Abstract. We present a detailed analysis of our recent observation of synchronous Rabi oscillations between the electronic ground state and Rydberg states in a mesoscopic ensemble containing roughly 100 ultracold atoms (Reetz-Lamour et al submitted, Preprint 0711.4321). The mesoscopic cloud is selected out of a sample of laser-cooled Rb atoms by optical pumping. The atoms are coupled to a Rydberg state with principal quantum number around 30 by a two-photon scheme employing flat-top laser beams. The influence of residual spatial intensity fluctuations as well as sources of decoherence such as redistribution to other states, radiative lifetime and laser bandwidth are analysed. The results open up new possibilities for the investigation of coherent many-body phenomena in dipolar Rydberg gases. As an example we demonstrate the van der Waals blockade, a variant of the dipole blockade, for a mesoscopic atom sample.
1. Introduction: on aims and obstacles

Since the early days of atomic physics, atoms in highly excited electronic states (‘Rydberg atoms’) have been extensively studied due to their exaggerated properties marking the borderline between classical and quantum physics [1]. Recently, Rydberg atoms have attracted interest as a possible implementation of quantum information processing based on neutral atoms [2]–[5]. Representing an alternative to trapped ions or nuclear spins in molecular complexes [6, 7], neutral atoms offer extraordinarily weak coupling to dissipative processes of the environment, thus promising long coherence times [8]. Several schemes have been proposed to entangle neutral atoms and to realize quantum gates [3, 9, 10]. Ensembles of ultracold Rydberg atoms appear to be particularly promising as they exhibit controllable electric dipole interactions over distances of many micrometres. The coherent character of these dipolar Rydberg–Rydberg interactions has been explored in ultracold gases over the last few years [11]–[15], paving the way for the implementation of fast two-qubit gates.

One prominent example for the strong Rydberg–Rydberg interaction is the so-called van der Waals blockade. The van der Waals blockade is the inhibition of multiple excitations in a mesoscopic ensemble due to the interaction-induced energy shifts which separate the single excitation from multiple excitations energetically. It is a favourable tool in quantum information protocols [3] and has resulted in a number of experimental investigations. It was first observed as a suppression of excitation in macroscopic clouds for strong van der Waals interaction at high principal quantum numbers [16, 17], later for both resonant and permanent dipoles [18, 19]. It is accompanied by a change in counting statistics [20] and a modified many-body Rabi frequency which has not been observed directly so far but evidence of which has recently been found [21].

Despite this success, the realization of single-qubit operations with Rydberg atoms has proven to be particularly demanding. While stimulated adiabatic passage has been used to achieve high Rydberg excitation probabilities [22, 23], Rabi oscillations of atoms starting from...
the ground state are hard to observe due to the small spatial overlap between the electronic wavefunctions of ground and highly excited states resulting in very small dipole transition matrix elements. In order to achieve sufficiently high Rabi frequencies despite the small dipole matrix elements between the electronic ground state and Rydberg states, one therefore needs high laser intensities provided by tightly focused beams. The Gaussian beam profile of a focused laser beam, however, exhibits a wide range of intensities and would therefore impede the observation of synchronous Rabi floppings in a mesoscopic sample [23]. One may overcome this by preparing the atomic cloud in very small dipole traps, rendering the atomic cloud smaller than the excitation laser and thus only using the central part of a focused Gaussian beam. This technique has recently been used for clouds of between 1 and 10 atoms [24].

We have recently demonstrated Rabi oscillations between ground and Rydberg states in a frozen mesoscopic sample of roughly 100 ultracold atoms by applying beam shaping to ensure a constant intensity distribution over the excitation volume [25]. We restrict the excitation volume to the area where the shaped beam has a sufficiently constant intensity by spatially selective optical pumping. In this way, we filter a signal of a subensemble of only 100 atoms out of a cloud of 10 million atoms. Here, we discuss the experimental details and thoroughly investigate important sources of decoherence. In addition, we demonstrate the van der Waals blockade for this mesoscopic system by observing a density-dependent suppression of excitation for strong van der Waals interaction at large principal quantum numbers $n$.

2. Experimental realization

The experiment is performed with a cloud of ultracold atoms in a vapour-cell magneto-optical trap (MOT) with about $10^7$ $^{87}$Rb atoms at densities of $10^{10}$ cm$^{-3}$ and a temperature below 100 $\mu$K. The excitation is achieved with two counter-propagating laser beams at 780 and 480 nm via

$$5S_{1/2}^{780 \text{ nm}} \rightarrow 5P_{3/2}^{480 \text{ nm}} \rightarrow nS_{1/2} / nD_{5/2},$$

(see figures 1 and 2). By detuning from the intermediate state $5P_{3/2}$, this state experiences negligible population which assures sufficiently long coherence times [23]. The excitation laser at 780 nm is collimated to a waist of 1.1 mm ensuring a constant Rabi frequency over the excitation volume. The Rabi frequency is determined from measured Autler–Townes splittings [23, 26, 27] with typical values of $\omega/2\pi = 55$ MHz.

The 480 nm laser alignment is discussed in sections 2.1 and 2.2. Its frequency is locked to a temperature stabilized resonator built from ZERODUR® which has a residual drift of $\sim 2$ MHz per hour. We compensate for this by repeating identical spectral measurements, determining the line centres and interpolating the resonator drift.

All lasers are switched by acousto-optical modulators (switching times $\sim 50$ ns) and guided to the experiment by single-mode polarization-maintaining optical fibres. This ensures an unaltered alignment independent of changes on the input side, e.g. when changing the laser wavelength for excitation of different Rydberg states or for different detunings from the intermediate state.

Typically, 30 $\mu$s after excitation all Rydberg atoms are field-ionized (field pulse height 770 V cm$^{-1}$) and detected with a microchannel plate detector whose signal is recorded with a boxcar integrator. The delay of 30 $\mu$s between excitation and detection suppresses a background of ‘hot’ (300 K) atoms, while it has no influence on the measured excitation probability of the

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Preparation of a mesoscopic subensemble through spatially selective optical pumping and Rydberg excitation with a spatially homogeneous laser intensity. A small tube of atoms within a cloud of $10^7$ ultracold atoms is marked by optical pumping with the pumper and repumper laser beam. A mesoscopic subensemble containing about 100 atoms within this tube is transferred to a Rydberg state by a near-resonant two-photon excitation with two counter-propagating laser beams at 780 and 480 nm, respectively. The excitation laser at 480 nm has a flat-top intensity profile shown in the inset, while the beam at 780 nm superimposed on the pumper beam has a Gaussian profile as all other beams.

We have verified that no spurious ions are present which is as expected for the short timescales and small atom numbers considered throughout this paper. After detection the MOT is turned back on and the whole cycle is repeated every 70 ms.

2.1. Beam shaping

The 480 nm beam is focused and shaped to a constant intensity distribution over the excitation volume: the output of a single-mode polarization-maintaining fibre is collimated by an achromat (focal length 80 mm and 1 inch diameter) to a beam size (1/e$^2$ intensity radius) of 6.9 mm and shaped by a diffractive optical element (DOE) which is cemented onto a plano-convex lens with a focal length of 100 mm (see figure 3(a)). The DOE effectively distorts the plano-convex lens to give a flat-top profile at one specific distance close to the actual focus of the beam. All elements from the fibre coupler to the DOE are mounted in a rigid cage that can be aligned as a whole with respect to the MOT (see figure 3(b)).

Figure 3(c) shows the calculated behaviour of the DOE. The flattop is realized only at one specific distance from the DOE. Before this point the beam profile is closer to a Gaussian beam, behind this point some of the beam rays cross (so-called caustics), which leads to an increased intensity at the beam edges. Even further along the beam is the actual focus.

We have aligned the elements within the cage by adjusting the distance between fibre output and achromat as well as the position of the DOE with respect to the centre of the collimated beam, and characterized the beam shape with a USB webcam (Philips ToUCam Pro PCVC740K). The CCD chip has been replaced by a black-and-white version (ICX098BL) with...
Figure 2. Level scheme for optical pumping and near-resonant two-photon excitation of $^{87}$Rb Rydberg states. Addressing of a mesoscopic subset of atoms involves depumping, spatially selective repumping and optical pumping in order to single out the 3-level subsystem $|5S_{1/2}, F = 2, m_F = 2\rangle \rightarrow |5P_{3/2}, F = 3, m_F = 3\rangle \rightarrow |nD_{5/2}, |m_J = \ell + 1/2, m_J = J\rangle$, with $\ell = 0$ (S) or 2 (D). Decoherence effects from the short lifetime of the intermediate state are minimized by the detuning $\delta$ from the intermediate resonance thereby reducing the system to an effective two-level system.

2.2. Beam alignment: selection of a mesoscopic subensemble

The previous characterization of the shaped beam allows us to confine the excitation volume to the flat-top region with a dedicated optical pumping scheme: the magnetic field of the MOT is turned off 3 ms before the trapping lasers are switched off and all atoms are pumped to the dark $|5S_{1/2}, F = 1\rangle$ ground state within 300 $\mu$s by a depumping laser (see figure 2). All atoms in a small cylindrical tube perpendicular to the excitation beams are transferred to the upper hyperfine component $|5S_{1/2}, F = 2\rangle$ of the ground state with a 1 $\mu$s pulse of a tightly...
Figure 3. Realization of a flat-top beam profile for the 480 nm excitation laser. (a) Schematic: the output of a single-mode polarization-maintaining fibre (1) is collimated by an achromat to a $1/e^2$-intensity radius of 6.9 mm. The beam is focussed and shaped by a DOE which is cemented onto a standard plano-convex lens (3). (b) Supporting cage with (1) fibre connector, (2) achromat and (3) diffractive element which is mounted on a two-axes translation stage shown in black. The cage is fixed on an adapted three-axes mirror mount (4) which is fixed to the vacuum chamber (5). (c) Beam-shaping principle with ray optics. The collimated input beam (1) is focussed and shaped into a flat-top beam profile at one specific distance (3). Before this region the shape is closer to a Gaussian beam (2), behind this point crossing rays (caustics) cause an increased intensity in the beam edges. (d) Measured beam profiles: (2) best resemblance of a flat-top, (1) and (3) profiles at a distance of 200 $\mu$m from the flat-top. The actual focus (4) is 640 $\mu$m further along the beam and is used for alignment (see section 2.2). Differences between the theoretical and experimentally realized intensity profiles (e.g. c(3) versus d(2) or c(4) versus d(3)) are a consequence of imperfections in the actual set-up, e.g. slight misalignment, inaccurate diameter of input beam, imperfect diffractive element.
Figure 4. The excitation volume is given by the overlap region of a tightly focused repumping beam (red) and the shaped 480 nm beam (blue). The lasers are aligned by first overlapping both foci (minimizing the excitation volume) and then moving the flat-top region onto the red beam by moving the blue focus by 640 µm along the beam direction (see figure 3(d)). For details, see text.

The red beam is produced from the output of a single-mode fibre with a mounting that allows alignment along the directions (1) and (2). After a rough alignment the focus of the repumping beam is moved slightly above the blue beam by a small separation in the direction (1). Moving the focus along the direction (2) we only get an overlap with the 480 nm beam (and thus a Rydberg signal), when the size of the diverging repumping beam is larger than the beam separation. In this way, we can probe the Rayleigh-range of the red laser and thus find its focal point. After this, we can measure the local width of the blue beam by moving the red laser along the direction (1). As the mounting of the 480 nm beam allows movement of its focus along the direction (3), we measure the 480 nm beam diameter in this way at several positions along the direction (3) and thus overlap the 480 nm beam focus with the red beam. Finally, we move the flat-top region onto the red beam by moving the blue focus by 640 µm along the beam.

With a saturation intensity of 3.6 mW cm\(^{-2}\), this translates into 12 scattered photons on average at the centre of the repumping beam, 3.7 photons at a distance of 10 µm from the beam centre, and less than 0.4 photons at 15 µm. Only four photons are needed to repump the atoms with more than 90% probability. Therefore, the density is almost constant up to 10 µm and then quickly falls to zero. The non-uniform density at this edge is important for a detailed description of blockade effects [28].

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Figure 5. (Inset) Spectral dependence of both the background signal determined with a blocked repumping beam (red, with Gaussian fit) and the signal from the mesoscopic sample (green, with Lorentzian fit, background already subtracted) for a detuning of $\delta = 2\pi \times 110$ MHz from the intermediate level (see figure 2). For the detunings used in this paper the background signal is usually between one tenth and one half the size of the signal from the mesoscopic cold ensemble. (Main graph) Peak position for the background signal (red dots) for different values of $\delta$ together with the peak position of the mesoscopic signal (green dots). The dashed green line shows the expected dependence for a coherent two-photon excitation, the dashed red line corresponds to the expectation for a hot gas as given by equation (1). Where error bars are not visible they are smaller than the dot size.

Figure 3(d)). We estimate an alignment accuracy of 50 $\mu$m in the direction (3) and an accuracy on the order of 10 $\mu$m in the direction (1). The excitation volume is given by the beam diameters resulting in a volume of about $10 \mu$m $\times$ $10 \mu$m $\times$ $100 \mu$m which (at our density of $10^{10}$ cm$^{-3}$) corresponds to 100 atoms in the excitation volume.

2.3. Background suppression

As our excitation volume is defined by the overlap of the repumping and the 480 nm beam (see figure 1), we can determine a background signal by blocking the repumping beam. The inset of figure 5 shows the spectral dependence of both the background signal (red) and the signal from the flat-top region (green) on the detuning $\Delta$ of the 480 nm laser. The detuning of the 780 nm laser from the intermediate 5P$_{3/2}$, $F = 3$ state is fixed at $\delta = +2\pi \times 110$ MHz. The signal from the flat-top region peaks at $\Delta \cong -\delta$, i.e. the two-photon resonance condition is fulfilled, which is the signature of coherent excitation [23]. By contrast, the background signal peaks at a different detuning. Figure 5 shows the dependence of the peak positions for different values of $\delta$. The spectral dependence shows that the background signal comes from the hot background gas from which the cold cloud is loaded: due to their thermal velocity hot atoms experience the excitation lasers at their Doppler-shifted frequency. For atoms with a projected velocity $v$ in the direction of the 780 nm laser, this laser is resonant with the transition 5S$_{1/2}$ $\rightarrow$ 5P$_{3/2}$ if the Doppler shift $-2\pi \times v\lambda$ is equal to $\delta$, with $\lambda = 780$ nm being the laser wavelength. As the two
excitation lasers are counter-propagating these atoms are then resonant with the 480 nm laser if it is detuned from the resonance by

\[ \Delta = 2\pi \times \frac{v}{480 \text{nm}} = -\frac{780 \text{nm}}{480 \text{nm}} \delta. \]  

(1)

The dashed red line in figure 5 shows this dependence in good agreement with the experimental values for the background spectra.

In this way, the thermal velocity can be used to separate the signal from the hot atoms spectroscopically from the signal of the cooled atoms in the flat-top excitation volume. We reduce the background even further by delaying the detection: after each excitation we wait for 30 \( \mu s \) before detection. In this time, most of the hot atoms have already left the detection region which allows for a significant background suppression. Longer delays are not possible as the lifetime of the states discussed here are of the same order (e.g. 26 \( \mu s \) for 35D).

All the figures shown in this paper have been acquired by subtracting the background of hot atoms, which is determined by repeating the corresponding measurement with a blocked repumping beam.

2.4. Observation of Rabi oscillations

Our excitation scheme allows excitation to either \( nS \) or \( nD \) states (\( n \) denotes the principal quantum number) depending on the helicity of the polarization: we excite atoms to \( nS \) states via

\[ |5S_{1/2}, F = 2, m_F = 2\rangle \xrightarrow{\sigma^+} |5P_{3/2}, F = 3, m_F = 3\rangle \xrightarrow{\sigma^-} |nS_{1/2}, m_J = 1/2\rangle, \]

and \( nD \) states via

\[ |5S_{1/2}, F = 2, m_F = 2\rangle \xrightarrow{\sigma^+} |5P_{3/2}, F = 3, m_F = 3\rangle \xrightarrow{\sigma^+} |nD_{5/2}, m_J = 5/2\rangle, \]

as depicted in figure 2.3

In figure 6, the measured fraction of excited Rydberg atoms in the 31D\( 5/2 \) state as a function of excitation time is shown. The excitation time is given by the pulse length of the 480 nm laser, as the 780 nm light is switched on (off) 100 ns before (after) the 480 nm laser to ensure constant intensity during the on-time of the latter. Figure 6 shows a measured Rabi oscillation. It is damped out quickly. For times larger than 200 ns the number of Rydberg atoms reaches a steady state which indicates the balance between excitation and down-stimulation. The same temporal dependence is observed for \( nS \) and \( nD \) states with \( n \lesssim 40 \). For these small \( n \) it does not depend on the atom density. This indicates that the observed Rabi oscillations are not affected by Rydberg–Rydberg interactions and that all atoms simultaneously perform identical Rabi oscillations. As seen in figure 6, the oscillations are strongly damped, which is a result of the remaining admixture of the intermediate state, the residual intensity distribution of the flat-top beam profile, and our finite laser bandwidth. We will discuss these sources for decoherence in section 3.4.

3 Note that \( |5P_{3/2}, F = 3, m_F = 3\rangle = |5P_{3/2}, I = 3/2, m_I = 3/2, m_J = 3/2\rangle \).
Figure 6. Rabi oscillations of atoms excited to the $31D_{5/2}$ state at a detuning from the intermediate state of $\delta/2\pi = 140$ MHz and a Rabi frequency for the lower transition of $\omega/2\pi = 55$ MHz. Each dot corresponds to an average of measurements over 28 experimental repetition cycles. The dotted line is the simulated excitation probability for a perfect flat-top, i.e. a single Rabi frequency. In this case the damping comes from a small but finite admixture of the short-lived intermediate $5P_{3/2}$ state (see section 3.4). The dashed line shows a simulation that additionally averages over the measured intensity distribution. Finally, the solid line also takes an effective laser linewidth of 2.4 MHz into account. The MCP signal is given in V ns, and the axis is scaled to overlap with the simulated curve. The scaling factor constitutes the detector efficiency measured in mV ns per atom. The only free parameter for the simulations is the Rabi frequency for the upper transition which is compared to ab initio calculations in section 3.2.

3. Systematics

3.1. Dependence on the detuning from the intermediate state

If the detuning from the intermediate level is sufficiently large, one can reduce the actual three-level system to a two-level system with an effective Rabi frequency of $\Omega_{\text{eff}} = \omega \Omega / 2\delta$, where $\omega$ ($\Omega$) denotes the Rabi frequency of the lower (upper) transition and $\delta$ denotes the detuning from the intermediate level. We have verified this dependence by measuring the effective Rabi frequencies for different detunings from the intermediate level. The effective Rabi frequencies are determined by a fit of a damped oscillation, $\frac{1}{2} [1 - \exp(-t/\tau_{\text{damp}}) \cos(\Omega_{\text{eff}} t)]$, $\tau_{\text{damp}} = 60$ ns, to measured Rabi oscillations. The resulting fit values together with the fit uncertainties are depicted in figure 7. They are in very good agreement with the expectation for an effective two-level system.

3.2. Comparison to ab initio transition matrix elements

The experimental values of the Rabi frequencies are compared to ab initio calculations, yielding a concise test of predicted transition matrix elements. The Rabi frequency scales with the blue laser intensity $I$ as $\Omega \propto \mu \sqrt{I}$, where $\mu$ is the matrix element between the states coupled by the laser light, i.e. the $5P_{3/2}$ and the Rydberg state.
Table 1. Comparison between measured Rabi frequencies and calculations based on \textit{ab initio} transition matrix elements.

| Final state | \( \Omega_{\text{exp}}/2\pi \) | \( \Omega_{\text{theo}}/2\pi \) |
|-------------|---------------------------------|---------------------------------|
| 30D_{5/2}   | 32.8 ± 1.7 MHz                  | 37.6 MHz                        |
| 31S_{1/2}   | 18.9 ± 0.6 MHz                  | 19.3 MHz                        |

Figure 7. Measured effective Rabi frequencies \( \Omega_{\text{eff}}/2\pi \) for different detunings \( \delta/2\pi \) from the intermediate state. The solid line shows the theoretical prediction for a blue Rabi frequency \( \Omega/2\pi = 27 \text{ MHz} \) and a red Rabi frequency \( \omega/2\pi = 55 \text{ MHz} \) as deduced from an Autler–Townes splitting.

To determine \( \mu \) we split the Rydberg electron wavefunctions \( \psi_{n,\ell,j,m} \) into a radial and a spherical part with the ansatz

\[
\psi_{n,\ell,j,m} = \sum_{m_s} \frac{1}{r} U_{n,\ell,j}(r) Y_{\ell,m_j-m_s}(\theta, \phi) C_{\ell,m_j,m_s,1/2,m_s}^{j,m_j,-m_j} \Theta_{m_s}^{1/2},
\]

where \( Y_{\ell,m} \) denotes the well-known spherical harmonics, \( C_{\ell,m_j,m_s,1/2,m_s}^{j,m_j,-m_j} \) is the Clebsch–Gordan coefficient and \( \Theta_{m_s}^{1/2} \) is the spin wavefunction. The radial part is obtained by numerically integrating the Schrödinger equation with the Numerov algorithm following [29] at the energy \( E_{n,\ell,j} \) (calculated with the according quantum defect). To obtain reasonable results for low-lying levels, we use a model potential that is fitted to one-electron energies [30]. The knowledge of the radial wavefunctions allows us to calculate the radial part \( \mu_{\text{rad}} \) of the dipole transition matrix element \( \mu = \mu_{\text{rad}} \times \mu_{\text{sph}} \) with

\[
\mu_{\text{rad}} = \left\langle \frac{1}{r} U_{5P3/2} \right| \left| \frac{1}{r} U_{n,\ell,j} \right\rangle.
\]

For large principal quantum numbers it scales as \( \mu_{\text{rad}} \simeq C_{\ell} (n^*)^{-3/2} \), where \( n^* \) denotes the principal quantum number reduced by the appropriate quantum defect. For \( n \geq 30 \), we find \( C_0 = 4.508 \text{ au} \) and \( C_2 = 8.475 \text{ au} \), while the spherical matrix elements for the stretched transitions as depicted in figure 2 are \( \mu_{\text{sph}} = \sqrt{1/3} \) for \( \ell = 0 \text{ (S)} \) and \( \sqrt{2/75} \) for \( \ell = 2 \text{ (D)} \). Table 1 shows
Figure 8. Quenching of Rydberg atoms by stimulated emission. At 150 ns, the laser at 780 nm for the lower transition is turned off, and Rydberg atoms in the $32S_{1/2}$ are stimulated to the $5P_{3/2}$ state by the laser light at 480 nm. Experimental values of the total Rydberg excitation with the corresponding statistical errors are shown as dots. The solid line shows the excitation probability of the $32S_{1/2}$ state as predicted by a model calculation with no free parameters. The scaling factor for the MCP signal was determined by comparing the corresponding Rabi oscillation to a simulated curve as in figure 6.

the comparison between the experimental values and the theoretical prediction. While there is excellent agreement for the $31S$ state, the measured value for the $30D$ state is slightly smaller than expected. The deviation is most likely caused by electric stray fields which induce an admixture of other $m_J$-states effectively reducing the spherical matrix element. This effect does not perturb the $S_{1/2}$ state as $m_J = 1/2$ is the only dipole-coupled state and the Stark effect on the intermediate $5P_{3/2}$ state is negligible.

3.3. Redistribution to other states

Redistribution of Rydberg atoms to other states due to population transfer processes has already been observed to reduce the efficiency of de-excitation (quenching) in a macroscopic cloud of Rydberg atoms [22]. By measuring the quenching for the $32S_{1/2}$ state as shown in figure 8, this process can be ruled out for the small mesoscopic ensemble described here. After 150 ns of excitation the 780 nm laser is turned off, while the laser for the upper transition is left on. Due to the short lifetime of the intermediate $5P_{3/2}$ state, population inversion is created. The blue laser stimulates transitions of the Rydberg state to the intermediate state, which then quickly decays to the ground state. Redistribution processes would lead to population in other Rydberg states which are not coupled by the blue laser light. As these atoms are still field ionized, they should result in a finite MCP signal even for long quenching times. As figure 8 shows, the total number of Rydberg atoms is almost fully quenched by stimulated emission. In addition, the temporal evolution of the Rydberg signal is in good agreement with the solution of the Bloch equations for a pure three-level system shown as a solid line. Redistribution of Rydberg population can therefore be ruled out as a major cause of decoherence in the mesoscopic ensemble.
3.4. Sources for decoherence

Besides redistribution of Rydberg atoms to other states, three factors cause the reduced contrast and the damping of Rabi oscillations: the residual inhomogeneity of the flat-top intensity profile, the small but finite admixture of the short-lived intermediate $5P_{3/2}$ state, and the finite linewidth of the laser source. Note that only the last two effects are sources of decoherence for all atoms while the first effect only represents ensemble averaging with full coherence for each individual atom.

The influence of the intermediate state and the residual intensity distribution on the damping of the Rabi oscillations are simulated by solving the optical Bloch equations (OBE) for the 3-level system and averaging over different blue Rabi frequencies according to the measured intensity distribution shown in figure 2. The only free parameters are the detection efficiency of the MCP and the average Rabi frequency of the upper transition, which agree well with theory (see section 3.2). The resulting simulations are depicted as dotted and dashed lines in figure 6 showing damped Rabi oscillations with a reduced contrast. The dotted line is a simulation for a single Rabi frequency and shows a reduced contrast due to the admixture of the intermediate state which can be decreased by increasing the detuning $\delta$ which in turn results in a smaller effective Rabi frequency. The dashed line additionally incorporates an averaging over different Rabi frequencies corresponding to the measured residual intensity fluctuation in the flat-top beam profile. The inhomogeneous distribution of Rabi frequencies alone does not fully explain the reduced measured contrast.

Indeed, the main contribution to the reduced contrast is the finite bandwidth of the laser sources. As the admixture of the intermediate level is small we can incorporate the laser bandwidth $\gamma$ into the simulation by solving the OBE for a two-level system with a linewidth of $\gamma$ and an effective Rabi frequency of $\Omega_{\text{eff}} = \omega \Omega / 2\delta$, where $\omega$ ($\Omega$) denotes the Rabi frequency of the lower (upper) transition and $\delta$ denotes the detuning from the intermediate level. This is a good approximation for $\delta \gg \omega$, $\Omega$ [31]. We have again averaged over the different values of $\Omega_{\text{eff}}$ as present in the imperfect flat-top. The corresponding model calculation for an effective incoherent excitation bandwidth of $\gamma = 2\pi \times 2.4$ MHz is shown as a solid line in figure 6 in excellent agreement with the experimental data. The experimental value for $\gamma / (2\pi)$ agrees well with the specified bandwidths of 2 MHz for the blue and 1 MHz for the red excitation lasers. In addition, we have performed beating measurements with two comparable, independent lasers at 780 nm which yielded a combined bandwidth of 2 MHz, in good agreement with the above values.

4. Van der Waals blockade

The excitation presented so far has been performed in a regime with negligible interactions at small $n$. For large values of $n \gtrsim 60$, we do not observe Rabi oscillations as the Rabi frequency, scaling with $n^{-3/2}$ becomes smaller than the dephasing rate. On the other hand, these higher lying states exhibit stronger Rydberg–Rydberg interactions which can result, for example, in a van der Waals blockade. Figure 9(c) shows how the excitation probability of the $75S$ state depends on the ground-state density. For a single (or non-interacting) atom we can solve the OBE for the independently determined experimental parameters and obtain a value represented by the dashed line. For low densities, the measured excitation probability agrees well with the prediction. With increasing density, we observe increasing suppression of the excitation probability.
Figure 9. Excitation blockade for a mesoscopic ensemble of atoms in the 75S state. The upper graphs show excitation spectra for the highest (a) and lowest (b) investigated densities together with Lorentzian fits. A higher density is clearly accompanied by smaller excitation probability and a significant line broadening. The excitation probabilities are calibrated by measurements at low $n$, which results in single-atom excitation probabilities. (c) Shows the maximum excitation for different ground-state densities. The dashed line corresponds to the theoretical excitation probability for non-interacting atoms. For higher densities the Rydberg–Rydberg interaction energy rises and moves the excitation out of resonance reducing the excitation probability on resonance. The corresponding line broadening (shown in (d) with an identical $x$-axis) also reflects the increasing interaction energy.
probability. It is accompanied by a significant line broadening which is shown as the inset in figure 9(d) as well as in the exemplary spectra in figures 9(a) and (b). For small densities, the measured linewidth agrees well with a convolution of our laser bandwidth (2.4 MHz) and saturation broadening (1.7 MHz).

The suppression of excitation marks the onset of an interaction-induced blockade. The underlying interaction has van der Waals character with a $C_6/R^6$ dependence on the interatomic spacing $R$ in contrast to dipole–dipole interaction with $R^{-3}$ character, which was originally proposed [3]. In comparison this means a shorter interaction range and in fact we see the suppression due to vdW interaction only for high principal quantum numbers $n$ as the vdW coefficient scales as $C_6 \propto n^{11}$.

These results complement our earlier measurements with macroscopic atom clouds [17] which showed a much stronger broadening at comparable densities and excitation suppression. In fact, the observed broadening in the macroscopic cloud cannot be explained by mere Rydberg–Rydberg interactions [32]. One possible explanation may be ions that are produced by interaction-induced collisions between the cold atoms after acceleration by the strong van der Waals forces between Rydberg atoms [33, 34]. For the mesoscopic system investigated here we have ruled out the presence of ions directly by selective field ionization. This is also expected for the small number of involved atoms and the short excitation times.

5. Conclusion

We have observed Rabi oscillations of a mesoscopic cloud of about 100 atoms in the coherent excitation of Rydberg states out of an ultracold atom cloud. The reduced contrast of the Rabi oscillation is traced back to residual intensity variations and the finite bandwidth of our laser system. While the latter can simply be solved improving the frequency stability of the laser systems, one option to bypass the first limitation is the use of liquid crystal elements [35] to dynamically optimize the diffractive pattern for better results.

In addition, we have demonstrated the van der Waals blockade at large principal quantum numbers for a mesoscopic cloud. This indicates the onset of a local blockade as has been observed before for macroscopic clouds [16]–[21]. The full blockade regime can be identified by an increased Rabi frequency $\sqrt{N} \Omega$, where $N$ and $\Omega$ denote the number of atoms and the atomic Rabi frequency, respectively [3]. The experiments presented here constitute a necessary prerequisite for the direct observation of this coherent many-body phenomenon. In future works, we will use a number of options to reach this regime by increasing the interaction strength through higher densities [21] and by using either resonant [18] or permanent [19] dipole–dipole interactions. Smaller excitation volumes [24] will provide ensembles where even the interaction between the farthest separated atoms dominates all other energy scales.

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