Spatially tunable spin interactions in neutral atom arrays

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Analog quantum simulations with Rydberg atoms in optical tweezers routinely address strongly correlated many-body problems due to the hardware-efficient implementation of the Hamiltonian. Yet, their generality is limited, and flexible Hamiltonian-design techniques are needed to widen the scope of these simulators. Here we report on the realization of spatially tunable interactions for XYZ models implemented by two-color near-resonant coupling to Rydberg pair states. Our results demonstrate the unique opportunities of Rydberg dressing for Hamiltonian design in analog quantum simulators.

Quantum simulators based on arrays of neutral atoms have proven to be among the most promising platforms to address non-trivial problems of strongly correlated many-body phenomena. This success is based on the optimally hardware-efficient analog implementation of the Hamiltonian under study [1–4], which is one of the reasons to employ these machines for useful tasks in the so-called NISQ (noisy intermediate-scale quantum) era of quantum processors. While such an emulation approach eliminates any overhead in control necessities or qubit numbers, it strongly restricts the use cases of a specific quantum simulator to problems rooted in the device-dependent Hamiltonian. Here, neutral atoms trapped in optical tweezer arrays with engineered geometries in two dimensions, laser coupled to Rydberg states to induce interactions [5], are among the most promising platforms [6–12]. Large system sizes have been demonstrated with long coherence times [12, 13], enabling the simulation of quantum magnets both in equilibrium [14, 15] and dynamically [16].

Rydberg atom-based simulators naturally implement Ising or XY-type Hamiltonians with power-law interactions [17–20]. Recently, Floquet-engineered XXZ spin coupling in bulk systems and optical tweezer arrays has been demonstrated [21, 22]. Control over the spatial interaction profile of Ising systems has also been realized by admixing Rydberg character to the ground state, so-called Rydberg dressing [23–30]. Lately, a sharply peaked interaction profile has been demonstrated by coupling to molecular Rydberg macrodimer potentials [31]. One of the biggest remaining challenges is to increase the systems’ flexibility via universally programmable analog qubit couplings.

We report on progress into this direction by the realization of freely tunable short-range XYZ-type spin interactions between atoms trapped in optical tweezer arrays. The effective spin-1/2 system is encoded in two electronic ground states and we introduce interactions by two-color Rydberg dressing. This allows to engineer the spin-spin couplings in each spin direction by the choice of the laser parameters. Our approach uses the spatially dependent van-der-Waals (vdW) interactions between different m_j-sublevels in the Rydberg pair state manifold to design distance and angular-dependent couplings of the XYZ Hamiltonian [33]

\[
\hat{H}_{\text{XYZ}} = \hbar \sum_{i<j}(J_{ij}^z \hat{\sigma}_i^z \hat{\sigma}_j^z + J_{ij}^{++} \hat{\sigma}_i^+ \hat{\sigma}_j^+ + J_{ij}^{+-} \hat{\sigma}_i^\pm \hat{\sigma}_j^\mp + \text{h.c.})
\]

(1)

The Pauli matrices \(\hat{\sigma}_i^z\), \(\hat{\sigma}_i^y\) = (\(\hat{\sigma}_i^x\) + \(\hat{\sigma}_i^y\)) and \(\hat{\sigma}_i^y = i(\hat{\sigma}_i^- - \hat{\sigma}_i^+)\) describe a spin-1/2 particle at position \(j\). This Hamiltonian distinguishes between three types of spin couplings \(J_{ij}^{\alpha}\): The diagonal interaction between dressed ground states \(J_{ij}^z\), the off-diagonal “flip-flop” \(J_{ij}^{++}\), and “flip-flop” \(J_{ij}^{+-}\) interactions. While dressing-induced Ising (\(J^z\)) interactions have already been studied in various experiments [26, 27, 30, 31, 34–36] and programmable long-range interactions have been demonstrated in optical cavities [37], we focus on programmable \(J_{ij}^{++}\) and \(J_{ij}^{+-}\) interactions (see fig. 1). With control over the laser parameters and the geometric arrangement of single atoms, we can engineer the relative coupling strength of the spin-spin interactions \(J_{ij}^{++}/J_{ij}^z\) and \(J_{ij}^{+-}/J_{ij}^z\) as visualized exemplarily in fig. 2. We are also able to switch off specific couplings globally by the choice of the laser detunings as discussed below. In a 2D configuration, the situation is even more complex: The angular dependence of the interaction provides a unique opportunity to control the nearest-neighbor- versus longer-ranged interaction and to realize models featuring various magnetic phenomena, including frustration and topology [7, 10, 33, 38–40]. Our approach also opens new pathways to quantum simulations with practical relevance for the inference of Hamiltonians underlying spectra obtained in nuclear magnetic resonance experiments in chemistry and biology [41].

The physical system we use is an optical tweezer array of single \(^{39}\)K atoms. The spins are encoded in the
hyperfine states $|↑⟩ = |4S_{1/2} F = 2, m_F = −2⟩$ and $|↓⟩ = |4S_{1/2} F = 1, m_F = −1⟩$. Both states are coupled individually to the Rydberg states $|r⟩ = |62P_{3/2}, m_j = −3/2⟩$ and $|r_i⟩ = |62P_{3/2}, m_j = 1/2⟩$ by off-resonant single photon excitation at 286 nm with the Rabi frequencies $Ω_↑$ and $Ω_↓$, and detunings $Δ^↑$ and $Δ^↓$ (see Fig. 1). The choice of beam polarizations suppresses single-atom Raman couplings. In this doubly laser-coupled system, rich spin-spin interactions emerge, which are rooted in the strong van der Waals (vdW) interactions between the addressed Rydberg pair states. For the derivation of the spin couplings $J_{ij}^{++}$ and $J_{ij}^{−−}$, we start with diagonalizing $H_{\text{Ryd}} = H_{\text{las}} + H_{\text{int}}$ in the Rydberg pair state basis [33]. Here, $H_{\text{las}}$ is the single atom Hamiltonian in the rotating frame. The vdW Hamiltonian $H_{\text{int}}$ leads to interactions between the different $m_j$ levels in the $62P_{3/2}$ manifold. We admix different components of the vdW pair eigenstates to the ground states by laser coupling to obtain the effective interactions between the ground states.

The interactions in Eq. 1 can be understood as four-photon processes by adiabatic elimination of all excited states (Supplementary Information and ref. [33]). For example, for the flip-flop interactions, the coupling of the $|↑↑⟩$ pair ground state to a Rydberg pair eigenstate $|Ψ_α^{(2)}⟩$ follows by adiabatic elimination of the singly excited state as $Ω_α^{↑↑} = (Ω_↑^2 − Δ α σ^↑ − Δ_α) / 2Δ^↑$, where $c_α^{↑↑} = ⟨Ψ_α^{(2)}|↑↑⟩$ is the wavefunction overlap of one eigenstate $|Ψ_α^{(2)}⟩$ in the Rydberg manifold with the asymptotic Rydberg pair state $|r↑r↓⟩$. The coupling of $|↓↓⟩$ follows analogously. For sufficiently large detuning of the lasers to any coupled state in the Rydberg manifold, we can furthermore eliminate the Rydberg pair eigenstates to arrive at an effective coupling between ground state atoms pairs $i$ and $j$:

$$J_{ij}^{++} = 2 \sum_α \frac{Ω_α^{↑↑}Ω_α^{↓↓}}{Δ_α^{(2)}} = \frac{(Ω_↑^2Δ^↓^2)}{Δ_α^{(2)}} \cdot c_α^{↑↑}c_α^{↓↓}$$

The Rydberg pair state detuning $Δ_α^{(2)}$ includes vdW interaction-induced shifts $U_{\text{vdW, α}}$. Spin flips from $|↑↑⟩$ to $|↓↓⟩$ and vice versa require a non-zero probability overlap $c_α^{↑↑}c_α^{↓↓}$ provided by the mixing of the $m_j$-sublevels.

The derivation of $J_{ij}^{−−}$ starts with two atoms in opposite spin states $|↑↓⟩$ or $|↓↑⟩$. Different from the flip-flop interaction case, there are two excitation paths to the Rydberg manifold. Via adiabatic elimination of the intermediate singly excited state we obtain an effective two-photon coupling $Ω_α^{↑↓} = Ω↑Ω↓c_α^{↑↓}c_α^{↓↑} / (1/4Δ↑ + 1/4Δ↓)$. Then, in fourth order perturbation theory, we obtain the flip-flop interaction:

$$J_{ij}^{−−} = 2 \sum_α \frac{Ω_α^{↑↓}Ω_α^{↓↑}}{Δ_α^{(2)}} = \sum_α \frac{(Ω_↑Ω_↓)^2}{16 (Δ↑Δ↓)^2} \cdot c_α^{↑↓}c_α^{↓↑}$$

For finite flip-flop interaction, we require a non-zero overlap of $c_α^{↑↓}c_α^{↓↑}$. In the case of symmetric detunings

Figure 1. Experimental setup and level schemes. a. Illustration of the experimental setting. The Rydberg excitation beams ($k$, and $k'$, light blue) are aligned along the magnetic field $B$, driving $σ^−$ transitions. They illuminate all tweezer groups, each with three linearly arranged tweezers (red), which are statistically loaded with atoms (black spheres). At the bottom of the illustration, a single shot fluorescence image is shown. b. Example of fluorescence images of fully loaded tweezer groups for various angles $θ$ at a distance of 5.2 μm. c. On the single-atom level, we couple the electronic ground states $|↑⟩$ and $|↓⟩$ to the Rydberg states $|r⟩$ and $|r⟩$ with Rabi frequencies ($Ω^↑$, $Ω^↓$) and detunings ($Δ^↑$, $Δ^↓$). d. Schematic for the flip-flop interaction $J_{ij}^{++}$ between two atoms $i$ and $j$. Via adiabatic elimination of the singly excited pair states, we reduce the four-photon process to an effective $Λ$-scheme. The pairs of ground state atoms are coupled with the effective Rabi couplings $Ω_α^{↑↓}$ and $Ω_α^{↓↑}$ to Rydberg pair states $|Ψ_α^{(2)}⟩$. $Δ_α^{(2)}$ is the two-photon detuning to each $|Ψ_α^{(2)}⟩$, which includes interaction induced shifts. e. Calculated eigenenergies of $H_{\text{Ryd}}$ depending on the atom pair distance $d$ at an angle of $θ = 90°$ (upper) and atom pair angle $θ$ at a distance $d = 6$ μm (lower). The color scale corresponds to the overlap $c_α^{↑↑}c_α^{↓↓}$. The solid lines at ±2 MHz mark the energy of the asymptotic Rydberg pair state $|r↑r↓⟩$ and $|↓↑r↓⟩$. The theoretical results are obtained by exact diagonalization of $H_{\text{Ryd}}$ using the “pairinteraction” software package [32].
Figure 2. Tunable XYZ interactions in 1D chains. In both panels we show the calculation of the same data points, highlighting the distance (a) and angle (b) dependence of the ratios \(J_{ij}^{++}/J_{ij}^z\) and \(J_{ij}^+/J_{ij}^z\) for one exemplary set of detunings \((\Delta^+, \Delta^-) = 2\pi \cdot (1.4, -0.6)\) MHz. The interactions are calculated for distances spaced by 100 nm and angles in steps of 1°. Quadrant II and IV show a smooth tunability, while the ratios in quadrant I and III are realized close to Rydberg pair state resonances. The latter requires higher stability of the control parameters, but may ultimately be beneficial for the achievable coherence [38].

\[ \Delta^+ = -\Delta^- \], the flip-flop interaction is generally canceled by destructive interference of the excitation paths. This provides us with sensitive control of \(J_{ij}^+\) by choosing the excitation laser detuning accordingly. In contrast, energy conservation restricts the flop-flop processes and requires the laser detunings to be set to \(\Delta^+ - \Delta^- = E_z\), with \(E_z\) the Zeeman splitting between \(|\uparrow\rangle\) and \(|\downarrow\rangle\) (see Fig. 3c).

To study the dependence of the interaction strengths on the geometric arrangement experimentally, we select the simplest possible setting of three in-line traps with various nearest-neighbor (nm) distances \(d\) and angles \(\theta\) (see Fig. 1a,b). Here, \(\theta\) is the angle between the interatomic separation vector \(d\) and the magnetic field \(B\), which is set to 1 G and defines the quantization axis. We use 14 replications of this pattern for increased statistics, where the inter-group spacing is 20 \(\mu\)m, larger than any interaction range in the system. With a first fluorescence image of the atom array, we check for the presence of an atom in the trap. We then prepare all atoms in the \(|\uparrow\rangle\) state and perform Raman sideband cooling. This allows us to minimize the trap induced inhomogeneities by working at the lowest possible tweezer depth of \(h \cdot 80\) kHz [42] (Supplementary Information). We then apply two-color Rydberg dressing for 50 \(\mu\)s. Next, we remove all \(|\uparrow\rangle\) atoms by a blowout pulse and detect only the remaining atoms in the \(|\downarrow\rangle\) state with a second fluorescence image. Comparing both fluorescence images allows us to infer the spin interactions by observing flipped spins and their correlations.

First, we aim to reveal the induced flop-flop interactions by choosing our detuning symmetric \(\Delta^+ = -\Delta^-\), to cancel the flip-flop terms. We map the spatial dependence of the interactions by preparing the atoms at different distances and angles. We do not observe significant single spin flips, confirming the suppression of single-atom Raman processes. The \(J^{++}\) interaction leads to pairwise spin-flips, which we observe in our setting between nearest neighbors. The distance dependence of the pairwise spin flips is shown in Fig. 3, where we scan the atoms' distances at \(\theta = 90^\circ\) and Rabi couplings \((\Omega^+, \Omega^-) = 2\pi \cdot (0.52, 0.36)\) MHz. The experimental data and the amplitude-scaled theoretical prediction are overall in good agreement. Differences in theory and experiment emerge from several line-broadening effects, such as the finite size of the atoms' thermal wavepacket in radial and axial direction in the traps (Supplementary Information), which results in an averaging over a range of atom pair separations and angles within the radial ground state wavepacket size of \(\sigma_{\text{rad}} = 0.15\) \(\mu\)m and the axial
thermal wavepacket size $\sqrt{2}\sigma_{\text{ax}}^0 \sqrt{k_B T / \hbar \omega_{\text{ax}}} \approx 0.86 \mu m$ for the axial trapping frequency $\omega_{\text{ax}} = 2\pi \cdot 1.7 \text{kHz}$. A second, equally important, effect is caused by the line shifts due to tweezer-to-tweezer inhomogeneities which result in an average trap depth difference of $|\Delta U| = \hbar \cdot (10.6 \pm 1.6) \text{kHz}$. In addition, laser phase noise currently limits the dressing time due to a 20-fold increased scattering rate appearing as atom loss [43].

We additionally map out the angular dependence of the flip-flop interaction for a fixed distance of 5.6 µm and Rabi couplings of $(\Omega^+, \Omega^-) = 2\pi \cdot (0.55, 0.30) \text{MHz}$. In this measurement, we cross a singularity in the spin-spin coupling at $\theta \approx 30^\circ$ caused by a Rydberg pair state resonance. We reproduce a peaked interaction around this resonance, shown in Fig. 4d, while the broadening effects explain the weak atom loss by direct Rydberg excitation on resonance (Supplementary Information).

In the second set of measurements, we switch on both flop-flop and flip-flop interactions by setting the detunings to $(\Delta^+, \Delta^-) = 2\pi \cdot (1.4, -0.6) \text{MHz}$ and Rabi frequencies to $(\Omega^+, \Omega^-) = 2\pi \cdot (0.5, 0.36) \text{MHz}$. For the analysis of the spin interactions, we take into account different initial states prepared in the statistical loading of the traps (see Fig. 4). These configurations of interest are chosen in postselection and correspond to either a fully loaded group ($|\uparrow\uparrow\uparrow\rangle$) or groups where only two out of three tweezer sites at the nearest neighbor distance are filled ($|\uparrow\uparrow\circ\rangle$, $|\circ\uparrow\uparrow\rangle$, where $\circ$ indicates an empty site). In the latter configuration, only flop-flop processes occur, while in the case of three atoms also flip-flop processes appear. Comparing the two fluorescence images before and after the dressing phase allows us to identify which interaction processes occurred. More precisely, assuming a $|\uparrow\uparrow\uparrow\rangle$ occupation at the beginning, a flop-flop interaction can produce a $|\uparrow\downarrow\downarrow\rangle$ ($|\uparrow\downarrow\uparrow\rangle$) arrangement, and flip-flop processes introduce the $|\downarrow\uparrow\uparrow\rangle$ state, after push out detected as $|\downarrow\circ\downarrow\rangle$. The pure flop-flop process results in the presence of two atoms at the nearest neighbor distance on the second image. We predict a different spatial dependence of $J^{++}$ and $J^{+-}$ (see Fig. 4b,c). Note that the detection method used here always requires flop-flop to be present in order to initiate the dynamics out of the fully polarized initial state.

Our data reveals the angular- and distance-dependent $J_{ij}^{++}$ interactions for an asymmetric detuning in Fig. 4d,f. In addition, we measure a peaked occurrence of the $|\downarrow\circ\downarrow\rangle$ pattern, which we identify as the flip-flop interaction shown in Fig. 4e. This reflects the tunability of our system by introducing $J_{ij}^{-+}$ interactions for a given laser detuning, atom pair distance, and angle. In addition, we probe our system such that the flip-flop interaction strength vanishes and only flop-flop interactions occur (see Fig. 4f,g). Here, we scanned the angular dependence of the interactions at a fixed distance of 5.3 µm without crossing a Rydberg pair resonance. The minimum in the signal, around 65°, is caused by interference on the two-atom level. Multiple Rydberg pair states $|\Psi_{\alpha}^{(2)}\rangle$ with admixtures $c_{\alpha}^{\uparrow\uparrow} c_{\alpha}^{\downarrow\downarrow}$ of opposite sign contribute such that $J^{++}$ vanishes.

In conclusion, we have demonstrated two-color Rydberg-dressing as a new technique to achieve tunable, XYZ-type short-range spin interactions in optical
tweezer arrays. Technical limitations currently prevent us from probing coherent interactions (see Supplementary Information). The two leading limitations stem from tweezer-to-tweezer line shifts due to array inhomogeneities and from laser phase noise. None of these are fundamental, in fact, other groups have reported tweezer arrays with less than 1.1% inhomogeneity [44], a factor of 10 improvement over our arrays. Laser phase noise can be filtered by optical cavities, as demonstrated for Rydberg excitation in ref. [45]. Furthermore, it has been shown that the observed Rydberg pair state resonances can be utilized to enhance the coherence of Rydberg dressing [38]. By implementing these measures, we estimate that a maximum figure of merit, measured as the product of the peak interaction strength and the coherence time, of up to one hundred is reachable with current laser technology. This will allow one to realize a flexibly programmable analog quantum simulation platform for many-body quantum spin problems. Not only the ratio of the spin-interactions in the different channels can be controlled, but also the ratio of nearest- to next-nearest-neighbor interactions. This is rooted in the non-monotonic spatial dependence of the interaction strength, which can also be used to design interactions in two-dimensions for the realization of a variety of frustrated geometries [33], static [39] or dynamic gauge fields [40].

Data and materials availability: The experimental and theoretical data and evaluation scripts that support the findings of this study are available on Zenodo [46].

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SUPPLEMENTARY INFORMATION

This supplementary information document provides information about the experimental sequence and laser setup, a detailed derivation of the interactions, and a discussion of our experimental limitations.

I. EXPERIMENTAL SEQUENCE

The optical tweezers are generated using a commercial 1064 nm laser aligned onto a liquid crystal spatial light modulator, which imprints a phase pattern onto the beam. An in-vacuum mounted objective (NA = 0.6) focuses the linearly-polarized beam, obtaining a tweezer array with controlled geometry, of which each trap has a waist of 0.9 µm. We load $^{39}$K atoms into the traps by alternating trapping and cooling light with a frequency of 1.4 MHz [47], one order of magnitude faster than the radial trapping frequency of $\omega_r = 2\pi \cdot 158$ kHz ($\omega_z = 2\pi \cdot 25$ kHz). On average, 50% of the traps are filled with a single atom [42, 43], and the experimental cycle rate is 1 Hz.

After a first fluorescence image probing the tweezer filling, we optically pump and prepare the atoms in the $|↑⟩ = |4S_{1/2}, F = 2, m_F = −2⟩$ state with $σ^−$-polarized pumping and repumping light on the D1-line. To quantitatively analyze experimental results, we compare both fluorescence images allowing us to deduce the spin interactions based on spin flips and their correlations.

II. RYDBERG LASER SETUP

The Rydberg dressing laser setup consists of a homebuilt ECDL laser at 1143.5 nm, which is amplified to 8 W via a commercial Raman fiber amplifier and then frequency-quadrupled in two consecutive, homebuilt cavity-enhanced doubling stages. This results in an output power of 1 W at 286 nm. This UV beam is then split into two paths with acousto-optical modulators (AOM) with frequencies of ±230 MHz, which we use for intensity stabilization and bridging the hyperfine ground state splitting. The beams are then overlapped and focused onto the atoms, with a horizontal waist of 40 µm and a vertical waist of 10 µm. The Rydberg excitation beams propagate parallel to the magnetic field and drive $σ^−$ transitions.

The lifetime of the dressed ground states is proportional to the Rydberg state probability. For our parameters and assuming a phase-noise-free laser, we expect a dressed (black-body radiation limited) lifetime of $\tau_{ds} = 1.7$ ms. In contrast, the experimentally observed lifetime is reduced to 70 µs (for $\Delta_1 = −2\pi \cdot 0.6$ MHz and $Ω_1 = 2\pi \cdot 0.4$ MHz) due to laser noise [43]. Atom loss due to excitation to Rydberg pair resonances is weak, as shown in Fig. S1. Here, the data has been postselected to only nearest neighbor tweezer pairs initially loaded. In the measurement, we did not apply a push out pulse, to realize spin-insensitive imaging. All Rabi frequencies have been measured before the respective set of measurements runs by driving Rabi oscillations without trapping light. The typical uncertainty of the Rabi frequency fits 0.01 MHz.

We then apply Raman sideband cooling (RSC) [42], which enables us to lower the trap depth to a minimum of $h \cdot 80$ kHz before gravity opens the trap. At these low intensities, the light shift induced inhomogeneities of the Rydberg excitation lines are reduced to a few kHz (see section IV B). We continue with a 50 µs long pulse of both Rydberg dressing lasers (details see section II). The magnetic field strength of $B = 1$ G leads to a Zeeman splitting of the Rydberg states $E_z = h \cdot 1.98$ MHz.

The Rydberg excitation beams couple the following ground states

\begin{equation}
\begin{aligned}
|↑⟩ &= |4S_{1/2}, F = 2, m_F = −2⟩ = |4S_{1/2}, m_j = −1/2⟩ |m_I = −3/2⟩ \\
|↓⟩ &= |4S_{1/2}, F = 1, m_F = −1⟩ = |4S_{1/2}, (m_j = −1/2⟩ |m_I = −1/2⟩ − \sqrt{3} |m_j = 1/2⟩ |m_I = −3/2⟩)/2
\end{aligned}
\end{equation}
The pair interaction Hamiltonian $\hat{H}_\text{int}$ arises from the dipolar interactions among the Rydberg states. We use the “pairinteraction” software package to diagonalize the interaction Hamiltonian [32] and to obtain the pair-separation $d$ and -angle $\theta$ dependent eigenstates $|\Psi_\alpha^{(2)}\rangle$ with eigenenergies $E_\alpha(d,\theta)$. The eigenstates can be developed in asymptotic pair states $|r^m r^n\rangle$ as $|\Psi_\alpha^{(2)}\rangle = \sum_{m,n} c_\alpha^{mn}(d, \theta) |r^m r^n\rangle$ with the distance- and angle dependent admixture $c_\alpha^{mn}(d, \theta) = \langle \Psi_\alpha^{(2)} | r^m r^n \rangle$. To improve the readability, we suppress the explicit $d, \theta$-dependency in the rest of the text.

The dipolar interactions between any pair of Rydberg states $|r^m r^n\rangle$ can be written in the pair basis $\{ |r^m r^n\rangle, |r^m r^n\rangle, |r^m r^n\rangle, |r^m r^n\rangle \}$ in the form

$$\hat{H}_\text{int} = \begin{pmatrix} V^{mm,mm} & 0 & 0 & V^{mm,nn} \\ 0 & V^{nn,nn} & 0 & V^{nn,nn} \\ 0 & 0 & V^{nn,nn} & 0 \\ V^{mm,nn} & 0 & 0 & V^{nn,nn} \end{pmatrix}.$$  \hspace{1cm} (S.3)

When adiabatically eliminating the Rydberg states (equivalent to 4-th order perturbation theory, c.f. ref. [33]), this form of the interactions is transferred to the dressed ground states and the effective Hamiltonian $\hat{H}_\text{eff}$ reads in the $\{ |\uparrow\uparrow\rangle, |\downarrow\downarrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle \}$ basis:

$$\hat{H}_\text{eff} = \begin{pmatrix} W^{\uparrow\uparrow,\uparrow\uparrow} & 0 & 0 & W^{\uparrow\uparrow,\downarrow\downarrow} \\ 0 & W^{\downarrow\downarrow,\uparrow\uparrow} & W^{\uparrow\downarrow,\downarrow\downarrow} & 0 \\ 0 & W^{\downarrow\uparrow,\downarrow\uparrow} & W^{\downarrow\uparrow,\uparrow\uparrow} & 0 \\ W^{\downarrow\uparrow,\downarrow\uparrow} & 0 & 0 & W^{\downarrow\uparrow,\downarrow\downarrow} \end{pmatrix}.$$  \hspace{1cm} (S.4)

In the following, we develop an intuitive picture for deriving the different entries of this effective interaction matrix. The general idea is a step-wise elimination of the Rydberg levels, starting with singly excited states to obtain a $\Lambda$-system. In the second step, we also eliminate the doubly excited states to arrive at an effective Hamiltonian for the dressed ground states. This procedure is illustrated in Fig. S2. Here, we assume that there are only four relevant (i.e. near-resonantly) laser coupled asymptotic pair states, the states $\{ |r^\uparrow r^\uparrow\rangle, |r^\downarrow r^\uparrow\rangle, |r^\downarrow r^\downarrow\rangle, |r^\uparrow r^\downarrow\rangle \}$.

A. The diagonal coupling terms

For the derivation of $W^{\sigma_-\sigma_-,\sigma_-\sigma_-}$, we start with adiabatic elimination of the single excited state $|+\rangle = (|\sigma\sigma\rangle + |\sigma\sigma\rangle)/\sqrt{2}$. For large atom distances, we obtain the effective two-photon Rabi couplings $\Omega^{\sigma} = (\Omega^{\sigma})^2/2\Delta^\sigma$. At short distances, the pair potentials in the $m_\sigma$-subspace of the 62$D_{5/2}$ manifold interact with each other via dipole-quadrupole interaction, which leads to avoided crossings and mixing of Rydberg states [48]. The corresponding admixture $c^{(2)}_{\alpha} = \langle \Psi_{\alpha}^{(2)} | r^\sigma r^\tau \rangle$ of $|r^\sigma r^\tau\rangle$ in close-by interacting pairstates $|\Psi_{\alpha}^{(2)}\rangle$ reduces the effective Rabi frequencies to $\Omega_{\alpha}^{\sigma} = \Omega_{\alpha}^2 c_{\alpha}^{(2)}$. We then adiabatically eliminate the Rydberg pairstates and subtract the asymptotic value of $W^{\sigma_-\sigma_-,\sigma_-\sigma_-}$ for $d = \infty$ to eliminate a constant offset.

$$W^{\sigma_-\sigma_-,\sigma_-\sigma_-} = \frac{(\Omega^{\sigma})^4}{4(\Delta^{\sigma})^2} \sum_{\alpha} \left( \frac{(\omega_{\alpha}^{\sigma})^2}{(\Delta_{\alpha}^{(2)})^2} - \frac{1}{2\Delta^{\sigma}} \right),$$  \hspace{1cm} (S.5)

with the Rydberg pair state detuning $\Delta_{\alpha}^{(2)} = 2\Delta^{\sigma} - E_{\alpha}$. The derivation of the $W^{\sigma_-\sigma_-,\sigma_-\sigma_-}$ (with $\sigma \neq \bar{\sigma}$) is similar: As there are two excitation paths from $|\sigma\bar{\sigma}\rangle$ to $|\Psi_{\alpha}^{(2)}\rangle$, the reduced two-photon coupling is
Figure S2. Stepwise adiabatic elimination. a. Elimination of singly excited Rydberg states. We show the level schemes corresponding to the coupling of different ground state spin-pairs to the Rydberg manifold. The singly excited states are adiabatically eliminated to obtain an effective pair state coupling $\Omega_{\sigma'\sigma}$ to the eigenstates of the dipolar interaction Hamiltonian $|\Psi^{(2)}\rangle$. b. Elimination of doubly excited states. The result of the first elimination step is an effective $\Lambda$-system, in which we eliminate the doubly excited states $|\Psi^{(2)}\rangle$ to obtain effective ground state coupling (for unequal initial and final states), or a light shift (for equal initial and final states).

\[
\Omega_{\sigma'\sigma} = \left(\frac{\Omega^{(2)}}{4\Delta^{(2)}} + \Omega^\sigma \Omega^{\bar{\sigma}} / 4\Delta \right) \cdot c_{\sigma'\sigma}.
\]

Again we adiabatically eliminate $|\Psi^{(2)}\rangle$ and remove a constant offset by subtracting the $d = \infty$ asymptotic value to obtain:

\[
W_{\sigma\bar{\sigma},\sigma\bar{\sigma}} = \sum_{\alpha} \frac{\Omega^{\sigma\sigma} \Omega^{\bar{\sigma}\bar{\sigma}}}{\Delta^{(2)}_{\alpha}} \cdot \frac{\Omega^{(2)}}{4\Delta^{(2)}} \cdot c_{\sigma'\sigma}.
\]

\[
\Omega^{\sigma\sigma} = \left(\frac{\Omega^{(2)}}{4\Delta^{(2)}} + \Omega^\sigma \Omega^{\bar{\sigma}} / 4\Delta \right) \cdot c_{\sigma'\sigma}.
\]

\[
W^{\sigma\sigma,\sigma\bar{\sigma}} = \sum_{\alpha} \frac{\Omega^{\sigma\sigma} \Omega^{\bar{\sigma}\bar{\sigma}}}{\Delta^{(2)}_{\alpha}} \cdot \frac{\Omega^{(2)}}{4\Delta^{(2)}} \cdot c_{\sigma'\sigma}\cdot c_{\bar{\sigma}\bar{\sigma}}.
\]

\[
W^{\sigma\bar{\sigma},\sigma\sigma} = \sum_{\alpha} \frac{\Omega^{\sigma\sigma} \Omega^{\bar{\sigma}\bar{\sigma}}}{\Delta^{(2)}_{\alpha}} \cdot \frac{\Omega^{(2)}}{4\Delta^{(2)}} \cdot c_{\sigma'\sigma}\cdot c_{\bar{\sigma}\bar{\sigma}}.
\]

\[
W^{\sigma\bar{\sigma},\sigma\bar{\sigma}} = \sum_{\alpha} \frac{\Omega^{\sigma\sigma} \Omega^{\bar{\sigma}\bar{\sigma}}}{\Delta^{(2)}_{\alpha}} \cdot \frac{\Omega^{(2)}}{4\Delta^{(2)}} \cdot c_{\sigma'\sigma}\cdot c_{\bar{\sigma}\bar{\sigma}}.
\]

B. The flop-flop off-diagonal terms

The flop-flop coupling terms $W^{\sigma\bar{\sigma},\sigma\sigma}$ are derived analogously. The effective Rabi frequencies for the flop-flop interactions are $\Omega^{\sigma\sigma} = \Omega_{\sigma\sigma}' + c_{\sigma'\sigma}$. Via adiabatic elimination of the Rydberg pair states, we obtain the flop-flop coupling term. For the off-diagonal terms, offsets at $d = \infty$ are absent since there are two different asymptotic pair state overlaps involved, and one of them must vanish asymptotically. We obtain:

\[
W^{\sigma\bar{\sigma},\sigma\bar{\sigma}} = \sum_{\alpha} \frac{\Omega^{\sigma\sigma} \Omega^{\bar{\sigma}\bar{\sigma}}}{\Delta^{(2)}_{\alpha}} \cdot \frac{\Omega^{(2)}}{4\Delta^{(2)}} \cdot c_{\sigma'\sigma}\cdot c_{\bar{\sigma}\bar{\sigma}}.
\]
Figure S3. **Tweezer inhomogeneities.** Distribution of the trap depth difference $|\Delta U|$ for two tweezers at the nearest neighbor distance and for the minimum trap depth of $\hbar \cdot 80 \text{ kHz}$. The Gaussian fit (solid line) reveals an average trap depth difference of $\langle \Delta U \rangle = \hbar \cdot (10.6 \pm 1.6) \text{ kHz}$. For our tweezers generated with 1064 nm light, the magnitude of the ponderomotive potential for the Rydberg states approximately equals the trap depth for the ground states but is of the opposite sign. Hence, the difference in the line shifts of the ground state-Rydberg transition for nearest-neighbor pairs is about $2|\Delta U|$.

D. Formulation of an effective spin Hamiltonian

The XYZ-Hamiltonian written in terms of Pauli matrices $\sigma^x, \sigma^y, \sigma^z$ reads

$$\hat{H}_{\text{XYZ}} = \hbar \sum_{ij} [J_{ij}^+ \sigma_i^+ \sigma_j^- + J_{ij}^- \sigma_i^- \sigma_j^+ + J_{ij}^z \sigma_i^z \sigma_j^z], \quad (S.9)$$

or alternatively in raising/lowering form

$$\hat{H}_{\text{XYZ}} = \hbar \sum_{ij} [J_{ij}^{\uparrow \downarrow} (\sigma_i^+ \sigma_j^- - \sigma_i^- \sigma_j^+) + J_{ij}^{\downarrow \uparrow} (\sigma_i^- \sigma_j^+ - \sigma_i^+ \sigma_j^-) + J_{ij}^z \sigma_i^z \sigma_j^z], \quad (S.10)$$

with $\sigma_i^z = (\sigma_i^- + \sigma_i^+)$, $\sigma_i^y = i(\sigma_i^- - \sigma_i^+)$, the flop-flop coupling $J_{ij}^{\uparrow \downarrow} = (J_{ij}^- - J_{ij}^+)$ and the flip-flop coupling $J_{ij}^{\downarrow \uparrow} = (J_{ij}^+ + J_{ij}^-)$.

By expanding Eq. S.10 in the ground state pair basis and comparing to Eq. S.4 one identifies

$$J_{ij}^{\uparrow \downarrow} = W^{\uparrow \downarrow \downarrow \uparrow}(d_{ij}, \theta_{ij}) + W^{\downarrow \uparrow \uparrow \downarrow}(d_{ij}, \theta_{ij})$$

$$- 2W^{\uparrow \downarrow \downarrow \downarrow}(d_{ij}, \theta_{ij})$$

$$J_{ij}^{\downarrow \uparrow} = 2W^{\uparrow \downarrow \downarrow \downarrow}(d_{ij}, \theta_{ij})$$

$$J_{ij}^z = 2W^{\uparrow \downarrow \downarrow \downarrow}(d_{ij}, \theta_{ij})$$

where we used $W^{\uparrow \downarrow \downarrow \downarrow} = W^{\downarrow \uparrow \uparrow \downarrow} = W^{\downarrow \downarrow \uparrow \uparrow}$, and $W^{\uparrow \downarrow \downarrow \downarrow} = W^{\downarrow \uparrow \uparrow \downarrow}$. For clarity, we also restored the pair-separation and -angle dependence here.

IV. EXPERIMENTAL LIMITATIONS

In our setup, laser noise and inhomogeneous line shifts due to the trapping laser are the main limitations preventing us from probing coherent interactions. In the following, we discuss these limitations and their consequences.

A. Laser noise

Phase noise of our Rydberg excitation laser results in an incoherently enhanced population of the Rydberg states. The Rydberg population is determined by $\beta^2 = \Omega^2/4\Delta^2$ for negligible noise, resulting in an excitation rate of $\beta^2 \gamma_r$. Here, $\gamma_r^{-1}$ is the Rydberg-state lifetime. In our experiment, we measure an about 20-fold increased scattering rate by observing the trap loss $[43]$.

B. Inhomogeneity of trap depths

To determine the depth of the tweezers, we measure the AC Stark shift on the D1-line by spectroscopy. We first prepare the atoms in the $|F = 2, m_F = 2\rangle$ state and set the magnetic field perpendicular to the optical beams such that we probe different polarizations. On resonance, the atoms are pumped to the $F = 1$ hyperfine manifold. Then we adiabatically rotate the magnetic field parallel to the direction of the laser beam and remove all atoms in the $F = 2$ hyperfine manifold with light resonant to the $|F = 2, m_F = 2\rangle$ to $|F' = 3, m_{F'} = 3\rangle$ cycling transition of the D2-line. We measure the light shift at an average trap depth of $200 \, \mu \text{K}$ and scale the results to the minimal depth used for experiments described in the main text. In Fig. S3 we show the nearest-neighbor tweezer trap depth difference $|\Delta U|$ of a $3 \times 14$ tweezer array.

C. In-trap wavepacket size

Tweezer inhomogeneities force us to work at a minimal trap depth of $\hbar \cdot 80 \, \text{kHz}$. This results in a radial (axial) trapping frequency of $\omega_{\text{rad}} = 2\pi \cdot 11 \, \text{kHz}$ ($\omega_{\text{ax}} = 2\pi \cdot 1.7 \, \text{kHz}$) with corresponding radial (axial) ground state wavepacket sizes of $\sigma_{\text{rad}}^0 = \sqrt{\hbar/(m\omega_{\text{rad}})} = 0.15 \, \mu \text{m}$ ($\sigma_{\text{ax}}^0 = 0.39 \, \mu \text{m}$).

The temperature of our Raman cooled atoms corresponds to $k_B T = \hbar \cdot 4.2 \, \text{kHz}$ as measured in ref. [42]. Since
the temperature is below the trapping frequency in radial
direction we use the ground state wavepacket size to esti-
 cate the radial pair-distance fluctuations $\sigma_{\text{rad}} \approx \sqrt{2}\sigma_{\text{rad}}^0$. The factor $\sqrt{2}$ takes the independent motion of the two
atoms into account.

The impact of the axial (out-of-plane) wave packet
sizes is much weaker and we neglect its effect in the main
text. Nevertheless, it explains the comparably strong
collapse-flop interactions for small angles for the measure-
ment shown in Fig. 3f of the main text. In Fig. S4 we show the effect of a thermal wavepacket of size $\sqrt{2}\sigma_{\text{ax}}^0 \sqrt{k_B T/\hbar\omega_{\text{ax}}} \approx 0.86\,\mu\text{m}$ on the flop-flop interac-
tions. We use the large temperature limit for the estima-
tion of the position fluctuations here since $k_B T > \hbar\omega_{\text{ax}}$. The out-of-plane fluctuations result in an averaging over
a range of angles, removing the zero in the interactions
at a mean angle of $\theta = 0^\circ$. 