

FIG. 6. Influence of the density and the blockade radius on the DTWA simulations. Left column: Simulations for the same blockade radius as in the main text for different particle numbers *N*. Right column: Simulations for the same particle number and various blockade radii. In all simulations the geometry of the cloud is the same.

two microwave photons at $2\pi \times 16\,\text{GHz}$ couple to $|\uparrow\rangle$ = $|62S_{1/2}, m_i = 1/2\rangle$. Here, a single photon Rabi frequency of $\Omega = 2\pi \times 48 \, \text{MHz}$ with a detuning $\Delta_{\nu} = 2\pi \times 170 \, \text{MHz}$ leads to a two-photon Rabi frequency of $\Omega_{2\gamma} = 2\pi \times$ 6.8 MHz. To realize the Ising model, the state $|61S\rangle$ has to be coupled to $|64S_{1/2}, m_i = 0.5\rangle$ but the detuning of $\Delta_{\nu} =$ $2\pi \times 1.426\,\mathrm{GHz}$ is too large and prevents an efficient coupling of the states with two microwave photons of the same frequency $2\pi \times 47 \,\text{GHz}$. Therefore, we combine two frequencies differing by $2\pi \times 1.563$ GHz such that the effective detuning to the intermediate state $|62P\rangle$ is $2\pi \times 136\,\mathrm{MHz}$. For a single photon Rabi frequency of $\Omega = 2\pi \times 30 \,\mathrm{MHz}$ this results in an effective two-photon Rabi frequency of $\Omega_{2\nu} = 2\pi \times 3.3 \,\mathrm{MHz}$ [see Figs. 1(b)–1(d) for the microwave photonic transitions]. The parameters of the laser waists and resulting geometries can be found in Table I.

APPENDIX C: DISTRIBUTION OF INTERACTION TIMESCALES IN THE SPIN SYSTEM

In the main text, we have highlighted that the typical timescale of the relaxation is given by the pair oscillation frequency $|J_{\parallel}-J_{\perp}|$. For the Heisenberg XXZ Hamiltonian, both exchange and Ising interactions exist. Therefore, another

possibility of rescaling would only involve J_{\perp} , which would disregard the anisotropy $\delta = J_{\parallel}/J_{\perp}$. In Fig. 5, we have compared both possibilities of rescaling time. The rescaling by the oscillation frequency shows a more precise collapse of the experimental data. This demonstrates that this frequency indeed determines the relevant timescale of the system. In addition, this indicates that the Rydberg interactions can be mapped onto the Heisenberg XXZ Hamiltonian with $\delta = -0.7$.

In Fig. 6, we show the sensitivity of the DTWA simulations to different densities and blockade radii. For most simulations, these parameters have only a small, quantitative effect on the simulated dynamics. A notable exception is the Ising system. Here, the Rydberg cloud is largely saturated and the blockade radius is the relevant length scale of the system. Therefore, a variation of the blockade radius changes drastically the early time dynamics. In contrast, the density of the sample featuring XX-interaction is low, therefore the blockade effect can be neglected. For the Heisenberg XXZ Hamiltonian, the simulations show that the blockade radius of 8.3 µm fits the observed dynamics slightly better than the value of 10 µm expected from the simplified excitation model assuming no phase noise of the laser.

Histograms showing the resulting distribution functions of couplings are shown in Fig. 7.