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From coherent collective excitation to Rydberg blockade on an atom chip

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Using time-resolved measurements, we demonstrate coherent collective Rydberg excitation crossing over into Rydberg blockade in a dense and ultracold gas trapped at a distance of 100 µm from a room-temperature atom chip. We perform Ramsey-type measurements to characterize the coherence. The experimental data are in good agreement with numerical results from a master equation using a mean-field approximation, and with results from a super-atom-based Hamiltonian. These results represent significant progress in exploring a strongly interacting driven Rydberg gas on an atom chip.

Rydberg atoms are being actively studied for their strong, tunable interactions, making them interesting tools for quantum information science, e.g. for quantum computing [1, 2] and quantum simulation [3, 4] and for realizing strong photophoton interactions [5–12]. Remarkable progress has been made both with ensembles [8, 13–15] and with individual atoms [16–18]. In dense samples the strong van-der-Waals interaction between Rydberg atoms limits excitation from the ground state to a single Rydberg excitation within some ‘blockade’ volume. At the same time atoms within this volume experience a coherent, collective coupling to the excitation light (as a ‘superatom’) with a Rabi frequency that is enhanced by \( \sqrt{N_b} \), where \( N_b \) is the number of participating atoms [19]. The coherence of the excitation competes with decoherence mechanisms such as spontaneous and black-body-induced decay of the Rydberg states [20–22], atomic motion, the finite bandwidth of the excitation lasers, and, particularly in a trapped gas, with dephasing through the inhomogeneous density distribution [19, 23, 24]. Yet, collective enhancements of Rydberg excitations were observed on both one- [25] and two-dimensional lattices [26], in optical tweezer arrays [16, 17, 27] and between physically separated ensembles that are individually dense enough to act collectively [14].

An ongoing challenge is to combine Rydberg-mediated interactions with atom chips [28], microfabricated devices designed to cool and trap neutral atoms close to surfaces. Such a combination would be an excellent starting point for hybrid quantum systems [29, 30], e.g. combining solid-state and neutral-atom-based qubits, but also for building compact quantum devices based on neutral atoms.

However, thus far coherent Rydberg physics on atom chips has been difficult to observe due to decoherence from inhomogeneous Stark shifts induced by stray electric fields emanating from the surface [31–34]. While first evidence of Rydberg–Rydberg interactions on an atom chip has been demonstrated in a cryogenic environment [35], the saturation of Rydberg atom number could not be observed, and the excitation bandwidth was too large to probe the initial coherence. Here, we demonstrate two key aspects of resonant Rydberg driving in a dense and ultracold gas, trapped at around 100 µm distance from a room-temperature atom chip.

Using time-resolved measurements we detect (i) the early-time coherent regime of collectively enhanced excitations, crossing over at a later time into (ii) a regime with essentially constant Rydberg fraction due to the blockade mechanism. In regime (i) we find a collective enhancement of the Rydberg
Rabi frequency by up to an order of magnitude compared to the single-atom case. This implies that there are up to 100 ground-state atoms per blockade volume in our system. In regime (ii) we have a Rydberg fraction much below unity (on the order of 1%), consistent with the inferred blockade volume and despite the strong resonant driving of the system. Via a Ramsey-type pulse sequence we directly probe the initial collective coherence and its decay. We describe the dynamics using both an effective mean-field master-equation approach covering the full time range, and a one-dimensional (1D) coherent superatom-based description for the onset of the excitation, and find good agreement. These results constitute important progress in realizing coherent driving to a strongly interacting Rydberg gas in an integrated quantum device, and in the presence of several decoherence mechanisms.

The experimental arrangement is very similar to that described in Ref. [33]. In short, we trap $^{87}$Rb atoms in the $|F, m_F \rangle = |2, 2 \rangle$ state at a distance of $\sim 100 \mu m$ from the surface of an atom chip. The magnetic trap is generated with a Z-shaped gold wire patterned on a silicon substrate [36]. The temperature and atom number of the atomic cloud are controlled by varying the final frequency of the radio-frequency (RF) sweep used for evaporative cooling. This results in a cloud of typically $2 \times 10^4$ atoms at a temperature of a few $\mu K$ in the final magnetic trap with radial and axial trap frequencies of $\omega_r/2\pi = 860 \text{ Hz}$ and $\omega_z/2\pi = 46 \text{ Hz}$, respectively, and with its trap bottom at $B_0 = 3.4 \text{ G}$. Thus, the atoms are confined in a highly elongated, cigar-shaped geometry with a typical width $\sigma_r = 2.6 \mu m$ and length $\sigma_z = 48 \mu m$ (at $T = 2 \mu K$); see Fig. 1(a).

For Rydberg excitation, we use a two-photon excitation scheme with a 780-nm (‘red’) and a 480-nm-wavelength (‘blue’) laser. These propagate in opposite directions along the length of the cloud and have beam waists ($520 \mu m$ and $90 \mu m$, respectively) that are much larger than the width of the cloud so that the (single-atom) Rabi frequency is constant across the cloud. We excite either the $|28D_{5/2}, m_J = 5/2 \rangle$ or the $|30S_{1/2}, m_J = 1/2 \rangle$ state. The red laser is blue detuned by $\Delta_r/2\pi = 100 \text{ MHz}$ from the intermediate $5P_{3/2} \rightarrow 5S_{1/2}$ transition to prevent off-resonant scattering from it. The blue laser is detuned by $\Delta_b \approx -\Delta_r$ such that the overall detuning $\Delta = \Delta_r + \Delta_b$ is close to resonance. To change the overall detuning, we vary $\Delta_b$. The intermediate state can be adiabatically eliminated because $\Delta_r \gg \hbar \Omega_r, \Omega_b$ [37], so that a two-level system with a Rabi frequency of $\Omega = \hbar \Omega_r/2\Delta_r$ remains (here $\Omega_r$ and $\Omega_b$ denote the red and blue Rabi frequency, respectively). The excitation lasers are stabilized using a reference cavity and have a linewidth of typically $10 \text{ kHz}$ [38]. The 780-nm beam is sent through a combination of a mechanical shutter and an acousto-optical modulator for excellent contrast and fast switching times, respectively. For switching the 480-nm beam a mechanical shutter suffices.

Losses from the trap are detected via absorption imaging of the remaining atoms after a 6-ms time of flight. A decrease in the atom number is interpreted as losses due to Rydberg excitation. Note the significant difference between magnetic trapping and the more common technique of optical trapping in this regard: in optical traps, all the sublevels of the electronic ground state are trapped, and decay of Rydberg states to the ground state does not necessarily lead to losses. In a magnetic trap, however, most ground state sublevels are not trapped, and Rydberg excitation and subsequent decay will lead to particle loss. Most experiments described here are performed using a series of pulses (see below), with a pulse separation longer than the Rydberg lifetime. This allows us to investigate shorter timescales, where the losses per pulse are small, while still having an observable loss signal.

Due to their large orbitals, Rydberg atoms have a strong van-der-Waals interaction given by $-C_b/r^6$, where $r$ is the interatomic distance, and $C_b$ is the (state-dependent) effective interaction coefficient [19, 33, 39–41]. For typical nearest-neighbor distances in an ultracold ground-state gas the strength of this interaction is larger than the excitation bandwidth, so it is not possible to excite a second Rydberg atom within some distance from a prior Rydberg excitation, leading to blockade. The relevant length scale over which Rydberg blockade is effective, known as the blockade radius [19, 42], is in this case given by $|C_b/\hbar \zeta|^{1/6}$, where $\zeta$ is the effective excitation bandwidth. For narrow-bandwidth lasers (and as long as other decoherence mechanisms can be neglected) $\zeta$ is given by the generalized Rabi frequency $\sqrt{N_b \Omega^2 + \Delta^2}$, where the Rabi frequency is enhanced due to the collective coupling. For our parameters the blockade radius is typically more than $1 \mu m$. Since the radial extent of our cloud is usually on the order of the blockade radius, the system is close to being 1D in terms of the geometrical extent of the Rydberg blockade, and we can only excite one Rydberg atom in the radial direction.

To demonstrate the presence of Rydberg blockade, we perform an experiment using the $|28D_{5/2}, m_J = 5/2 \rangle$ state, see Fig. 1(b). This state has a lifetime of $15.2 \mu s$ (including blackbody radiation at $T = 300 \text{ K}$ [43]). We compare the losses after a single $37.5-\mu s$ pulse to those after a series of 75 pulses with a duration of 500 ns apiece, in which case the pulse separation is longer than the Rydberg lifetime. In a naive picture that ignores the blockade and coherent behavior, one would expect these to lead to the same results since the pulse areas are the same. In Fig. 1(b) we can see that this is clearly not the case, the continuous pulse has far fewer losses compared to the pulse train. This can be interpreted in terms of the blockade strongly limiting the total Rydberg atom number during the continuous pulse. This effect is lifted when the pulse is split up in smaller segments, enhancing the losses, so that the Rydberg resonance becomes clearly visible as a loss feature. The results are in very good agreement with a mean-field master equation model (see below).

Much more information can be extracted from the full time evolution while on resonance. For comparison, we look at the $|30S_{1/2}, m_J = 1/2 \rangle$ state as well. The parameters of this state are similar to those of the $28D_{5/2}$ state, except its van-der-Waals interaction is isotropic and repulsive [40, 41, 43]. For short times, we used multiple pulses and extracted the average losses per pulse afterwards, see Fig. 2.
For the single-atom Rabi frequencies used here (typically around $2\pi \times 100$ kHz) three different regimes in the excitation evolution are clearly visible in Fig. 2. Specifically, we can distinguish between a coherent onset (where the scaling of losses with time is superlinear), a blocked intermediate regime (where this scaling is sublinear), and a linear regime at later times. The latter is best understood as the regime where the cumulative number of atoms that has left the trap after decaying from the Rydberg state is larger than the number of atoms in the Rydberg state at any time. This lost atom number grows linearly since the process is governed by a (constant) decay rate. Note that for short pulse times, we are sensitive to losses below the $10^{-3}$ level (using 500 pulses), i.e. to the loss of only a few atoms per pulse.

The experimental results are explained well using a master-equation model following Ref. [44], see also [45]. In short, the ground state and Rydberg state of each atom can be written as the spin-down and spin-up state of a pseudospin $1/2$. The Rydberg–Rydberg interactions and the coherent coupling by the excitation lasers are then described by a spin Hamiltonian. By assuming an ‘exclusion region’ around every Rydberg excitation which is otherwise embedded in a sea with a homogeneous Rydberg density, one can make what amounts to a self-consistent mean-field approximation. The effects of decoherence are included via Lindblad terms which are added to the master equation for the single-particle density matrix, effectively describing the system in a local-density approximation. We numerically integrate this equation at different densities, and average the results over the cloud.

The results of the model [solid lines in Figs. 1(b) and 2] are in good agreement with the experimental data once we include a decoherence rate on the order of $10^6$ s$^{-1}$ (see Supplemental Material, and the discussion below). In particular, the initial fast, superlinear rise in losses originating from the initial coherent Rydberg population buildup is reproduced, as well as the subsequent saturation of the Rydberg population, leading to a plateau in the relative losses as a function of time, until the cumulative losses via the Rydberg state start to dominate and the loss becomes linear with time. The key to observing the initial coherent rise is that while the single-atom Rabi frequency is lower than the decoherence rate, the collective Rabi frequency is enhanced (in our case by up to an order of magnitude) to the point that it dominates the short-time behavior.

We have also simulated the system using the full many-body Hamiltonian describing the excitation of interacting Rydberg atoms, whilst ignoring inelastic and decohering processes. In principle, this requires an exponentially large basis making calculations intractable even for a relatively small number of atoms. The Rydberg blockade, however, prohibits a large fraction of these basis states to be populated, meaning the calculation can be simplified dramatically. We do so by dividing the cloud in so called ‘superatoms’: entities consisting of multiple ground-state atoms that couple to the light field collectively [44, 45]. Furthermore, we restrict ourselves to a one-dimensional description, in which the atoms are assumed to lie on a line. This way, it is computationally feasible to propagate the Hamiltonian over the first few $\mu$s which is sufficient to analyze the coherent start. To compare to the experimental data, the calculated Rydberg fraction at the end of the pulse is then interpreted as the lost fraction. The results are shown as dotted lines in Fig. 2. These confirm the short-time results of the mean-field model. For intermediate times (after the initial fast rise), the dotted lines show how the inhomogeneous density along the length of the cloud leads to a dephasing of the collective Rabi oscillations at various positions in the cloud, even in the absence of other decoherence mechanisms. The difference in saturation level between the mean-field and fully coherent models can be (partially) attributed to the fact that the latter is strictly one-dimensional, while the mean-field model does not impose any restrictions on the number of Rydberg excitations that can be present in the radial direction.

The differences that we observe between the $S$ and $D$ state can be explained in terms of the different Rabi frequencies used during the excitation. The fact that the $28D_{5/2}$ state has an anisotropic interaction does not appear to matter much, as our models use a root-mean-square averaged value and still produce a good result.

In order to demonstrate the coherence explicitly, we have performed Ramsey-type experiments. We employed a double-pulse scheme, where we varied the detuning and used two pulses of 250 ns with a 250 ns gap in between them. To increase the signal we repeated such a pulse pair 300 times. The observed Ramsey fringes are shown in Fig. 3(a). The results of the mean-field approach (solid lines in Fig. 3) are in excellent agreement with our measurements. On the other hand, the coherent propagation of the Hamiltonian shows the...
same qualitative behavior, with much stronger contrast. This can be attributed to the absence of decoherence in that model.

Convolving the measured spectrum with a Gaussian effectively simulates inhomogeneous spectral broadening. In this way, we can determine the effective linewidth $\sigma$ and find best results for a Gaussian with a width of $\sigma = 0.42$ MHz. We measure the decoherence time directly in a final experiment, namely a Ramsey measurement in the time domain. To this end, we detune the lasers to the red by $\Delta/2\pi = -2$ MHz, and again use two 250 ns pulses, but now vary the delay time between them. Repeating the pulse pair 100 times, we observe a decaying sines, see Fig. 3(b). From a fit to this we can extract a coherence time of $1/\gamma = 0.44 \mu$s.

In the future, finer control of the electric field components can be gained by adding more on-chip electrodes. This would allow compensating electric field gradients (thereby reducing the dominant source of dephasing), and significantly extend the coherence time. In parallel, adding an optical lattice along the length of the cloud (as in Ref. [23]) will turn the system into a linear array of tens to hundreds of atomic ensembles on a chip, with the distance between sites smaller than the characteristic blockade radius. An intriguing further possibility would then be to use an electric field gradient along the length of the cloud to allow for site-specific control over the excitation within such an array. Furthermore, it should be relatively straightforward to extend these results to colder and denser clouds and Bose-Einstein condensates, to reach more deeply into the one-dimensional regime where the radial cloud size is significantly smaller than the blockade radius. It would also be interesting to extend the Ramsey sequence used here to more advanced (spin-echo-like) pulse sequences, in order to further distinguish and characterize the various dephasing and decoherence mechanisms mentioned above.

In conclusion, we have directly observed blockade and coherence in the excitation of Rydberg atoms in a mesoscopic ensemble of atoms trapped on an atom chip. We have analyzed the measured spectra and the dynamical evolution both by solving the many-body Hamiltonian and a mean-field master equation. We are able to characterize and quantify the amount of decoherence, which we attribute to inhomogeneous fields resulting from the present chip design. These results open up promising routes towards quantum information science with Rydberg atoms in integrated quantum devices.
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SUPPLEMENTAL MATERIAL

Superatom Hamiltonian and mean-field master equation

Here we describe both the super-atom-based model and the mean-field master-equation approach in more detail. These approaches are directly based on Ref. [44] and Ref. [24], respectively. In a typical experimental run, we couple $N \sim 10^4$ ground state atoms to the Rydberg state. Since interactions play a role, we cannot hope to understand our system by resorting to a description that relies on $N$ two-level systems. Instead, our theoretical treatment should ideally consider all possible correlations that can be present. Given the large number of particles in our system, this is clearly intractable, and we have to reduce the size of the basis somehow. We start our discussion with the full Hamiltonian; for $N$ particles, it is given by

$$
\mathcal{H} = \sum_{j=1}^{N} \left( \frac{\hbar \Omega}{2} \sigma_x^{(j)} - \frac{\hbar \Delta}{2} \sigma_z^{(j)} + \mathcal{H}_{\text{int}}^{(j)} \right), \quad (S1)
$$

here $\Delta$ and $\Omega$ are the laser detuning and Rabi frequency, respectively, and $\sigma_x^{(i)}$ are the Pauli spin matrices of the $i$th atom. The last term describes the Rydberg–Rydberg interactions, it is given by

$$
\mathcal{H}_{\text{int}}^{(j)} = -\frac{C_6}{2} \sum_{k \neq j} \frac{\rho^{(j)} \rho^{(k)}}{|x_j - x_k|^6}. \quad (S2)
$$

Here $C_6$ is the van der Waals-interaction coefficient of the specific Rydberg state, $\rho^{(j)} \equiv |r_j \rangle \langle r_j|$ is a projection operator onto the Rydberg state, and $x_j/k$ are the atom positions; the sum runs over all other atoms $k$. We assume the system to be in the frozen gas limit, meaning the displacement during the excitation is negligible.

The dimension of the Hilbert space that this Hamiltonian samples can be reduced by factoring the basis into superatoms [13, 44, 45]. Physically, $N_b$ ground state atoms within one blockade sphere couple collectively to the light field at an enhanced Rabi frequency $\Omega_c = \sqrt{\Omega_0 \Delta}$. They will oscillate between an overall ground state ($\mid g \rangle \equiv |g_1 g_2 \cdots g_{N_b} \rangle$) and the $W$-state (a symmetric superposition of all possible states with a single Rydberg excitation):

$$
\mid W \rangle = \frac{1}{\sqrt{N_b}} \sum_{j=1}^{N_b} |g_1 \cdots r_j \cdots g_{N_b} \rangle. \quad (S3)
$$

This same principle can be used to reduce the basis size, even though the concept of a superatom is somewhat ill-defined in a dilute system such as ours, where the blockade radius is smaller than the system size. The principle of operation is as follows: we begin with a randomly sampled distribution of atoms satisfying some global parameters such as density and system size; we then find the two nearest-neighbors which are combined into a virtual superatom. This superatom gets an increased ‘weight’ signifying the number of ground-state atoms that constitute it. The square root of this number denotes its enhanced coupling to the light field. This process is iterated, each time finding the nearest (super)atoms and combining them into a new superatom while adjusting the weight according to the total number of atoms that constitute the superatom.

There are some physical ‘common sense’ requirements: it is, for instance, unphysical to have a smaller number of virtual superatoms than actual superatoms one would have in a physical system for the simple reason that this would lead to an underestimation of the total number of Rydberg atoms. Using more virtual superatoms than there are actual ones, on the other hand, does not lead to an overestimate, since in that case they will simply have a residual interaction, naturally limiting the number of Rydberg atoms.

The distribution of superatoms, then, is used to generate a basis, using which a density matrix $\rho$ and Hamiltonian matrix $\mathcal{H}_{\text{S}}$ can be generated. The dynamical behavior is described by the von Neumann equation:

$$
\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar}[\mathcal{H}_{\text{S}}, \rho], \quad (S4)
$$

(where $[\hat{A}, \hat{B}] \equiv \hat{A}\hat{B} - \hat{B}\hat{A}$ denotes the commutator) from which we can extract the number of Rydberg atoms as a function of time.

The distribution of atoms in our experiment is Gaussian, this means that for a given peak density, there is a large variety of other densities influencing the behavior. For instance, at the edges of the cloud, where the density is lower, the Rabi frequency is not enhanced as much as it is in the center. This leads to a slower Rabi oscillation. It would not be feasible to account for all of these different scalings in one simulation—the required number of superatoms would simply be too large.

We account for this by solving Eq. (S4) for a distribution with constant density, assuming periodic boundary conditions. Sampling several such ‘infinite systems’ at different densities, and averaging the result, should give a fair idea of what happens in a physical system. Using this approach, a typical calculation such as shown in Fig. 2 takes one day on a 16-core node of our cluster using an Intel® Xeon® E5-v2 processor.

This microscopic description works well for relatively short timescales; its validity is limited by the coherence time. One solution would be to add decoherence in the form of a master equation with Lindblad operators, but alternatively we can resort to effectively single-atom rate equations, which have recently proven their merit for driven-dissipative systems like these [22, 47–49]. They have the additional advantage of lower computational costs.

We choose a midway between these alternatives: a single-atom master equation where the Rydberg interactions are added as a mean-field approximation. This is a simple model that works surprisingly well for both long and short time scales, and has proven its merit in explaining different scaling behaviors observed elsewhere [24].

We start by removing the complicated spatial correlations from the problem by replacing the interaction term in the Hamiltonian (S1) with an interaction with the ’mean field’ of all
other Rydberg excitations in the system (under the assumptions that there is at most one excitation per blockade volume). As shown in Ref. [24], this reduces Eq. (S2) to a single-atom term of the form

\[ \mathcal{H}_{\text{int}}^{(i)} = V_j \sigma_z^{(i)} / 2, \]  

(S5)

where the mean-field interaction energy \( V_j \) is given by:

\[ V_j = -C_b \left( \frac{4\pi}{3} \text{mn}_0 \right)^2. \]  

(S6)

This leads to the single-particle, mean-field Hamiltonian:

\[ \mathcal{H}_{\text{MF}}^{(j)} = \frac{\hbar \Omega}{2} \sigma_x^{(j)} - \frac{1}{2} (\hbar \Delta - V_j) \sigma_z^{(j)}. \]  

(S7)

Here \( m \) is the magnetization, which is nothing but the average Rydberg fraction. When we describe the system in terms of three levels (a ground state \(|g\rangle\), a Rydberg state \( |r\rangle \), and a loss state \(|l\rangle \)) this is equivalent to the \( \rho_{rr} \) entry in the single-atom density matrix.

The single-atom, mean-field, Liouville–von Neumann master equation is now given by

\[ \frac{\partial \rho^{(j)}}{\partial t} = -\frac{i}{\hbar} \left[ \mathcal{H}_{\text{MF}}^{(j)}, \rho^{(j)} \right] + L \rho^{(j)} \rho + \frac{1}{2} \left[ L^\dagger \rho, \rho^{(j)} \right] + L^{(j)}_{\text{ph}}, \]  

(S8)

where \( \{ A, B \} \equiv \hat{A} \hat{B} + \hat{B} \hat{A} \) denotes the anti-commutator, and \( L \) is the jump operator describing spontaneous decay at a rate \( \gamma \) from \( |r\rangle \) into \( |g\rangle \) and \( |l\rangle \) with a branching ratio \( b \):

\[ L = \begin{pmatrix} 0 & \sqrt{(1-b) \gamma} & 0 \\ 0 & 0 & 0 \\ 0 & \sqrt{b \gamma} & 0 \end{pmatrix}. \]  

(S9)

Our linewidth, \( \gamma_0 \) is generally limited by experimental factors such as stray fields or the finite laser linewidth. We add this using the phenomenological Lindblad operator

\[ L^{(j)}_{\text{ph}} = \begin{pmatrix} 0 & -\gamma_0 \rho_{rg} / 2 & 0 \\ -\gamma_0 \rho_{rg} / 2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \]  

(S10)

Equation (S8) constitutes a system of differential equations that we can solve in order to obtain state populations as a function of time. In the incoherent limit it is equivalent to the rate equation description [22, 47–49], this can be derived by so-called ‘adiabatic elimination’, that is: assuming the off-diagonal elements of the density matrix are constant (i.e. \( \partial \rho_{jk} / \partial t = 0 \forall j \neq k \)), solving those equations, and finally substituting the results in the expressions for the populations (i.e. \( \rho_{jj} \)).

We have used this approach to analyze our experimental results, see the main text. Since the density in our system is not homogeneous, we modeled the system for different densities and took a representative average. We also assumed that all Rydberg atoms that are excited leave the system before they decay, hence we set \( b = 1 \) throughout. In Table SI we have listed the parameters that were used in each figure, insofar as those are not mentioned in the main text. The density distributions that we used for averaging are based on the atom numbers, temperatures, and trap frequencies mentioned.

<table>
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<th>State</th>
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<th>( \gamma_0/2\pi ) (10^6 s(^{-1}))</th>
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</tr>
<tr>
<td>Fig. 2</td>
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</tr>
<tr>
<td></td>
<td>30S(_{1/2})</td>
<td>40</td>
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<tr>
<td>Fig. 3(a)</td>
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<tr>
<td>Fig. 3(b)</td>
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<td>Lifetime (( \mu s ))</td>
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<td>( C_b / \hbar ) (MHz \mu m(^6))</td>
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<td>-25.0</td>
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Table SI. Overview of Rydberg state parameters as used in simulations. The \( C_b \) coefficient of the 28D\(_{5/2}\) is anistropic, we have tabulated the root-mean-square value after calculating it for different angles.

### Rydberg state parameters

Table SII contains the relevant experimental parameters of the Rydberg states that we have investigated. All values are calculated using the Alkali Rydberg Calculator (ARC) [40]; the interaction coefficients are verified using PairInteraction [41]. These open-source packages were also used in combination with our own calculation [33] to calculate the dipole matrix elements and validate the Rabi frequencies in Table SII. Additionally, these were verified using an electromagnetically induced transparency (EIT) measurement.

Black-body photons from the environment cause enhanced decay towards nearby Rydberg states, leading to a reduction in lifetime [39]. This was accounted for in the calculated values, assuming an environment at \( T = 300 \) K.