Supplementary material: Anomalous broadening in driven dissipative Rydberg systems

E. A. Goldschmidt,[1] T. Boulier,[2] R. C. Brown,[2, *] S. B. Koller,[2, †]
J. T. Young,[2] A. V. Gorshkov,[2, 3] S. L. Rolston,[2] and J. V. Porto[2]

[1] *United States Army Research Laboratory, Adelphi, Maryland 20783 USA*
[2] *Joint Quantum Institute, National Institute of Standards and Technology*
and the University of Maryland, Gaithersburg, Maryland 20899 USA
[3] *Joint Center for Quantum Information and Computer Science,*
National Institute of Standards and Technology and the University of Maryland, College Park, Maryland 20742 USA

RABI FREQUENCY CALIBRATION

We calibrate the Rabi frequencies of the two optical fields using the light shifts due to each field. For the 795-nm laser, which is detuned $\Delta/2\pi \approx 235$ MHz below the $|5s; F = 2, m_F = -2\rangle \rightarrow |5p_{1/2}; F = 1, m_F = -1\rangle$ transition, we observe the intensity dependent shift, δ_{795}, of the $|5s; F = 2, m_F = -2\rangle \rightarrow |5s; F = 1, m_F = -1\rangle$ microwave transition. The light shift $\delta_{795} = \Omega_1^2/4\Delta$ is a factor of $\Delta_{\text{hyperfine}}/\Delta \approx 30$ larger for the $|5s; F = 2, m_F = -2\rangle$ state compared to the $|5s; F = 1, m_F = -1\rangle$ state so we take the effect on the microwave transition as the light shift on the $|5s; F = 2, m_F = -2\rangle$ state. We observe a shift that is linear in intensity I, as expected, and use this to calibrate the Rabi frequency $\Omega_1(I) = 2\sqrt{\delta_{795}(I)\Delta}$ which we vary from $2\pi \times 0.2$ MHz to $2\pi \times 10$ MHz to control the two-photon Rabi frequency.

To obtain the Rabi frequency of the 485-nm field, detuned $\Delta/2\pi \approx 235$ MHz above the $|5p_{1/2}; F = 1, m_F = -1\rangle \rightarrow |18s; F = 2, m_F = -2\rangle$ transition, we observe the intensity dependent shift, δ_{485}, of the two-photon $|5s; F = 2, m_F = -2\rangle \rightarrow |18s; F = 2, m_F = -2\rangle$ transition keeping the 795-nm light intensity (and frequency) constant. The 485-nm field is far from any transition coupling the ground $5s$ state to any optically excited state, so the shift of the $|18s; F = 2, m_F = -2\rangle$ state is the dominant contribution and $\delta_{485} = \Omega_2^2/4\Delta$. We observe a shift that is linear in intensity, as expected, and use this to calibrate the Rabi frequency $\Omega_2(I) = 2\sqrt{\delta_{485}(I)\Delta}$ which we hold at the maximum value given available laser power and beam diameter, $\Omega_2/2\pi \approx 7$ MHz.

LATTICE FILLING

Loading 4×10^4 atoms into the 3D optical lattice as described in the main text leads to overfilling (more than one atom per site) in a fraction of the lattice sites. However, we determined that the filling fraction does not affect the broadening, which is only determined by the overall density. We made this determination by comparing a Mott insulator state with no more than one atom per site to a state with a Poissonian distribution of atoms per site, but the same total number of atoms in the participating ground state (and thus the same global density). Only the overall atom number was pertinent to the broadening. We also compared random transfer of half of the atoms to the participating ground state with transfer of all atoms on every other site in 2D in a checkerboard fashion [34]. These two cases lead to a factor of two difference in the filling fraction per lattice site. We again saw that the total atom number controlled the broadening without regard to the microscopic configuration.

18s LIFETIME

We observe 780-nm fluorescence on the $5p_{3/2} - 5s$ transition following excitation to the 18s Rydberg state and fit a decaying exponential to extract a lifetime. The bulk of the fluorescence is due to the ≈ 30 % of the decay via $18s - 5p_{3/2} - 5s$ for which the 28-ns lifetime of the $5p_{3/2}$ is negligible compared to $\tau_0 = 3.5$ μs, the natural lifetime of the 18s. However, ≈ 1 % of the population decays via channels of the form $18s - np - n's - 5p_{3/2} - 5s$ for which the lifetimes of the intermediate states delay the final 780-nm photon leading to effective lifetimes $\approx 4\times$ longer [26] and an increase in the measured lifetime by ≈ 10 %. In addition, radiation trapping, in which photons are reabsorbed and reemitted one or more times before leaving the cloud, can alter the measured lifetime. By measuring fluorescence following resonant excitation on the $5s_{1/2} - 5p_{3/2}$ transition, we place a limit of < 0.5 μs, shorter than the measured lifetimes, on the characteristic time for radiation trapping, and thus expect little or no alteration of the measured lifetime due to this effect.

In all cases studied, including resonant excitation at different two-photon Rabi frequencies and detuned excitation at different detunings, the extracted lifetime is 8 % to 20 % longer than τ_0, consistent with the natural lifetime and suggesting no shortening of the lifetime due to superradiance or other purely homogeneous effects. In addition, the amount of fluorescence is consistent with the optical pumping signal under all different conditions. This further suggests that superradiance is not a significant effect as it would lead to a Rydberg density depen-

dence of the fractional decay via the $5p_{3/2}$ state compared to other intermediate states.

DIPOLE VS VAN DER WAALS SCALING

In order to calculate β_3, we have only included the $17p$ and $18p$ states and have excluded interactions that do not conserve total magnetic quantum number. The former should add $\approx 1\ \%$ to the total interaction and the latter are not resonant due to Zeeman splitting caused by a non-zero magnetic field. However some m non-conserving interactions have an energy mismatch that is less than the largest observed linewidth and may play a role.

If the broadening were due to van der Waals interactions between $18s$ atoms, one expects scaling $\Gamma = C_6\rho_{18s}^2$. We define an interaction volume $\beta_6 = \sqrt{C_6/\Gamma_0}$, in which case we assume $\Gamma = \beta_6^2\rho_g^2 R_0^2/\Gamma_0$. Combined with the observed relation $R_0 = \Omega^2/\Gamma$, we express Γ and R_0 in terms of the independently controlled variables ρ_g and Ω:

$$
\begin{aligned}
\Gamma &= \left(\Omega^4/\Gamma_0\right)^{1/3}\ \left(\rho_g\beta_6\right)^{2/3} \\
R_0 &= \left(\Omega^2\Gamma_0\right)^{1/3}\ \left(\rho_g\beta_6\right)^{-2/3}.
\end{aligned}
\tag{S1}
$$

Figure S1 shows Γ and R_0 in terms of these expressions. There is a large mismatch in values between the data and the van der Waals scaling. In addition the data is not linear in the expressions and the rate R_0 does not collapse to a single function for all Ω and ρ_g.

DIPOLE BROADENING IN VARIOUS REGIMES

We consider how broadening due to dipole interaction with spontaneously created atoms in nearby Rydberg states affects a variety of possible operating regimes. For systems with small atom number, the time until the first atom is created in a nearby Rydberg state can be long compared to the lifetime if $bN_0 \ll 4\delta^2/\Omega^2$. However, since the dressed Rydberg interaction is of order Ω^4/δ^3 it may be difficult to access interesting many-body effects in this regime.

Additionally, this effect remains at higher principal quantum number: $\beta_3 \propto n^{*7}$ where $n^* = n - \delta_{qd}$ is the effective principal quantum number and δ_{qd} is the quantum defect [41]. The contaminant density at which the dipole interaction overtakes the dressed interaction $(\Omega^2/\delta C_3)$ scales as n^{*-4}. Assuming reasonable limits on attainable Rabi frequency $(\Omega/2\pi < 100\text{ MHz})$ and dressed interaction strength $(\Omega^4/\delta^3 < 2\pi \times 10\text{ kHz})$, this corresponds to a single spontaneous Rydberg atom in a typical 10-μm diameter ultracold atom system for $n > 40$. Systems at higher principal quantum number may allow some additional time in an absolute sense due to their longer lifetimes (which scale as $< n^{*3}$), but are still subject to
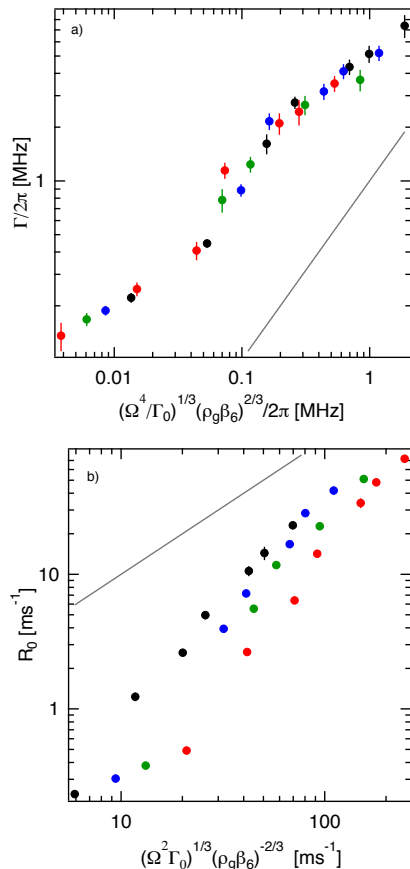

FIG. S1. (a) Measured Γ and (b) R_0 for different Ω and ρ_g vs expected scaling for van der Waals dominated interaction. Linear scalings with unit slope are indicated with solid lines.

this effect and cannot access the expected single-atom scattering rates.

Since the van der Waals interaction is derived from the dipole interaction with nearby states, any configuration with large van der Waals interaction must have dipole interactions at least as large. Even in the case of operation at a Förster resonance to maximize the interaction [11, 14], the uncontrolled, spontaneously created population in nearby Rydberg states causes interactions larger than any dressed interaction and must be considered in determining the timescales available for coherent operations.

INHOMOGENEOUS GUTZWILLER MEAN-FIELD THEORY

Treating each atom as a three level system with states g, p, and s, corresponding to ground, $18s$, and np states respectively, we model the system with the following

Hamiltonian:

$$H = -\delta \sum_i \sigma_i^{ss} + \Omega \sum_i (\sigma_i^{sg} + \sigma_i^{gs}) + \sum_{ij} V_{ij} \sigma_i^{sp} \sigma_j^{ps} + h.c.,$$
(S2)

where $V_{ij} = \frac{C_3}{r_{ij}^3}(1 - 3\cos^2\theta)$ is the dipole-dipole interaction with θ the angle between the z-axis and the relative position $\mathbf{r}_{ij}$. The system evolves according to the master equation

$$\dot\rho = -i[H, \rho] + \mathcal{L}_s + \mathcal{L}_p + \mathcal{L}_R,$$
(S3a)

$$\mathcal{L}_s = \Gamma_s \sum_i \left(\sigma_i^{gs} \rho \sigma_i^{sg} - \frac{1}{2}\{\sigma_i^{ss}, \rho\} \right),$$
(S3b)

$$\mathcal{L}_p = \Gamma_p \sum_i \left(\sigma_i^{gp} \rho \sigma_i^{pg} - \frac{1}{2}\{\sigma_i^{pp}, \rho\} \right),$$
(S3c)

$$\mathcal{L}_R = \Gamma_R \sum_i \left(\sigma_i^{ps} \rho \sigma_i^{sp} - \frac{1}{2}\{\sigma_i^{ss}, \rho\} \right),$$
(S3d)

where $\mathcal{L}_s$, $\mathcal{L}_p$, and $\mathcal{L}_R$ are Lindblad terms corresponding to decay from s to g, from p to g, and from s to p respectively.

Using an inhomogeneous Gutzwiller mean-field approximation, we assume the density matrix has the form

$$\rho = \bigotimes_i \rho_i,$$
(S4)

which assumes there are no correlations between different atoms. The method is inhomogeneous in the sense that each atom has its own density matrix, whereas in homogeneous Gutzwiller mean-field theory all atoms have the same density matrix. This results in an effective local Hamiltonian

$$H_i^{eff} = -\delta\sigma_i^{ss} + \frac{\Omega}{2}(\sigma_i^{sg} + \sigma_i^{gs}) + \sum_j V_{ij}\sigma_i^{sp}\langle\sigma_j^{ps}\rangle + h.c.$$
(S5)

In this picture, the interactions behave as an effective driving term between the s and p states whose strength and phase are determined by the $\langle\sigma^{ps}\rangle$ coherences of the surrounding atoms.

We determine the steady state of these equations numerically by initializing a cubic lattice of randomized density matrices for each site and evolving the system according to the master equation and effective Hamiltonian. This was done for a variety of Ω and δ based on the experimental measurements. The interaction strengths were reduced due to computational constraints, but the nearest neighbor interaction strength remained at least two orders of magnitude above Ω and all decay rates, compared to four in the experiment.

In all cases, we find that the $\langle\sigma^{ps}\rangle$ coherences all decay to zero in steady state, in which case the system behaves as if there are no interactions. This is a consequence of the flip-flop interactions and would not occur if the interactions were of the form

$$\sum_{ij} V_{ij}\sigma_i^{ss}\sigma_j^{pp}.$$
(S6)

Collective decay between the s and p states does result in nonzero $\langle\sigma^{ps}\rangle$ coherences in steady state, but the effect of interactions in this case is small and the experimental results indicate that collective decay is not the source of the observed broadening.

Future work to solve this model using techniques beyond Gutzwiller mean-field will hopefully provide a more full understanding of the observed broadening and inform a more detailed study of the early time dynamics of the system.

––––––––––

* Present address: National Institute of Standards and Technology, Boulder, Colorado 80305 USA
† Present address: Physikalisch Technische Bundesanstalt, 38116 Braunschweig, Germany