical light fields [26, 51, 52].

In the following, we will first describe the basic properties of non-interacting Rydberg atoms and deduce from this the applicable energy and time scales for meaningful experiments. These considerations also set the preconditions and limitations to the techniques for the excitation of ultracold trapped atoms to Rydberg states and their subsequent detection. In the next section, we will include the interaction among Rydberg states and will derive a mean-field model for the strongly interacting case, which describes quite well the experimental findings. Finally, we will summarize experimental tools and results on strongly interacting quantum systems based on Rydberg atoms.

2. General properties of Rydberg atoms

The properties of Rydberg atoms are similar to that of ground-state atoms but are severely enlarged with the principal quantum number $n$ [53]. Table 1 shows an overview of some important values and their corresponding scaling.

2.1. Electronic structure and lifetimes of highly excited rubidium atoms

The largest fraction of all laboratories working with ultracold atoms use alkalis, since their simple hydrogen-like structure makes their excited states accessible and controllable with only a few laser frequencies. For most of them, the deviation from a hydrogen atom is caused by the core electrons, which add a repulsive Coulomb potential for the Rydberg electron and a simple $1/r$ Coulomb potential is no longer applicable. This effect can be faced in a phenomenological manner by introducing a quantum defect $\delta(n, j, l)$, which results in an altered Rydberg formula:

$$E(n, j, l) = -\frac{R'}{(n - \delta(n, j, l))^2} = -\frac{R'}{(n\gamma^2)^2}, \quad (1)$$

where $R' = Ry/(1 + m_e/m_{\text{nucleus}})$ is the specific Rydberg constant for the element in question, e.g., $R' = 109\,736,605 \text{ cm}^{-1}$ for rubidium ($R' = 109\,737,316 \text{ cm}^{-1}$ for hydrogen). The effective quantum number $n'\gamma$ mostly depends on $l$ and is in lowest order for rubidium determined by $\delta(l = 0) = 3.13$, $\delta(l = 1) = 2.64$, $\delta(l = 2) = 1.35$, $\delta(l = 3) = 0.016$.
and \( \delta(l > 3) \approx 0 \) [57–59]. More accurate values for \( nS \) and \( nD \) states have recently been measured [60]. As a consequence of the \( l \)-dependent quantum defects, the degeneracy of the \( s, p, d \) and \( f \) states is lifted and only states with \( l > 3 \) show a hydrogen-like behaviour. With the knowledge of the energies of the individual excited states, it is possible to solve the Schrödinger equation analytically [61] or numerically to obtain also the radial wavefunctions. For that purpose, one has to add a term \( V_p = -\frac{Z}{r} e^2 \) to account for the Coulomb potential of the core electrons in terms of a core polarizability, e.g., \( \alpha_0 = 9.023 \) au in the case of rubidium [62]. Since the probability of finding a Rydberg electron inside the core is quite small, the quality of calculated wavefunctions are often of sufficient accuracy to be used to reliably compute dipole matrix elements, while improved model potentials exist for other cases [63]. These dipole matrix elements are crucial for the calculation of the excited state lifetime, the polarizability, the van der Waals interaction and many more properties [53].

The lifetime of Rydberg states is determined by the radiative decay to lower lying levels but also by transitions to higher (and lower) lying states induced by blackbody radiation as shown in figure 1. The major contribution to the lifetime is given by spontaneous decay events to low-lying levels where the product of interaction strength and density of states of the electromagnetic field scaling as \( \omega^3 \) outweighs the decreasing dipole moments \( \propto n^{1.5} \ldots \propto n^{1.55} \). For Rydberg state \( n \geq 40 \), the absorption and stimulated emission of thermally occupied infrared modes to neighbouring states start to dominate the lifetime. The latter effect can be eliminated by using a cryogenic environment as it is done in cavity QED experiments with Rydberg atoms [65].

The transition rate between two states \( i \) and \( f \) with a dipole matrix element \( |i| r |f \rangle \), separated by the energy \( \hbar \omega_{if} \), is typically expressed in terms of an Einstein \( A \) coefficient for the spontaneous decay

\[
A = \frac{2e^2 \omega_{if}^3}{5\hbar^2 c^4} |i| r |f \rangle |
\]

and an Einstein \( B \) coefficient \((B = AN(\omega))\) for stimulated emission as well as absorption of blackbody radiation. The number of blackbody photons \( N(\omega) \) per mode at a given temperature \( T \) is given by

\[
N(\omega) = \frac{1}{\frac{\hbar}{k_B} T + 1}.
\]

**Table 1.** With some explicit values for the 43S state in rubidium, it is possible to get an idea for the framework and the boundary conditions for an experimental setting involving Rydberg atoms. With the given scaling laws \((n^*)^4\), one can also find an estimate for the physical situation with other Rydberg states.

<table>
<thead>
<tr>
<th>Property</th>
<th>Expression</th>
<th>((n^*)^4)</th>
<th>\text{Rb(5S)}-ground state</th>
<th>\text{Rb(43S)}-Rydberg state</th>
</tr>
</thead>
<tbody>
<tr>
<td>Binding energy (E_{n^*} )</td>
<td>(-nhc/\omega n^*)</td>
<td>((n^*)^{-2})</td>
<td>4.18 eV</td>
<td>8.56 meV</td>
</tr>
<tr>
<td>Level spacing (E_{n^<em>} - E_{n^</em>+1})</td>
<td>((n^*)^{-3})</td>
<td>2.50 eV (5S–6S)</td>
<td>413.76 (\mu\text{eV} = 100.05 \text{GHz} ) (43S–44S)</td>
<td></td>
</tr>
<tr>
<td>Orbit radius (r )</td>
<td>(\frac{1}{2} (3(n^*)^2 - l(l + 1)) )</td>
<td>((n^*)^2)</td>
<td>5.632 (a_0)</td>
<td>2384.2 (a_0)</td>
</tr>
<tr>
<td>Polarizability (\alpha )</td>
<td>((n^*)^2)</td>
<td>(-79.4 \text{ MHz (V cm}^{-1})</td>
<td>(-17.7 \text{ MHz (V cm}^{-1})</td>
<td></td>
</tr>
<tr>
<td>Lifetime (spontaneous decay) (\tau )</td>
<td>(\tau = \frac{1}{\omega} (n^*)^6)</td>
<td>((n^*)^5)</td>
<td>(5P_{1/2})–(5S_{1/2}): 26.2 ns</td>
<td>43.2 (\mu\text{s} ) at 300 K including BBR</td>
</tr>
<tr>
<td>Transition dipole moment (\langle S</td>
<td>\vec{r}</td>
<td>nS\rangle)</td>
<td>((n^*)^{-1.5})</td>
<td>5(P_{1/2})–(5P_{3/2}): 2.227 (e\alpha_0)</td>
</tr>
<tr>
<td>Transition dipole moment (\langle nP</td>
<td>\vec{r}</td>
<td>(n+1)S\rangle)</td>
<td>((n^*)^{-2})</td>
<td>4707 au</td>
</tr>
<tr>
<td>van der Waals coefficient (\gamma )</td>
<td>((n^*)^{1.5})</td>
<td>-1.697 (10^9) au</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 1. Stacked bar chart for the calculated transition rates of the 43\(S_{1/2}\) state of rubidium (top) and a 30\(\times\) enlarged view (bottom). The Einstein \(A\) coefficients for the spontaneous decay are shown in blue \(nP_{1/2}\) and light blue \(nP_{3/2}\) for all energetically lower lying states \((5 \leq n \leq 42)\). The sum of these coefficients results in a lifetime due to spontaneous decay of 80.8 \(\mu\text{s}\). An additional loss of population is caused by blackbody radiation, which favours neighbouring states shown in red \(nP_{3/2}\) and orange \(nP_{3/2}\). The total lifetime (including blackbody radiation at 300 K) is then reduced to 42.3 \(\mu\text{s}\), which corresponds to a Lorentzian linewidth of 3.76 kHz and is in good agreement with more elaborate calculations carried out in [64].
Rydberg state and the second sum runs over all allowed dipole transitions, however neglecting transitions to ionized states. A measurement of the radiative lifetime by an integral detection of remaining Rydberg states after some delay of the excitation could be falsified by nearby Rydberg states, which have been populated by blackbody radiation. To measure the lifetimes of Rydberg states accurately, it is therefore necessary to implement state-sensitive detection schemes [66, 67].

2.2. Rubidium Rydberg atoms in electric fields

The large polarizability of Rydberg states is a welcome feature for experimentalists, since it allows for simple and fast manipulation of the excited-state energies by applying small electric fields \( E \). With the knowledge of the dipole matrix elements, it is straightforward to find the new eigenenergies in the presence of an electric field [68]. The corresponding Stark map, as shown in figure 2, exhibits two kinds of Stark-shifted states. If the degeneracy is lifted by the quantum defect, the Stark effect is quadratic at low fields \( E_{\text{Stark}} = \alpha E^2/2 \) with the polarizability \( \alpha \), and linear in the case of degeneracy, which corresponds to a permanent dipole moment. In the case of rubidium, the polarizability for the 43S states is \( \alpha = -17.7 \text{ MHz (V cm}^{-1})^{-2} \), which shifts this state at a field of only 1 V cm\(^{-1}\) already 350 linewidths to the red. For states with higher angular momentum, the manifold of magnetic substates discloses a unique pattern in the presence of an electric field [69], which can be used to calibrate the actual electric field [70].

In zero electric field, any atom in a single-parity eigenstate does not exhibit a permanent dipole moment. Nevertheless, fluctuations of the electron distribution lead to momentary dipole moments, which induce dipole moments in the other atoms. In second-order perturbation theory, the interaction energy is proportional to the square of the dipole moment for each atom divided by the energy spacing of the pair states [71]. The largest contribution is given by the dipole moments to nearby Rydberg states, which results in a scaling behaviour with respect to the principal quantum number as \( C_6 \sim n^{11} \). A numerical evaluation of the interaction potentials gives, e.g., for two rubidium Rydberg atoms in an S-state and 30 < \( n < 95 \) [72] in atomic units, a simple scaling law

\[
C_6 = n^{11} (11.97 - 0.8486n + 3.385 \times 10^{-3}n^2).
\]

Various interaction parameters have been calculated with perturbative methods [71] but also by diagonalization [73–75]. For rubidium in the 4S state, the van der Waals coefficient is \( -1.697 \times 10^{19} \text{ au} = -h \times 2441 \text{ MHz} \mu\text{m}^6 \), which is not only much larger than in the ground state 5S \( (C_6 = \pm 4707 \text{ au} = +h \times 677 \text{ nHz} \mu\text{m}^6) \) but is also opposite in sign resulting in a repulsive potential. This is a quite useful feature since attractive forces essentially lead to inelastic collisions and ionization [76–80]. At small distances, the van der Waals interaction becomes comparable to the energy differences between the relevant pair states, the Förster defect, and perturbative methods are no longer valid. At smaller distances, the interaction potential adopts a dipolar \( \sim 1/r^3 \) character, which is valid down to the LeRoy radius where the exchange interaction of the electrons can no longer be neglected. For the 4S state in rubidium, the Förster defect is 3 GHz [81], which corresponds to the van der Waals interaction energy at a distance just below 1 \( \mu\text{m} \). However, the distance between two rubidium atoms in evaporatively cooled clouds may easily reach smaller values leading to interaction energies of many tens of GHz, an energy much larger than all other relevant energies scales in the system. Even at a distance of 5 \( \mu\text{m} \), the interaction energy is still 150 kHz, which establishes in combination with a comparable coupling strength of the driving laser fields and sufficiently dense atomic samples the domain of strongly interacting Rydberg gases.

3. Rydberg excitation of ultracold atomic gases

For experiments with Rydberg atoms in ultracold gases, additional equipment and methods for excitation and detection of Rydberg states are required, without affecting the cooling and trapping procedure. The required level of performance of these new tools is determined by the properties of the Rydberg atoms. For example, the laser light for the Rydberg excitation has to be sufficiently intense and narrow in frequency to achieve a coherent evolution within the natural lifetime. The 4S in rubidium has a lifetime (including BBR at 300 K) of 42.3 \( \mu\text{s} \), which corresponds to a linewidth of 3.76 kHz and the excitation laser should be accordingly narrow. The lifetime also sets the timescale for the experiments and by this the desired coupling strength of the laser system. To observe, e.g., a full Rabi oscillation within the radiative lifetime of

![Figure 2. Calculated Stark map including LS coupling with respect to the 43S_{1/2} state in ^{87}\text{Rb}. Only states with } L \geq 4 \text{ exhibit hydrogen like linear Stark fans. For lower } L \text{ the states are shifted quadratically as it can be seen in the inset for the 43S_{1/2} state.}
the 43S state in rubidium, the Rabi frequency $\Omega_R$ has to be accordingly strong and the linewidth of the excitation sufficiently narrow. Given that the dipole matrix elements for excitation into Rydberg states are typically quite small, a sufficient strong laser source with several tens of milliwatts is appropriate. But many proposals, e.g., for quantum computing and quantum simulation will only be manageable with several hundred milliwatts of available laser power.

After excitation, one also wants to detect the produced Rydberg atoms efficiently. Absorptive and fluorescing methods, as typically used for ground-state atoms, are impractical since the spontaneous scattering rates are roughly a thousand times smaller. One exception is the Rydberg states of alkaline earths, where the second non-excited electron can be used for optical detection [82]. The most common method relies on efficient and fast ion detection with multichannel plates (MCPs) or channeltrons, which can reach counting efficiencies of 90%. To do so, one has to field ionize the Rydberg states first either with a strong electric field or by a far infrared laser pulse [83]. The field plates inside the vacuum chamber to ionize the atoms and to guide them to the ion detectors are also very useful to apply moderate electric fields to shift the Rydberg levels in and out of resonance with the driving laser fields, to subtract unwanted ions or to implement a Ramsey interferometer [84].

The other contributor, ultracold atomic gases, is 15 years after the first observation of Bose–Einstein condensation [85, 86] of the shelf in many atomic physics laboratories. There exist countless tools and methods to control the internal and external degrees of freedom, where most of them also get along with the technical constraints of highly excited Rydberg states. For precision spectroscopy, trapped gases at extremely low temperatures are an ideal sample. The Doppler effect is largely reduced, the interaction time with the probing light field is limited either by the excited state lifetime or by the lifetime of the trap, which can be in minutes. Actually on the typical timescales of Rydberg experiments of a few microseconds, the ultracold atoms move only a fraction of the wavelength of the exciting lasers, which has led to the expression of frozen Rydberg gases. Additionally, the high densities of fully polarized samples make the interaction potential among Rydberg states accessible to a spectroscopic precision limited only by the excited state lifetime. Such measurements can also be taken in zero electric and magnetic fields by switching of all trapping potentials (magnetic or dipole) during excitation.

### 3.1. Two-photon excitation of ultracold atoms into Rydberg states

The excitation into Rydberg states with principal quantum numbers ranging from $n = 20$ up to the ionization threshold is usually accomplished by a two-photon and sometimes by three-photon excitation schemes. For a single-photon excitation of rubidium to Rydberg states, one would have to use light at 297 nm, which is difficult to produce and would also not allow one to access the desired S-states. In rubidium, there are, in principle, two two-photon excitation schemes feasible; here, we use light at 780 and 480 nm for the 5s–5p–ns/nd scheme as shown in figure 3 but also the alternative route 5s–6p–ns/nd with 420 and 1016 nm lies in the range of standard diode laser systems.

In all experiments, we detune the red laser far from resonance to minimize spontaneous scattering of photons via population of the 5p state, which has a lifetime of 26 ns [87]. In doing so, we choose a blue detuning to avoid all the lower lying hyperfine levels of the 5p state as shown in figure 3. For large enough detunings $\delta$, the intermediate state can be eliminated and one ends up with an effective two-level system with a total coupling strength $\Omega_R = \sqrt{\Omega_i^2 + \Omega_b^2} + \Delta^2$. To obtain coherent coupling between the ground state and the Rydberg state, it is necessary to achieve an effective Rabi frequency $\Omega$ well larger than the linewidth of the Rydberg state (3.76 kHz for 43S) or of the driving laser fields. This is easily achieved for the 5s–5p transition with only a few milliwatts of laser power but the dipole matrix element for the second step (5p–43s) is roughly $10^3$ times smaller requiring $10^6$ more laser power for a comparable coupling strength.

The typical excitation scheme excites in a first step a spin polarized sample in the 5S1/2 $F = 2, m_F = 2$ state...
into the $5P_3/2 \rightarrow F = 3, m_F = 3$ state and in the second step into the $nS_{1/2} \rightarrow J = 1/2, m_J = 1/2$ Rydberg state. Note that the hyperfine structure of the highly excited state cannot be resolved and we return here to the $J, m_J$ basis. Following the standard notation \cite{88, 89}, we can compute the dipole matrix element for the second step by

$$
\langle Fm_f | er_q | Jm_J \rangle = \langle J | er | J \rangle \times \sqrt{(2F + 1)(2J + 1)(-1)^{J+L+1+m_F+m_J+q}} 
\times \left( \begin{array}{ccc} J & I & F \\ m_f + q & m_F - m_J - q & -m_F \\ J' & 1 & J \\ m_f & q & -m_F - q \end{array} \right). 
$$

(5)

The reduced matrix element $\langle J | er | J \rangle$ is related to the pure radial part $\langle R_{dl} | er | R_{dl} \rangle$ as

$$
\langle J | er | J \rangle = \langle R_{dl} | er | R_{dl} \rangle \times \sqrt{\frac{\text{max}[L, L']}{2L + 1}} \left( \begin{array}{ccc} L & L' & 1 \\ J' & J & S \end{array} \right). 
$$

(6)

For the transition from the $5P_{3/2}$ into the $43S_{1/2}$ state, we obtain the purely radial matrix element $\langle R_{5,1} | er | R_{4,0} \rangle = 0.0178 \text{ 86a} \text{-} \text{a} \text{a}$ leading to a total dipole matrix element of

$$
\langle 5P_{3/2}, F = 2, m_F = 2 | er_{q=+1} | 43S_{1/2}, J' = 1/2, m_J = 1/2 \rangle = 0.0103 \text{ 31a} \text{-} \text{a} \text{a}. 
$$

(7)

State-of-the-art diode laser systems based on tapered amplifiers with subsequent frequency doubling deliver about $P = 300 \text{ mW}$ of blue light, which corresponds to an intensity of $I = 9.55 \text{ MW m}^{-2}$ at the centre of a focused Gaussian beam with a width of $w_0 = 100 \mu \text{m}$. With the electric field $E = \sqrt{2I/\delta e}$, we can compute the coupling strength $\Omega_c = dE/h = 2\pi \times 11.2 \text{ MHz}$. In combination with a coupling strength for the first transition of $\Omega_1 = 2\pi \times 100 \text{ MHz}$ and a detuning $\delta = 400 \text{ MHz}$, we obtain a resonant $(\Delta = 0)$ two-photon coupling strength $\Omega_\text{gap} = 2\pi \times 1.4 \text{ MHz}$.

For higher intensities, one can consider dye jet lasers or dye amplifiers based on Bethune cells where the latter has been shown to produce coupling strengths in the GHz regime \cite{90}.

Another important consequence of rather narrow linewidths of Rydberg states is that the Doppler broadening becomes comparable to the temperature of ultracold atomic clouds in the micro-Kelvin regime. In figure 4, the FWHM widths $\Delta \nu = \sqrt{\delta \rho / 2|m^2| (v_b^2 + v_z^2)}$ of a Doppler broadened spectrum for a collinear ($\parallel$) and a counter-propagating ($\perp$) excitation scheme are shown, which has to be convoluted with the $T = 0 \text{ K}$ spectrum. Here, $h\nu_b$ and $h\nu_v$ are the energy spacing of the lower and the upper transitions, respectively.

3.2. Experimental setup for frozen Rydberg gases

3.2.1. Laser system. As already mentioned above, we excite Rydberg states with principal quantum numbers ranging from $n = 20$ up to the ionization threshold by a two-photon excitation scheme. The lower transition from the $5S$ ground state to an intermediate $5P$ state is accomplished with a wavelength at 780 nm and the second step to the desired Rydberg state with roughly 480 nm. Light at 780 and 960 nm is produced by the standard extended cavity diode laser setups in the master–slave configuration. The slave lasers are seeded via acousto-optical modulators (AOM) in the double-pass configuration that allows one to tune the light frequency over a range of 60–320 MHz, respectively, within a few microseconds. The infrared light is further amplified using a tapered amplifier and then frequency doubled (TA-SHG 110, Toptica Photonics AG, Germany) to generate the needed light at 480 nm.

Note that the experimental results on strongly interacting Rydberg gases \cite{19, 25, 91–93} have been accomplished with a precursor to our current setup based on a transfer cavity with a total linewidth of $\lesssim 1 \text{ MHz}$. With the new setup shown in figure 5, we were able to reduce the linewidth of the combined two-step excitation scheme to 60 kHz, which is an improvement of more than one order of magnitude compared to the old setup. The key elements of the new setup are two passively stable reference cavities \cite{94} on which we lock both lasers, respectively. Therefore, we use a Pound–Drever–Hall scheme \cite{95} with a fast feedback (bandwidth $\approx 10 \text{ MHz}$) on the current of the laser diode and a slow feedback on the grating of the extended cavity. To maintain a preferably constant length of the plano-concave cavities, the mirrors with a reflectivity of 0.995 were mounted on one single Zerodur spacer with a length of 100 nm, which corresponds to a free spectral range of 1.50 GHz and a finesse of 626. To avoid thermal drifts, we mounted the cavity inside a high vacuum chamber, vibrationally isolated by nitrile o-rings, at a pressure better than $10^{-9} \text{ mbar}$. Finally, the whole setup is set into an actively temperature stabilized case. One concave mirror is additionally equipped with piezos to tune the length of the resonator. This setup is suitable to hit all the desired wavelengths for arbitrary Rydberg states, but may be bypassed by the usage of a more broadband AOM-based setup, the sidebands of widely tunable EO modulators, or by locking to higher TE modes of the cavities. The latter comes...
Figure 5. Laser system for the two-photon excitation of $^8$Rb into Rydberg states. The red light at 780 nm is generated by a standard external cavity diode laser system. The production of blue light in the range of 475–483 nm makes use of a master–slave setup, where a standard diode laser setup delivers infrared light at 960 nm, which is amplified by a tapered amplifier. A subsequent frequency-doubling cavity delivers the desired wavelength. Both lasers are stabilized (780 and 960 nm) to a passively stable resonator.

Figure 6. The stability of the laser system is derived from the passive length stability of a Zerodur-glass spacer of a Fabry–Perot interferometer (FPI). To avoid thermal drifts, the whole FPI is placed inside a UHV chamber, which is again placed in a constant-temperature environment. The length of the plane-concave resonator is 100 mm and the finesse has been measured to $F = 626$. The spacer contains in total four FPIs, where two of them are additionally equipped with piezo rings for fine tuning of the length. To minimize thermal drifts, the piezo rings are mounted in a self-compensating way.

along with the problem that each TE mode exhibits a slightly different lineshape requiring adapted parameters for the PID locking circuit. By the special arrangement of the piezos, which exhibit a 100 times larger temperature coefficient than Zerodur, shown in figure 6, one can compensate reasonably well for thermal drifts. An even more advanced setup would involve ultrawide expansion (ULE) glasses as a spacer to reduce the thermal drifts even further. Additionally, we use very stable high-voltage supplies (T1DP 005 106 EPU, iseg Spezialelektronik GmbH, Germany) with a voltage stability below $10^{-4}$. Since the piezos only approach their equilibrium length exponentially after a sudden change of the applied voltage, we have to wait for a few hours to be within the stability range of our locking scheme. The combination of all these elements result in a frequency stability of 100 kHz mK$^{-1}$, which can be further improved by the usage of ULE glass and leaving out the piezo rings.

During the experiment, we can tune the frequency by the double-pass AOMs between the master and the slave lasers, which results in an overall scan range of 640 MHz of the blue light and 60 MHz of red light.

The short-time stability of the laser system has been studied by measuring the linewidth $\Delta f$ of the light emitted by the red and infrared slave lasers, respectively, using a delayed self-heterodyne interferometer (DSHI) [96]. The setup is depicted in the inset of figure 7. In one path, the laser light is shifted in frequency using an AOM at 80 MHz; in the other arm, the light is delayed using a single-mode optical fibre of 10.5 km corresponding to a delay time of $\tau = 52 \mu$s. For coherence times $1/\Delta f$ shorter than the delay time $\tau$, the two beams are completely decorrelated. The delayed light can then be treated as a second independent but otherwise identical laser. The spectrum $S_I(\omega)$ of the beat signal from the DSHI is then given as the cross correlation of the original laser spectrum $S_E$ [97]:

$$S_I(\omega) = \int_{-\infty}^{\infty} S_E(\omega + \omega')S_E(\omega') d\omega'.$$  (8)
Figure 7. Spectral power density of the beat signal from the slave laser at 780 nm with master laser locked (blue) and free running (red), measured with a DSHI as depicted in the inset. The curves are centred around the frequency of the AOM at 80 MHz. The signal of the free running laser shows a Voigt profile with a fitted linewidth of 210 kHz (black line), whereas the curve of the locked laser shows typical features occurring if the laser linewidth is well below the resolution limit of the DSHI (here \( \approx 19 \) kHz).

By assuming a model for the laser spectrum \( S_I \), this allows us to determine the linewidth of the laser quantitatively down to the resolution limit \( 1/\tau = 19 \) kHz, given by the delay time. The timescale of this measurement is also determined by the delay time \( \tau = 52 \mu s \), which is adapted to the typical timescale of our experiments. For linewidths below the resolution limit, the spectrum of the beat signal shows typical features such as a well pronounced peak at the centre and a modulation at the wings whose periodicity depends on the delay time \( \tau \) only. In this range, only qualitative measurements are possible unless the laser shows a pure Lorentzian spectrum [98]. Both diode lasers in our new setup show such features. Considering the frequency doubling and the residual broadening of AOMs used for switching the combined linewidth of our Rydberg excitation laser system is below 60 kHz, which agrees with the observed linewidths of experimental Rydberg spectra.

In the end, the light is brought to the atoms by polarization maintaining fibres and overlapped by dichroic mirrors. At the position of the atoms, the 1/e² radius of the 780 nm light was set to 550 \( \mu m \) and the one at 480 nm to 35 \( \mu m \). The maximum available laser power in the blue light (480 nm) is about 100 mW at the position of the atoms. In most experiments, the power of the 780 nm light is reduced well below one milliwatt to avoid excitation into the 5P<sub>3/2</sub> state. At a detuning of 400 MHz (see figure 11) and a typical laser power of, e.g., 50 \( \mu W \), the spontaneous scattering rate reduces to below 1000 s⁻¹. The effective two-photon Rabi frequency at this setting is \( 2\pi \times 250 \) kHz. Another important aspect is the uniformity of the illumination of the atoms. An atomic cloud at, e.g., 3.4 \( \mu K \) confined in our magnetic trap at an offset field of 0.89 G, as used in [19], has a Gaussian shape with a radial width of \( \sigma_p = 8.6 \mu m \). At these parameters, 85% of the atoms experience at least 80% of the maximum two-photon Rabi frequency.

3.2.2. Main vacuum chamber. Our vacuum chamber for exciting and detecting Rydberg atoms in an ultracold cloud of atoms is inspired by the design of the MIT group described in [99]. The vacuum chamber consists mainly of two parts, where the main chamber is shown in figure 8, which now includes field plates to apply various electric fields and MCPs for detection of charged particles. The effusive oven assembly is not shown, which is operated at high vacuum (10⁻⁷ mbar) and delivers a thermal beam of gaseous rubidium atoms to an increasing field Zeeman slower. The precooled atoms are loaded into a magneto-optical trap in the ultrahigh vacuum part of the setup (<2 \times 10⁻¹¹ mbar). After a short molasses,
we transfer the atoms into a magnetic trapping potential. By forced evaporative cooling, we produce for most experiments described here ultracold samples of several $10^6$ atoms at a few $\mu$K in the $F = 2$, $m_F = 2$ state. This is done to obtain a pure Gaussian density distribution in the harmonic trapping potential, which simplifies the subsequent analysis of our data. Typically, we work at peak densities of up to $10^{14}$ cm$^{-3}$, which is about four orders of magnitude larger than in experiments with laser cooled samples. If desired, we can also extend our evaporative cooling to quantum degeneracy and produce Bose–Einstein condensates with a few $10^5$ atoms. Note that for the excitation of Rydberg states in a Bose–Einstein condensate, one has to include in addition the bimodal density distribution of partially condensed clouds into the analysis as it has been done in [92].

The main chamber depicted in figure 8 has to fulfill several boundary conditions simultaneously. First of all, a good optical access in three dimensions for laser cooling is mandatory. Two further optical axes are added for imaging, which are equipped with larger view-ports (CF 63) to obtain a better optical resolution. The numerical apertures in both imaging axes are 0.17, which yields an optimal resolution of 5.6 $\mu$m for a wavelength of 780 nm. Altogether 11 optical view-ports are available which are suitable for wavelengths ranging from 300 nm to 2.5 $\mu$m. At the same time nearby magnetic coils for magnetic trapping are necessary. We use a cloverleaf style Ioffe–Pritchard trap [99, 100], which is located inside two recessed bucket windows outside the vacuum. The inner spacing of the two coil assemblies is 32 mm. The two pinch coils of the cloverleaf trap produce an axial curvature of $B^\prime = 0.56$ G cm$^{-1}$ per ampere and leaves a radial gradient of $B^\prime = 0.61$ G cm$^{-1}$ per ampere. At typical operation conditions with 400 A in all coils and an offset field of 1.5 G, we obtain for $^{87}$Rb trapped in the $F = 2$, $m_F = 2$ state trapping frequencies of 18 Hz axially and 250 Hz radially.

The main goal of the setup described here is the investigation of Rydberg atoms excited from ultracold gases or Bose–Einstein condensates. For the manipulation and detection of the highly excited atoms, we included eight field plates and two MCPs close to the atoms inside the vacuum. The details of this add-on will be discussed in detail below. Because of the arrangement of the field plates, we had to relocate the radio-frequency coils used for evaporative cooling further away from the atoms as can be seen in figures 8 and 9.

We use two coils made of polyimide-coated copper wires each consisting of two loops. Remanent charge on the insulating coating can cause disturbing electric fields if they are too close to the atoms, which is also avoided by the larger distance of the coils. Nevertheless, the coils produce at the position of the atoms an average magnetic field of 5 mG (for frequencies from 1 to 30 MHz), when driven with 2 W at 50 $\Omega$ without impedance matching. This is sufficient to drive the magnetic dipole transitions for evaporative cooling.

A very useful addition to the setup is a helical antenna with a forward gain of 17.9 dBi operated from outside the vacuum chamber and optimized for 6.8 GHz, which corresponds to the hyperfine splitting of $^{87}$Rb. By applying 1 W of RF power to this antenna, we are able to monitor Rabi flopping in the GHz regime. We use this antenna to adjust the density of the $F = 2$, $m_F = 2$ atoms by transferring a certain fraction by a Landau–Zener sweep to the magnetically untrapped $F = 1$, $m_F = 1$ state. During this process, we do not observe a severe change of the temperature, which implies also an invariant density distribution. By this, we avoid the influence of all size effects during Rydberg excitation.

### 3.2.3. Electric-field manipulation and detection of Rydberg atoms

The high sensitivity of Rydberg atoms to electric fields opens the possibility of manipulating the internal states of the Rydberg atoms by field plates [101]. To produce electric-field configurations as versatile as possible, we installed eight field plates close to the atoms. The spatial arrangement can be seen in figures 8 and 9. Each of these plates can be addressed individually, which allows us to generate various field configuration. For example, for a constant electric field of 1 V cm$^{-1}$ pointing along the $z$-axis, one has to charge plates A–D to $-2.6$ V and plates E–H to $+2.6$ V cm$^{-1}$, which produces in addition a curvature below 0.5 V cm$^{-2}$ in all directions. This corresponds to an additional electric field of 50 mV cm$^{-1}$ at a displacement of 1 mm with respect to the geometrical centre. Actually, we observe such an offset field around this order of magnitude [20], which can be explained by a spatial mismatch of the magnetic trapping potential with respect to the electric field plates. In principle, it is possible to compensate for such additional fields, but it requires an elaborate calibration procedure in all three dimensions, since the direction of the perturbing field is most likely unknown.

The eight field plates are made up of stainless steel with a thickness of 0.5 mm. They are glued (Epotek 377, Epoxy Technology) with 1 mm thick ceramic spacers onto the recessed buckets. The dimensions of the spacers have to be small enough that they are completely hidden behind the field plates from the viewpoint of the atoms. Any insulating surface can accumulate charge and falsify the desired field configuration. To charge the field plates, they
are spot welded to stainless steel wires, which are radially led outwards as can be seen in figure 8. At the edge of the recessed bucket, the wires are fixed in position each by a short ceramic tubing, which is also glued to the buckets and are subsequently connected to kapton insulated copper wires. These copper wires are then finally connected to one of the fourfold high-voltage feedthroughs. To avert breakthroughs inside the chamber induced by sharp edges, we rounded off all four edges of each plate with a radius of 1.5 mm. Finally, we etched and electro-polished all field plates including the spot-welded wires to burnish also small spikes. The polishing was done in an acid bath consisting of one part of 96% sulfuric acid, two parts of 85% phosphoric acid and six parts of distilled water. After 2 min at a current of 5 A about 70 μm of stainless steel from the plates was removed and they exhibited a semi-gloss surface. After installation of the field plates and evacuating the chamber, we measured no current leakage up to 3000 V for all plates.

During the experimental process, it is necessary to switch the applied voltages within short times. To do so, we use bipolar high-voltage switches (HTS-6103 GSM, Behlke Electronic GmbH, Germany), which have an intrinsic rise time of 60 ns. The push–pull circuit of the switch has to be adjusted to match the capacitive load of 50 pF of each field plate as well as the 300 pF load of the high-voltage coaxial cable, which connects the switch to the high-voltage feedthroughs.

For a high-detection sensitivity of Rydberg atoms, we installed two MCPs (Type BO12VA, El-Mul Technologies Ltd, Israel) inside the vacuum chamber. After field ionization of the Rydberg atoms, we use one MCP to detect the ions. The second MCP is designated to detect simultaneously the electrons. To improve the amplification even further, we use MCPs in a Chevron configuration, which consist of two successive glass plates with a small spacing in between. The electron current arriving at the anode is converted by a large resistor to a voltage signal, which limits our minimum sensitivity to about 100 ions. In principle, one could distinguish between single-ion events, but the noise level of the signal limits our minimum sensitivity to about 100 ions. Since we shoot around 10^12 ions per year into the detector, we do observe aging effects of the MCP, which makes a re-calibration from time to time necessary. Due to the large distance of the MCP to the position of the atom cloud, we can easily distinguish between Rb^+ and Rb^+_2 ions by their different time of flights due to their differing masses [102].

3.3. Excitation of Rydberg states in a magnetic trap

Due to the confinement of the cold atomic cloud in a magnetic trap, Rydberg atoms are excited in an offset magnetic field. The experimental sequence for Rydberg excitation in a magnetic trap is shown in figure 10. The ultracold cloud consists typically of 4 × 10^6 atoms at a temperature of 3 μK confined in a cigar-shaped harmonic trapping potential with an axial trapping frequency (along the z-axis) of 18 Hz and a radial trapping frequency of 310 Hz. The magnetic offset field at the centre of the trapping potential was set to 1.0 G during the cooling and trapping sequence. The cloud is spin polarized in the F = 2, m_F = 2 ground state with respect to the quantization axis given by the magnetic field after reloading from the MOT to the magnetic trap. Since evaporative cooling is performed via RF coupling of neighbouring m_F states, a residual occupation of the weakly bound F = 2, m_F = 1 state is created in the cooling sequence.

For highly excited states, the hyperfine coupling is significantly decreased and we remain with the coupling of the electron spin and the orbital momentum to a total momentum

Figure 11. The level scheme of the relevant levels $5S_{1/2}$, $5P_{1/2}$ and $43S_{1/2}$. (Left) A schematic of the two-photon transition including the frequency splittings (in MHz unless otherwise noted) between the hyperfine states (nuclear spin $I = 3/2$). (Right) The magnetic sublevels together with their respective Zeeman shift in $f$-basis for $5S_{1/2}$, $5P_{1/2}$ and in $j$-basis for $43S_{1/2}$.

Atoms are excited to the Rydberg state via a two-photon excitation, detuned by about 400 MHz from the intermediate $5P_{1/2}$ state (red and blue solid arrows in figure 11). The $nS_{1/2}$ Rydberg states exhibit the same magnetic moment as the ground state. Therefore, the transition from the ground state to this state is insensitive to magnetic fields. Polarizations of both excitation lasers are chosen as indicated in figure 11 to suppress excitation via different paths (grey dashed lines) to other magnetic substates. However, the direction of the magnetic field is spatially inhomogeneous due to the geometry of the magnetic trap. This causes spatially dependent excitations into the other $m_J$ Rydberg states by admixtures of other polarizations.

Atoms in the $F = 2$, $m_F = 1$ ground state can also be excited to the same Rydberg states. Due to the Zeeman splitting of the ground state, these transitions show different dependences on the magnetic field. To determine the relevant transitions and explore the dependence on the magnetic offset field, Rydberg spectra at different magnetic field strength are taken as shown in figure 12). The magnetic offset field was ramped up shortly before the excitation and detection sequence. The spectra show four distinct resonances of which three are dependent on the magnetic field (solid lines). The resonance at $-5.5$ MHz marked by the dashed line is due to ultralong-range Rydberg molecules.

4. Strongly interacting Rydberg gases

The strength of interactions in a system is usually characterized by the dimensionless quantity $na^3$, with $n$ being the density and $a$ an effective range of the interaction potential. Strongly interacting systems with $na^3 \gg 1$ are often difficult to describe theoretically. This can be understood as strong interactions typically lead to strong quantum correlations, where an effective description in terms of non-interacting quasiparticle excitations is no longer valid. The first steps to understanding the essential properties of strongly interacting systems are usually based on mean-field theory; however, especially in
low-dimensional systems, this can lead to unreliable results. Nevertheless, it is possible to derive a mean-field approach to strongly interacting Rydberg gases that reproduces the correct critical exponents for the underlying quantum phase transition if the dipole blockade is explicitly taken into account [22, 25]. In the case of interacting Rydberg gases, the effective range of the van der Waals interaction between Rydberg states is on the order of $a \approx 5 \, \mu m$, which has to be compared with the interparticle distance between the atoms in a Bose–Einstein condensate, $1/√n \approx 100 nm$, leading to $n a^3 = 1.25 \times 10^8$, which is deep in the strongly interacting regime.

4.1. Phenomenological description of strongly interacting Rydberg gases

The interaction energy per particle in a gas of highly excited Rydberg atoms with density $n$ is given in the case of a van der Waals interaction by $-C_6 n^2$. For two rubidium atoms in the 43S state, the van der Waals coefficient is $C_6 = -1.697 \times 10^{19} \, \text{au} = -h \times 2441 \, \text{MHz} \, \mu m^6$. If we now take a typical density of $n = 10^{12} \, \text{cm}^{-3}$ (mean distance $1/√n = 1 \, \mu m$) in an ultracold atomic cloud and assume that all atoms reside in a Rydberg state, then the average interaction energy reaches $h \times 2441 \, \text{MHz}$ per particle. This energy has to be compared to the other energy scales of the system like the temperature in the $\mu K$ regime, which corresponds to a kinetic energy of a few kHz, the trapping frequencies of few Hz to kHz and potentially to the chemical potential of a Bose–Einstein condensate of a few $\text{kHz}$. All these energies are comparable with the spectral width of the excited Rydberg states given by their lifetimes. The crucial parameter for coherent excitation dynamics is a sufficiently strong coupling strength of the exciting laser fields $\Omega_R$. The energy $h\Omega_R$ determines now in combination with the interaction energy $-C_6 n^2$ all bulk properties of the driven system as, e.g., the steady-state maximum Rydberg density. By equating these two energies $-C_6 n^2 = h\Omega_R$, e.g., for the 43S state and a driving field of $\Omega_R = 1 \, MHz$, one can estimate a minimum distance between two Rydberg atoms of $3.67 \, \mu m (n_b = 2 \times 10^{10} \, \text{cm}^{-3})$, a distance severely larger than the distance between two ground-state atoms. In other words, the excitation to Rydberg states is strongly blockaded and only one atom out of $n_b/n \approx 1$ atom can be excited to a Rydberg state.

This line of arguing is equivalent to the picture shown in figure 13, where the power broadening $\Omega_R$ of the exciting laser field can compensate the van der Waals repulsion up to a minimum distance.

Figure 13 gives a good illustration of the blockade mechanism during excitation of two atoms but is only correct approximately, since it neglects the many body aspect of the excited state as described in figure 14. Within the blockade region, one excitation is distributed over all atoms $N_b$, which results in a collective state $|W\rangle = |\psi_e\rangle = 1/\sqrt{N_b} \sum_{N_0} |g_{1}, g_{2}, g_{3}, \ldots, e_{1}, \ldots, g_{N_b}\rangle$, where we have dropped a spatially dependent phase factor $\exp(-ikr)$ of the excitation light assuming a plane wave. The effective coupling $\Omega$ to the collective excited W-state shows now a $\sqrt{N_b}$ enhancement due to the collective dipole matrix element $\langle g|ed|W\rangle$ with the ground state $|g\rangle = |g_{1}, g_{2}, g_{3}, \ldots, g_{N_b}\rangle$. This W-state is actually the only bright state that couples to the light field, whereas the other $N_b - 1$ states are dark states as can be easily seen for $N_b = 2, 3$ and 4:

- $N_b = 2$
  - bright: $1/\sqrt{2} (|rg\rangle + |gr\rangle)$
  - dark: $1/\sqrt{2} (|rg\rangle - |gr\rangle)$

Figure 14. Resonant excitation of an ensemble of two-level atoms. When exciting an ensemble of $N$ non-interacting atoms with a coupling strength $\Omega$, the atoms undergo coherent Rabi oscillations with the frequency $\Omega$. In the case of interactions and an ensemble size smaller than the blockade radius, only one excitation is possible and the excited collective state reads $W = 1/\sqrt{N} \sum |g_{1}, g_{2}, g_{3}, \ldots, e_{1}, \ldots, g_{N_b}\rangle$. As a consequence, collective Rabi oscillations arise with an $\sqrt{N}$ faster oscillation and by a factor $N$ reduced amplitude.
As one can easily see, in contrast to the bright W-state, the density of Rydberg states is not unique. Due to the interaction energy per Rydberg atom in such a lattice, the Rydberg atoms are arranged in a hexagonal closed packing. The interaction energy per Rydberg atom in this lattice is now
\[ Z = \frac{1}{6} \left( +|r_{ggg}| + |g_{rrg}| + |g_{rgg}| \right) \]
\[ + \frac{1}{2} \left( +|r_{ggg}| - |g_{rrg}| + 2|g_{rgg}| \right) \]
\[ + \frac{1}{2} \left( |r_{ggg}| - |g_{rrg}| + 0|g_{rgg}| \right) \]

\[ N_B = 3 \]

\[ N_B = 4 \]

The physical situation according to the experimental approach is depicted in figure 15, where the inhomogeneous density distribution have to be treated as individual atoms.

We start with an inhomogeneous atomic cloud with all atoms in the ground state and then switch suddenly the coupling light field to the Rydberg state. Then, the evolution of the excited state fraction \( f_R \), which is the total number of excited Rydberg atoms divided by the number of ground-state atoms \( N = \int n(r) \, dV \), follows:

\[ f_R(t) = \int \left( \frac{n_r(r)}{n(r)} \right) \sin \left( \sqrt{\frac{n_r(r)}{n(r)}} \, \Omega_R(r) t/2 \right) \, dV, \]

where we have used (11) and (12). The integral can be understood as a local density approximation, which is valid as long the variation in density is small compared to the blockade radius. The collective description given by the integral is only correct at sufficient high densities, where the excitation blockade is effective. In the low-density regime, where the interatomic distances are larger than the blockade radius, one has to include the single-particle dynamics separately. Note that in this picture of noninteracting superatoms, a homogeneous sample \( (n = \text{const}) \) \( f_R(t) \) would oscillate with the collective Rabi frequency. This is unlikely to happen for a thermal ensemble of atoms due to the random spatial normal distribution of atoms leading to a band structure of the excited states. An ordered array of atoms as e.g. given by a Mott-insulator state will exhibit a less complex distribution of the excited states with a few discrete energies and coherent Rabi oscillations may be observable. The temporal evolution of \( f_R(t) \) for a Gaussian density distribution results in a superposition of Rabi oscillations with varying collective frequencies and resembles a simple saturation behaviour with an initial linear increase with a rate \( R \) and an exponentially approaching saturation value \( f_{R,\text{sat}} \) as

\[ f_R(t) = f_{R,\text{sat}}(1 - e^{R/t_{\text{sat}}}). \]

This temporal evolution agrees well with an experimental observations and also with the corresponding numerical simulations. For very short times \( t < \Omega^{-1} \), the interaction energy lies below the saturation broadening and the system evolves quadratically with \( f_R(t) \sim \Omega_R^2 t^2 \). In the case of a
Gaussian distribution, one can extract scaling laws for $R$ and $f_{R,\text{sat}}$ with the help of equations (11) and (12) as a function of the peak density $n_0$ and the single-atom Rabi frequency $\Omega_R$:
\[
R \sim n_0^{1/5} \Omega_R^{2/5},
\]
\[
f_{R,\text{sat}} \sim n_0^{-4/5} \Omega_R^{2/5}.
\]
Keeping these results in mind, one notes that some care has to be taken when investigating the strongly interacting limit given by $n a^3 \gg 1$. It might be tempting to associate $n$ with the Rydberg density $n_r$, leading to the relation $n_r a^3 \sim 1$. Such a reasoning, however, ignores the fact that the system is driven between the ground state and the Rydberg state. We will show in the subsequent theoretical analysis that the global energy scale dominating the dynamics is $C_0 n^2$, with $n$ being the ground state density. Treating the driven system as a gas of Rydberg atoms and ignoring the ground state atoms misses the important fact that the number of Rydberg excitations is not a conserved quantity. This is in strong contrast to experimental situations in which an atomic gas is brought into the strongly interacting limit by means of a Feshbach resonance; there, the number of atoms is indeed conserved. Nevertheless, there do exist several proposals, which try to convert some part of these extreme interaction energies into effective interaction potential for ultracold atoms by admixing a small fraction of Rydberg states to the ground state [47–49, 104]. To get an idea of the actual length scales of the blockade radius, in figure 16 we depict its dependence for different values of the coupling field $\Omega$, the ground state density and the principal quantum number.

4.2. Mean-field theory of strongly interacting rydberg gases

The dynamics on the timescale of the experiment is well described by the Hamiltonian in the rotating frame of the laser excitation with the Rabi frequency $\Omega$ and the detuning $\Delta$:
\[
H = -\frac{\hbar}{2} \sum_i \sigma_i^{(i)} + \frac{\hbar \Omega_R}{2} \sum_i \sigma_i^{(i)} - C_p \sum_{j<i} \frac{P^{(i)}_{ee}(p)}{|r_i - r_j|^3},
\]
where the counter-rotating terms have been dropped [105]. It is instructive to first look at the Hamiltonian in the classical limit, i.e. $\Omega_R = 0$. For negative detuning $\Delta$, the ground state is a paramagnet with all spins pointing down since adding a Rydberg excitation will cost energy. However, for positive $\Delta$, the many-body ground state will have some Rydberg excitations present. For a fixed fraction of Rydberg excitations $f_R$ in the system, the energy will be minimal when the interaction energy is minimized, which is the case in a crystalline arrangement. At $\Delta = 0$, there is a continuous phase transition from the paramagnetic to the crystalline phase [22]. In the strongly interacting limit, it is convenient to use the interaction energy $E_0 = C_p |p|^{(i)}$ as the global energy scale and express the Rabi frequency and the detuning as dimensionless quantities:
\[
\alpha = \frac{\hbar \Omega_R}{|C_p| n^{(i)}} \quad \text{and} \quad \eta = \frac{\hbar \Delta}{|C_p| n^{(i)}}.
\]

Figure 16. (a) The dependence of the blockade radius for the 43S state according to equation (11) for a single superatom ($Z = 1$) as a function of the ground-state atom density $n_j$ and for various coupling strengths $\Omega_R$ (red: 100 Hz, green: 10 kHz, blue: 1 MHz, orange: 100 MHz; top to bottom). (b) The blockade radius is plotted for S-states as a function of the principal quantum number for a fixed coupling strength of $\Omega_R = 10$ kHz and various densities $n_j$ (red: $10^{-4}$ m$^{-3}$, green: $10^4$ m$^{-3}$, blue: $10^6$ m$^{-3}$, orange: $10^8$ m$^{-3}$; top to bottom). The dashed line indicates in both figures the region, where the interparticle distance $1/\sqrt{2}$ is larger than the blockade radius $r_b$, which is emphasized by the black line in (a).

The most straightforward method to deal with a system close to a continuous phase transition is mean-field theory. However, we cannot simply apply the conventional Landau theory, i.e. performing a Taylor expansion of the energy in the order parameter, as there are no correlations in Landau theory, which is incompatible with the dipole blockade. Instead, we make an explicit ansatz for the pair-correlation function
\[
g_2(r_i - r_j) = \frac{\langle P^{(i)}_{ee}(p) P^{(j)}_{ee}(p) \rangle}{f_R^2}.
\]
In the blockaded region with the distance $|r|$ being much smaller than the average Rydberg spacing $a_R$, the pair-correlation function vanishes. On the other hand, at large distances $|r| \gg a_R$, the correlation disappears and consequently $g_2(r) = 1$. The transition from a strong suppression to the uncorrelated regime is very sharp and the pair correlation function can be modelled by a step function
\[
g_2(r) = \theta (|r| - a_R).
\]
Then, the mean field theory containing this pair-correlation function is obtained by replacing the microscopic interaction in the Hamiltonian (17) by the mean interaction of the neighbouring atoms:

\[ p_{ee}^{(i)} p_{ee}^{(j)} \approx [p_{ee}^{(i)} f_R + p_{ee}^{(j)} f_R - f_R^2]g_2(r_i - r_j). \]  

(21)

which reduces the Hamiltonian to a sum of local Hamiltonians \( \mathcal{H}_m^{(i)} \) at each site \( i \). In the strongly interacting regime with \( \alpha, \eta \ll 1 \), the number of atoms in the blockaded regime is large, i.e. \( d^2 n \gg 1 \). This allows us to replace the summation over the surrounding atoms \( j \) by an integral over space with a homogeneous atomic density \( n \):

\[
\sum_j g_2(r_i - r_j) \frac{-C_p}{|r_i - r_j|^p} = A_d n \int_0^\infty d^d r r^{d-1} g_2(r) \frac{|C_p|}{r^p} \\
= \frac{A_d}{p - d} \int d^d r |C_p| r^{d-p},
\]

(22)

where \( A_d \) is the surface area of the \( d \)-dimensional unit sphere.

Then, we obtain the Hamiltonian for the \( i \)th atom:

\[
\frac{\mathcal{H}_m^{(i)}}{E_0} = \frac{\eta \sigma_z^{(i)} + \alpha \sigma_z^{(i)}}{2} + \frac{A_d}{p - d} \left( -\eta \sigma_r^{(i)} + f^2 \right) \\
= \frac{A_d}{2(p - d)} \left( -\eta \sigma_r^{(i)} + f^2 \right) = \mathbf{h} \cdot \mathbf{\sigma}^{(i)} + h_0.
\]

(23)

In the limit of strong blockade, the correlation length \( \xi \) is equal to \( a_R \) as this will minimize the interaction energy for a given Rydberg fraction. The correlation function \( g_2(r) \) thus satisfies the normalization condition

\[ n f_R \int dr [1 - g_2(r)] = 1, \]

(24)

which provides the relation

\[ a_R = (V_d f_R n)^{-1/d}, \]

(25)

with \( V_d \) being the volume of the \( d \)-dimensional unit sphere. The effective Hamiltonian is equivalent to a spin in a magnetic field \( \mathbf{h} \), which possesses both a transverse component \( h_t \) and a longitudinal component \( h_z \), and a constant energy offset \( h_0 \). We can diagonalize the Hamiltonian using a spin rotation, i.e. \( \mathcal{H}_m^{(i)} = \mathbf{h} \sigma_z^{(i)} + h_0 \), where \( h = \sqrt{h_t^2 + h_z^2} \). Here, \( \sigma_z^{(i)} = \cos \theta \sigma_x^{(i)} + \sin \theta \sigma_y^{(i)} \) denotes the Pauli matrix in the new basis, while the rotation angle can be expressed in terms of the magnetic field components as \( \tan \theta = h_z/h_t \). The ground state of the system is then given by \( \langle \sigma_z \rangle = -1 \), leading to the self-consistent equation

\[ \alpha = \frac{1}{p - d} |4A_d V_d^{d-1} f^{p/d+1/2} - p \eta f^{1/2}|. \]

(26)

This equation can be cast into the form

\[ \alpha \sim f_R^d |B - \eta f_R^{1/b}|, \]

(27)

from which the critical exponents \( d = p/d + 1/2 \) and \( b = d/p \) can be determined. Solving the mean-field equation for \( f_R \) leads to the equation of state shown in figure 17, with the limiting cases

\[ f_R \sim \eta^{d/p} \quad \text{for} \quad \alpha = 0, \]

(28)

Figure 17. Excited state fraction \( f_R \) of a strongly interacting Rydberg gas according to mean-field theory \((d = 3, p = 6)\).

\[ f_R \sim \alpha^{2d/(2p+d)} \quad \text{for} \quad \eta = 0. \]  

(29)

These critical exponents can also be derived using a universal scaling function [25].

So far, the mean-field analysis has been centred onto the ground-state properties of the system. However, in typical experiments, the Rabi frequency is not varied adiabatically such that the system follows its many-body ground state; instead, the driving lasers are switched to full power instantaneously, leading to a complex relaxation dynamics towards a stationary state. The resulting critical exponents are identical to those for the ground state. The phenomenological superatom model can then be derived by calculating the energy gap \( h \), which for \( \eta = 0 \) scales like \( h \sim \sqrt{N_b} \), with \( N_b \) being the number of blockaded atoms.

4.3. Numerical simulations of strongly interacting Rydberg gases

To get a quantitative assessment of the validity of mean-field results, it is instructive to obtain exact numerical results on the basis of the full spin Hamiltonian (17). For a system with \( N \) atoms, the dimension of the full Hilbert space grows exponentially like \( 2^N \); therefore, the exact behaviour can only be calculated for a relatively small number of atoms. However, the strong van der Waals repulsion suppresses the occupation probabilities of many basis states, which allows us to significantly reduce the Hilbert space: for each basis state, we compute the van der Waals energy and remove the state if its van der Waals energy is larger than a cutoff energy \( E_C \). This reduction leads for \( N = 100 \) to approximately \( 10^5 \) relevant basis states compared to the \( 10^{10} \) basis states of the full Hilbert space. Convergence of this method can be checked by increasing \( E_C \).

Within mean-field theory, we have already seen that the critical properties are identical whether we study ground-state properties or the relaxation to a stationary state after a sudden increase of the Rabi frequency. As we are also interested in dynamical properties, we perform a numerical integration of the time-dependent Schrödinger equation with the initial state being the product state of all spins pointing down. We place the atoms randomly according to a uniform distribution.
into a box of volume $V$ having periodic boundary conditions. Then, the resulting dynamics obtained using a fourth-order Runge–Kutta method is shown in Figure 18. One can see the expected relaxation towards a stationary state, although for $N = 50$ atoms there are considerable finite-size effects present in the system, especially concerning the saturation value of the Rydberg fraction $f_R$. However, these can be reduced by averaging over many different initial positions of the atoms according to the same distribution function.

Using our numerical simulation, we can also test the validity of our modelling of the pair-correlation function as a step function. As can be seen from Figure 19, the assumption is qualitatively correct although deviations can be clearly seen. There is a pronounced peak near the correlation length $\xi$ and also the normalization constraint of equation (24) leading to the visibility $v$ for different times $\tau$ and different densities. As predicted by mean-field theory, the visibility only depends on $\alpha^{-2/5}\tau$.

It is now possible to investigate how the scaling of the saturated Rydberg fraction $f_{sat}$ changes when the dimensionless parameter $\alpha$ is varied. For this, the value of $f_{sat}$ needs to be averaged over many random configurations of the atoms. Then, varying $\alpha$ by changing both the density $n$ and the Rabi frequency $\Omega$ leads to a clear data collapse to single line, with the numerically obtained critical exponents being in excellent agreement with mean-field results [22].

### 4.4. Simulations of rotary echo techniques

So far, the theoretical analysis has been centred around the derivation of scaling relations that can be compared to experimental results. For more information about the dynamics of the system, it is instructive to probe its coherence properties. Such an analysis is complicated by the experimental constraints that the only readily accessible quantity is the visibility $v$.

So far, the theoretical analysis has been centred around the derivation of scaling relations that can be compared to experimental results. For more information about the dynamics of the system, it is instructive to probe its coherence properties. Such an analysis is complicated by the experimental constraints that the only readily accessible quantity is the visibility $v$.

Figure 18. Relaxation dynamics of the Rydberg fraction $f_R (p = 6, d = 3, N = 50, \alpha = 1/25, \eta = 0)$.

![Figure 18](image1)

Figure 19. Pair-correlation function $g_2 (r)$ in the stationary state ($p = 6, d = 3, N = 50, \alpha = 1/25, \eta = 0$). The length scales corresponding to the cutoff energy $E_c$ and the mean Rydberg spacing $a_R$ are shown as vertical lines.

![Figure 19](image2)

Figure 20. Numerical results for a rotary echo setup. (a) Time evolution of the Rydberg fraction $f_R$ with (red) and without (blue) changing sign of the Rabi frequency at $\tau_p = 0.5 \Omega^{-1}$. (b) Scaling of the visibility $v$ for different times $\tau$ and different densities. As predicted by mean-field theory, the visibility only depends on $\alpha^{-2/5}\tau$.

![Figure 20](image3)
scaling behaviour of the visibility. As predicted by mean-field theory, there is a data collapse when the visibility is expressed as a function of the intrinsic timescale $\alpha^{-2/5}\tau$.

The concept of the visibility can also be used to determine a (possibly time dependent) dephasing rate $\gamma_d$, defined by the rate equation

$$\frac{d}{d\tau} v(\tau) = -\gamma_d(\tau) v(\tau).$$  \hspace{1cm} (31)

Assuming $\gamma_d(\tau)$ to be a power law in $\tau$, we can directly express the rate up to a constant factor as

$$\gamma_d = -\frac{\log v}{\tau}.$$  \hspace{1cm} (32)

This decay rate can now directly be compared with experimental results as only the Rydberg fraction $f_\text{R}$ has to be measured [93].

5. Experimental results on strongly interacting Rydberg gases

Strongly interacting Rydberg gases are best explored in dense ultracold clouds confined either in magnetic traps or in dipole traps with various methods. But also studies on laser cooled ultracold clouds confined either in magnetic traps or in dipole traps with various methods as described in more detail in the experimental results as only the Rydberg fraction $f_\text{R}$ has to be measured [93].

Figure 21. A strongly interacting Rydberg gas for, e.g., the 33S, 43S and 53S states in rubidium is limited to a certain domain in the parameter space determined by the density $n$ of the ground-state atoms, the coupling strength $\Omega_0$ between the ground-state atom and the Rydberg state, and the coherence time $\Gamma$. The large section of the 43S state, this is only relevant at densities above $10^{25}$ m$^{-3}$. However, for the 43S state, this is only relevant at densities above $10^{25}$ m$^{-3}$, which are hard to prepare in the first place. At reduced densities, the system becomes weakly interacting, whenever the blockade radius $r_0$ is smaller than the distance between two atoms establishing the unblockaded regime. For some cases, there will also be a limit in the regime of very high Rabi frequencies $\Omega_0$ due to multi-photon ionization processes. In the regime of strongly interacting Rydberg gases represented by the white area, several experiments have been conducted with various methods as described in more detail in the text. The grey shaded area represents only the parameter space of all our experiments with Rydberg atoms in the 43S state [19, 91–93].

The grey shaded area in figure 21 embraces the parameter space in which we performed our experiments on the 43S state in the strongly interacting regime. The focus of these experiments has been lying on the excitation blockade effect and on the coherence properties of a strongly interacting Rydberg gas. In our first experiment on strongly interacting Rydberg gases [19], we studied the excitation dynamics into Rydberg states of magnetically trapped clouds close to quantum degeneracy. In this work, as well as in all follow-up experiments, the state of choice was 43S of rubidium, which yields a purely repulsive van der Waals potential. By this, the Rydberg atoms are well protected against inelastic collisions as it happens for attractive D-states [76–80]. Nevertheless, some ions are also produced during the excitation into the 43S state and it is necessary to apply some electric field ($\sim 2$ V cm$^{-1}$) to extract the ions rapidly from the atomic cloud. As a result, we could observe in [19] a coherent evolution into the strongly blockaded regime by means of collective
excitations and explained the scaling behaviour within the phenomenological formalism of strongly interacting Rydberg atoms described above. In this experiment, we used atomic clouds of ground-state atoms with extensions much larger than the blockade radius, which were suddenly exposed to the excitation light field. The evolution of the excitation dynamics did not show any collective Rabi floppings, as suggested in figure 14, but exhibited a saturation behaviour with an initial excitation rate $R$ and a saturation value $f_R$. One reason for the absence of Rabi oscillations is the density distribution of the atomic cloud resulting in a spatial variation of the collective Rabi frequency $\Omega$. As we ionize the Rydberg atoms neither space- nor state-selective, we only monitor the integrated evolution over the entire cloud. Another reason is given by the random distribution of the ground-state atoms leading to broad bands in the density of states for the excited states. The proof of the collective character of the excitation and its saturation due to the strong van der Waals interactions relies on the scaling laws given by equations (15) and (16). The effect on the excitation dynamics with respect to the coupling strength $\Omega_1$ is easily observable by adjusting the corresponding laser intensities. To obtain the dependence with respect to the ground-state atom density, we used a Landau–Zener sweep to remove atoms from the cold clouds without changing the temperature. By this, we keep the effective volume of the atomic cloud constant and are invulnerable to size effects. With access to these two parameters, we were able to prove that we really observe coherent collective excitation into a strongly blockaded system [19].

Since the excitation rate and the steady-state population depend on the ground-state density, it is also possible to monitor the phase transition of a classical thermal cloud to a Bose–Einstein condensate by recording the excitation dynamics as a function of temperature. The main effect arises due to a redistribution of the atoms when crossing the critical temperature $T_c$. Below this point, a major fraction of the thermal cloud is suddenly transferred into the much smaller Bose–Einstein condensate, which leads to a significant variation of the excitation dynamics around $T_c$ [92]. This effect can be described in quite good agreement with the experimental observations by the local density approximation given in equation (13).

Until now, the coherent character of the excitation has only been deduced from the scaling behaviour of the excitation rate as a function of the intensity $n$ and the coupling strength $\Omega_R$ in [19]. By applying more complex pulse sequences as e.g. in the case of a rotary echo, it is possible to study the coherence properties of the excitation dynamics in more detail. The key advantage of the rotary echo is its insensitivity to variations in the coupling strength. In the case of a magnetically trapped cloud of cold atoms, we have to deal with a Gaussian density distribution, which leads according to equation (12) also to a spatial distribution of the collective coupling strength $\Omega$. In our rotary echo sequence, we flip the phase after a certain excitation time $\tau$ by $180^\circ$ resulting in an inversion of the excitation dynamics. At a time of $2\tau$, all atoms have returned coincidental in the ground state. However, the flipping of the phase does not change the sign of the van der Waals interaction among the excited Rydberg states, which still leads to a dephasing during excitation. This method allows now to discriminate between dephasing caused by inhomogeneities and dephasing caused by interactions. The rotary echo method has been applied to the excitation dynamics of 43S states [91] and also for various D states between $n = 40$ to $n = 45$ in the vicinity of a Förster resonance [21]. An alternative spectroscopy method immune to inhomogeneities in the coupling strength is given by Ramsey spectroscopy, which has been recently employed to study the decay mechanisms of highly excited Rydberg molecules [67] as well interaction effects close to a Förster resonance [20, 107].

Before exploiting now even more the highly developed methods and complex pulse sequences of nuclear magnetic resonance spectroscopy, one can also use the well-known electro-magnetically induced transparency as a coherent spectroscopy method to investigate the effect of interactions among Rydberg states. We have employed this method for dense samples excited to the 43S state [93] to analyse dephasing due to interactions, but it has also been shown to be useful to demonstrate interaction-induced optical nonlinearities in atomic samples at much lower densities [18].

A very peculiar effect of strongly interacting Rydberg gases is the blockade radius, which shall produce highly correlated density distributions of the excited Rydberg states. A first direct observation of these correlations has now been achieved by ionizing the Rydberg atoms after excitation into the strongly blockaded regime and a subsequent blow up imaging of the resulting ions by a fluorescence detector [108].

Various aspects of the interaction mechanisms among Rydberg atoms, especially Förster resonances, have already been studied by several groups with optically trapped atoms [109–111] and laser cooled atoms at somewhat smaller densities [10, 11, 16, 71, 107, 112–117]. The added value of these experiments demonstrate the strength of off-resonant and resonant interactions in various approaches for many different tasks in atom physics and quantum optics and will soon go beyond the quest for a fundamental understanding of the underlying physics.

6. Conclusion

This tutorial reviews only a small fraction of modern Rydberg physics and other review articles [9, 118] should be consulted to obtain a more complete picture. The research field dedicated to interacting Rydberg atoms is rapidly growing and splits into self-contained research fields. Much effort is made to realize scalable quantum computers [119, 120] and quantum simulators [39, 40] based on individually trapped single atoms, which might also be expanded to Rydberg states of ions [121]. Actually, ensembles of strongly interacting Rydberg may also be useful as a source of cold ions since the random potential heating is reduced by the blockade mechanism [27]. Although there have been quite a few studies on the nature of Förster resonances, there happens to be a lack of experiments actually using these resonances to address a specific physical problem. Possible fields of applications are quantum computing and quantum simulation. A natural
research topic will be the study of energy transport in a quantum network [122] as it happens also in many biological systems [123].

The nonlinearity induced by the interactions does not only produce interesting new states of material quantum matter but will also show accordingly a backaction on the light field. This has been observed already for laser-cooled clouds with EIT spectroscopy [18, 124] in terms of optical nonlinearities. There exist various proposals to use the blockade effect as an optical nonlinearity on the single-photon level to generate non-classical states of light fields as, e.g., Fock states [26, 52, 125, 126] and cat states [51].

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