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SUPPLEMENTARY INFORMATION
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Mesoscopic Rydberg-blockaded ensembles in the superatom regime and beyond

I. BENCHMARKING OF RATE EQUATION SIMULATIONS

To describe the complex many-body dynamics of strongly interacting Rydberg atoms, often a classical rate equation description is used [1–4]. Intuitively, such an approach is valid in the presence of strong decoherence and a formal derivation can be found, for example, in [5].

We here present results for a model system that exhibits important aspects of the superatom physics, but is so small, that the full quantum dynamics can be simulated. The model is displayed in Fig.1(a) and is comprised of two very small clusters of N atoms each. The size r of the individual clusters is small, such that the Rydberg-Rydberg interaction within the cluster strictly suppresses multiple excitations. On the other hand, the distance between the two ensembles is much larger ($l \gg r$) and therefore the interaction U between excitations in different clusters is finite. Atoms are excited with Rabi frequency Ω and subject to spontaneous decay Γ and decoherence with rate Γ_d. Exact simulations are feasible in the small Hilbert space of dimension $(1+N)^2$.

In Fig.1(b) we show results for the mean number of Rydberg excitations $\langle N_\mathrm{r}\rangle$ and their statistics $g_2(0)$ as a function of the driving Ω in the stationary state. At strong dephasing of $\Gamma_\mathrm{d}/2\pi = 140$ kHz (red), rate equation (full line) and exact results (squares) agree within small errors. In the limit of weak driving we calibrate the rate equation model to the experiment. For intermediate values of Ω we find a pronounced plateau indicating the blockade mechanism, where at most one of the two clusters is excited. Finally, for large drivings, the blockade breaks down and the number of excitations increases again. In absence of decoherence (black) the rate equations yield incorrect results for both the number of excitations and their statistics.

For off resonant excitation, the mesoscopic superatom and the model system exhibit antiblockade and pronounced bunching of Rydberg excitations occurs. We have choosen the detuning such that it exactly cancels the interaction $\Delta_0 = -U$ and the second excitation process is resonant. Both, statistics and excitation number are again well reproduced by the rate equations in the presence of decoherence across the range of driving Rabi frequencies, but the classical ansatz fails in absence of decoherence.

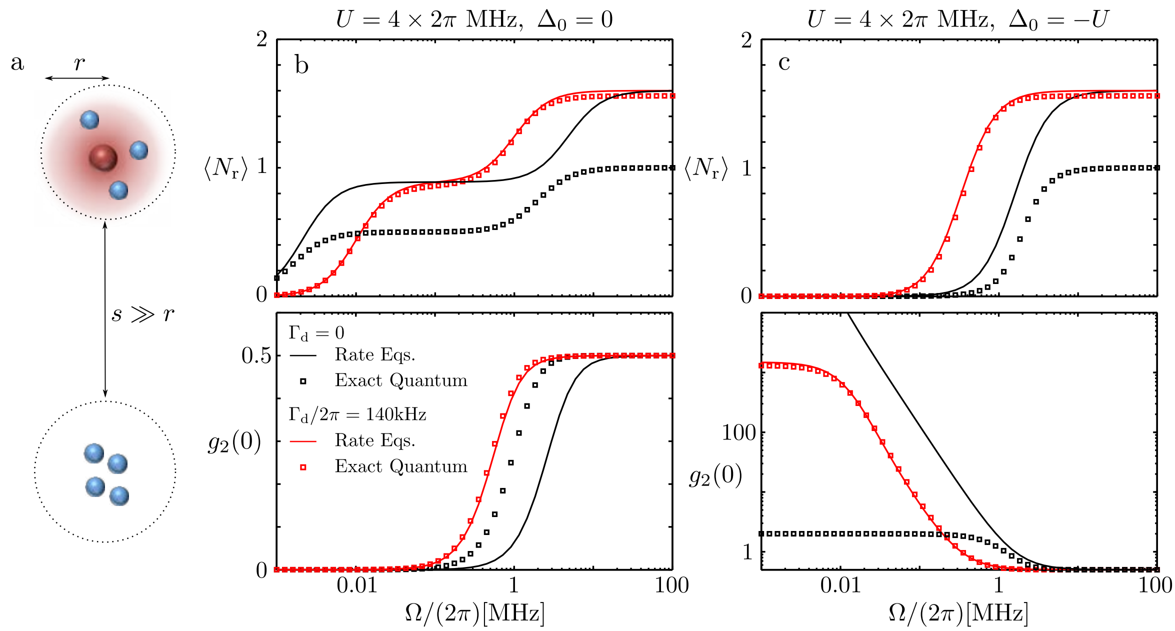

FIG. 1: (a) Model system for validating the rate equation simulations with cluster of 4 atoms. (b) Mean number of Rydberg excited atoms and two excitation correlations for resonant excitation with decoherence $\Gamma_\mathrm{d}/2\pi = 140$ kHz (red) and $\Gamma_\mathrm{d} = 0$ (black). Solid lines are results of the rate equation model and exact quantum simulation results are displayed as squares. (c) Results for off resonant excitation with $\Delta_0 = -U$, $U = 4 \times 2\pi$ MHz, symbols as in (b).

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A comprehensive comparison of rate equation results and full quantum simulations can be found in [6]. For a recent experiment, well described by rate equation simulations see [4].

II. COHERENT DYNAMICS

As shown in the previous section, the long time excitation statistics is well described by rate equation simulations. However on short time scales the number of excited atoms is more sensitive to coherent effects as shown in Fig.2. Parameters are choosen as in Fig.1(a) with driving $\Omega/2\pi = 270$ kHz, such that $g_2(0) = 0.1$ and we are in the steady-state blockade regime. The initial dynamics reveals coherent, collective Rabi oscillations, which are damped by the decoherence. In the experiment, the blockade conditions break down for such large values of the Rabi frequency, and coherent dynamics cannot be observed. Choosing a higher lying Rydberg states can preserve the blockade conditions even for large values of Ω. However, care has to be taken in order to keep the decoherence rate small.

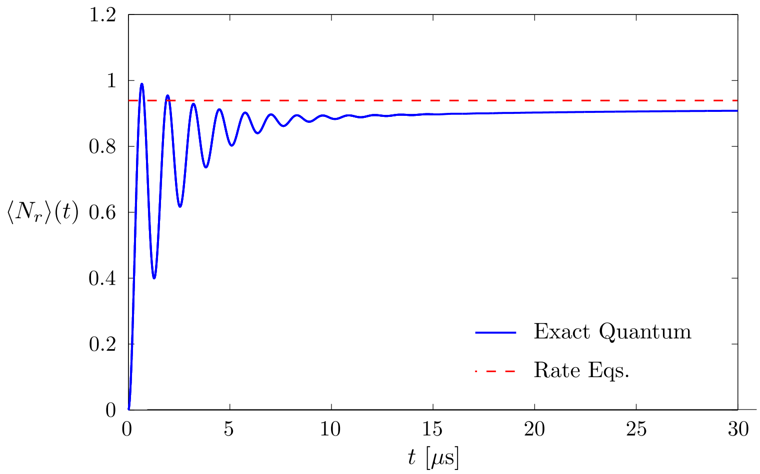

FIG. 2: Expectation value of Rydberg excitations as a function of time for the model system depicted in Fig. 1. For a decoherence rate of $\Gamma_d/2\pi = 140$ kHz, interaction $U/2\pi = 4$ MHz and $\Omega/2\pi = 270$ kHz the full quantum simulation (blue) reveals damped coherent dynamics. At longer times, exact results converge towards the rate equation model results (dashed red) in the stationary state.

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