Supplemental material: Observation of Interactions between Trapped Ions and Ultracold Rydberg Atoms

N. V. Ewald,[1] T. Feldker,[1] H. Hirzler,[1] H. A. Fürst,[1] and R. Gerritsma[1]

[1] *Van der Waals-Zeeman Institute, Institute of Physics, University of Amsterdam, 1098 XH Amsterdam, Netherlands*

(Dated: May 13, 2019)

CALCULATION OF LOSS SPECTRA

Atoms

To simulate the atom loss spectrum we sample atom starting positions from the atomic density distribution for random times and calculate the Rydberg excitation rate taking all electric fields into account. Atom positions are sampled from a 3D Gaussian distribution of widths $25 \times 25 \times 200\,\mu\text{m}^3$ and the starting time relative to the rf-phase of the Paul trap from the interval $[0,\ 2\pi/\Omega_{\text{rf}}]$ for 5×10^7 atoms. The spectral density of the atomic distribution is calculated by making a histogram of Stark shifts, calculated from the electric fields at the starting location and time of the atoms, with appropriate bin size.

The electric field at the atom position $\vec{r}_{\text{a}} = (x_{\text{a}}, y_{\text{a}}, z_{\text{a}})^{\text{T}}$ contains a contribution from the Paul trap, $\vec{E}_{\text{rf,a}}$ and one from the ion $\vec{E}_{\text{i,a}}$ at position $\vec{r}_{\text{i}} = (x_{\text{i}}, y_{\text{i}}, z_{\text{i}})^{\text{T}}$:

$$\vec{E}_{\text{rf,a}} = \frac{m_{\text{i}}}{2e}\begin{pmatrix}(-\omega_z^2 + q\Omega_{\text{rf}}^2\cos(\Omega_{\text{rf}}t))\ x_{\text{a}}\\ (-\omega_z^2 - q\Omega_{\text{rf}}^2\cos(\Omega_{\text{rf}}t))\ y_{\text{a}}\\ 2\omega_z^2\ z_{\text{a}}\end{pmatrix}, \text{and} \tag{1}$$

$$\vec{E}_{\text{i,a}} = \frac{e}{4\pi\epsilon_0 r_{\text{ia}}^3}\begin{pmatrix}x_{\text{a}} - x_{\text{i}}\\ y_{\text{a}} - y_{\text{i}}\\ z_{\text{a}} - z_{\text{i}}\end{pmatrix}, \tag{2}$$

where m_{i} is the ion mass, r_{ia} the distance between atom and ion, Ω_{rf} the trap-drive frequency, q the dynamic stability parameter and ω_z the axial trap frequency. The Stark shift experienced by the Rydberg atom is given by:

$$U_{\text{tot,a}} = -\frac{1}{2}\alpha_{\text{Ryd}}(\vec{E}_{\text{rf,a}} + \vec{E}_{\text{i,a}})^2, \tag{3}$$

where α_{Ryd} is the polarizability of the Rydberg state, and $\alpha_{24S} = 313\,\text{Hz}/(\text{V/m})^2$.

Ions

The ion loss spectrum requires a full dynamical simulation to model the ion loss induced by Rydberg atom-ion collisions. We assume that the ion is positioned at the trap center (justified later). We dice a random atom position and time as for the calculation of the atom loss spectrum. The initial velocity of the atom is sampled from a thermal distribution with $T = 10\,\mu\text{K}$. To model a collision, we simulate the dynamics of both atom and ion by solving Newton's equations. The force acting on the atom is given by:

$$\vec{F}_{\text{tot,a}} = -h\nabla_{\text{a}}U_{\text{tot,a}}, \tag{4}$$

where ∇_{a} denotes the gradient with respect to the atomic coordinates and h is Planck's constant. The ion experiences both the force due to the Paul trap $\vec{F}_{\text{rf,i}} = e\vec{E}_{\text{rf,i}}$ and the interaction force with the polarized Rydberg atom $\vec{F}_{\text{a,i}} = -h\nabla_{\text{i}}U_{\text{tot,a}}$, which results in:

$$\vec{F}_{\text{tot,i}} = \vec{F}_{\text{a,i}} + \vec{F}_{\text{rf,i}}. \tag{5}$$

Atom and ion dynamics are propagated using an adaptive step-size Runge-Kutta algorithm of fourth order [1]. This allows for fast propagation when the atom-ion distance is large and for slow and accurate propagation for small distances (when the forces become large). The simulation stops either if the ion-atom distance drops below a minimum distance $r_{\text{ia}} < r_{\text{min}}$, if the collision takes longer than $t_{\text{stop}} = 180\,\mu\text{s}$, or if the atoms leaves the interaction region $r_{\text{ia}} > r_{\text{escape}} = 10.5\,\mu\text{m}$ without colliding (glancing collisions). We set the minimum distance to $r_{\text{min}} = 200\,\text{nm}$, roughly corresponding to the distance where the potential barrier between the atom and ion opens for the $24S$ state [2]. We simulate 10^7 events for each loss spectrum. More details on our simulation methods can be found in [1, 3].

To create an ion loss spectrum, we calculate the occurrence of collisions for each initial Stark shift, resulting from the atoms' starting conditions. We choose an appropriate frequency bin to create a histogram of collision probabilities, weighted with the probability that the Rydberg state has not decayed at collision time t_{col}, yet:

$$P_{\text{Ryd}} = e^{-t_{\text{col}}/\tau_{24S}}. \tag{6}$$

Here, $\tau_{24S} \approx 11\,\mu\text{s}$ is the lifetime which we calculated using Ref. [4]. Fig. 1 shows initial Stark shifts for initial distances from the trap center r_{ia} for 8468 atoms that lead to ion loss (color dots), where P_{Ryd} is presented in the color code. Naturally, P_{Ryd} is largest for atoms sampled close to the ion and decreases for larger r_{ia} until the initial distance to the ion gets too large to be bridged within the Rydberg state's lifetime. Note that as explained in the main text and therein depicted in Fig. 2a, atoms excited outside the region dominated by the elec-

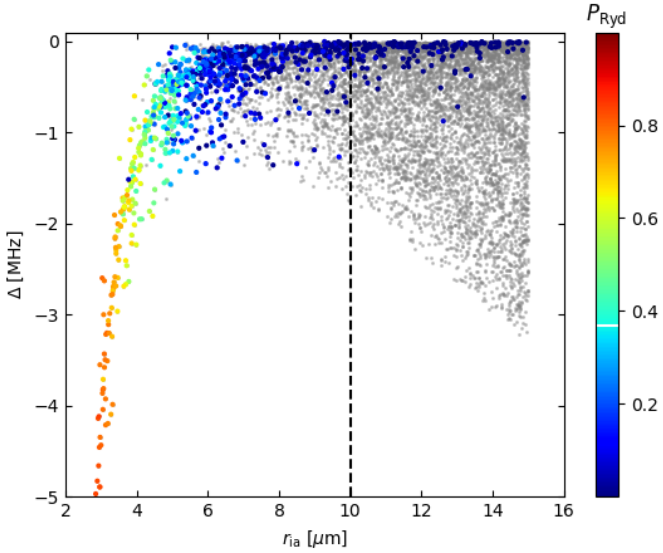

FIG. 1. Rydberg atom-ion collision probability for the $24S$ state for different two-photon detunings Δ and starting distances, thus initial Stark shifts, from the trap center r_{ia}. Atoms that approach the ion to within a distance of $0.2\,\mu$m are color-coded with the probability P_{Ryd} that the atom is still in the Rydberg state when the collision occurs (color bar legend). For larger initial distances, more and more atoms do not collide with the ion (gray).

tric field of the ion, will be accelerated away from the ion by the trap. We therefore restrict the atom sampling to a homogeneous sphere of radius $r_{0,\mathrm{max}} = 10\,\mu$m (vertical dashed line), consequently reducing the computational effort. Rydberg atoms created further away from the ion in the trap-dominated region thus do not contribute to the ion loss signal, but to the atom loss signal only. Here, due to the linear increase of the trap's electric field amplitude with distance, the maximal Stark shift at the excitation position increases linear with distance, seen as a negative slope giving a lower bound to the gray sampling dots in Fig. 1.

$24P$ State Simulation

For the calculation of the P state spectra, we follow a similar procedure. To account for the Stark admixing of the $24D$ state, we scale the probability of Rydberg excitation accordingly. Since the state admixing of the $24D$ state is to first order linear in the electric field experienced by the Rydberg atom, we scale the probability of exciting the Rydberg with the local electric field squared. Note that the polarizability $\alpha_{24P} = 2470\,\mathrm{Hz/(V/m)^2}$ and lifetime $\tau_{24P} \approx 22\,\mu$s are different from those of $24S$. To account for these differences, $r_{0,\mathrm{max}}$ and r_{escape} were increased to $20\,\mu$m and $20.5\,\mu$m, respectively, for the $24P$ state simulations.

Parameter	Value	Comment
ω_z	$2\pi \times 27\,\mathrm{kHz}$	axial trap frequency
Ω_{rf}	$2\pi \times 1\,\mathrm{MHz}$	trap-drive frequency
q	$\in [0, 0.4]$	stability parameter
T_{a}	$10\,\mu$K	atom bath temperature
$r_{0,\mathrm{max}}$	$10\,\mu$m	atom launch sphere rad.
r_{escape}	$10.5\,\mu$m	atom escape sphere rad.
t_{stop}	$180\,\mu$s	max. simulation time
r_{min}	$200\,\mathrm{nm}$	collision distance
α_{24S}	$313\,\mathrm{Hz/(V/m)^2}$	polarizability of $24S$
α_{24P}	$2470\,\mathrm{Hz/(V/m)^2}$	polarizability of $24P$

TABLE I. Parameters used for the numerical simulations of the Rydberg atom-ion collisions.

Fitting Procedure

Rydberg excitation suffers several mechanisms of line broadening caused by e.g. magnetic fields, interactions, finite temperature, etc. The symmetrically broadened Rydberg spectrum for $q = 0$ is fitted by a Gaussian $g_{\mathrm{G}}(\Delta)$ of FWHM $= 6.1\,\mathrm{MHz}$ ($\sigma = 2.6\,\mathrm{MHz}$ in the computation).

To account for broadening effects in our simulation, including the Paul trap for $q \neq 0$, we convolve the modelled atom and ion spectra $f_{\mathrm{mdl}}(\Delta)$ with a Gaussian of the extracted width (see Fig. 2 of the main text). Furthermore, a saturation parameter s is included to take into account atom densities and two-photon Rabi frequencies of the Rydberg transition. We calculate:

$$f_s(\Delta) = \frac{1}{2}\int_{-\infty}^{\Delta} (1 - e^{-s\cdot f_{\mathrm{mdl}}(\Delta')})\; g_{\mathrm{G}}(\Delta' - \Delta)\; \mathrm{d}\Delta', \quad (7)$$

for a set $\{s_1, ..., s_{\mathrm{n}}\}$ and interpolate the resulting $f_{s_1}(\Delta), ..., f_{s_{\mathrm{n}}}(\Delta)$ to obtain a two-dimensional function, dependent on s and Δ. This function is fitted to the measured data, with fit parameters s and a frequency offset Δ_0. Note that we assume that only a single collision can take place, as described in the main text. Table I summarizes the parameters used in the simulations.

EXPERIMENTAL CHECKS

Several mechanisms limit the region around the ion where Rydberg excitation can result in a collision and charge transfer. First, the finite lifetime of the Rydberg

atom limits the distance it can travel. Secondly, the electric fields of the Paul trap cause Rydberg atoms that are created far away from the ion to be accelerated away. On the other hand, motional excitation of the ions complicates matters. In particular, collisions with background gas molecules can cause the ions to enter highly excited motional states where they can sample large electric field regions in the trap. To check whether such events play a role in our experiment, we observe the ion's fluorescence signal right before the Rydberg excitation. In this way, we eliminate from the data presented in Fig. 4 in the main text instances where the ions are hit by a background gas molecule and sent into a large orbit during the preparation of the atom cloud. Voltage ramps are used to transport the ions in and out of the cloud such that fluorescence detection can be performed without risking the loss of ions due to collisions. The axial transport distance is about 400 μm, well outside the atom cloud. Observing the ions' fluorescence directly after interaction with the ultracold ground state atoms indicates that the presence of the atom cloud does not lead to detectable heating.

Independent measurements with isotope-labelled crystals containing one dark ion of another isotope show that the ion crystals do not melt when interacting with ultracold ground state atoms nor during transport. Note that under typical Paul trap operation ($q = 0.27$), a two-ion crystal melts if the ions have a radial oscillation amplitude beyond 12 μm, well within the atom cloud.

These measurements justify our assumption that the ions are located near the trap center when Rydberg excitation happens and do not venture into regions of large electric fields within the trap.

REFERENCES

[1] H. Fürst, N. V. Ewald, T. Secker, J. Joger, T. Feldker, and R. Gerritsma, Journal of Physics B: Atomic, Molecular and Optical Physics **51**, 195001 (2018).

[2] V. N. Ostrovsky, J. Phys. B: At. Mol. Opt. Phys. **28**, 3901 (1995).

[3] H. A. Fürst, *Trapped ions in a bath of ultracold atoms*, Ph.D. thesis (2018).

[4] N. Šibalić, J. D. Pritchard, C. S. Adams, and K. J. Weatherill, Computer Physics Communications **220**, 319 (2017).