SUPPLEMENTARY INFORMATION

Equilibrium positions and static potential energy

The trap potential of two ions with state-dependent trap frequencies ω^α with $\alpha = \{\uparrow, \downarrow\}$ resembling two possible internal states, reads

$$V^{\alpha\beta} = \frac{1}{2} m \left(\omega^\alpha\right)^2 Z_1^2 + \frac{1}{2} m \left(\omega^\beta\right)^2 Z_2^2 + \frac{\mathrm{C}}{|Z_1 - Z_2|} - 4e\gamma \left(Z_1 r_1 + Z_2 r_2\right), \quad (1)$$

where Z_j is the position of the j-th ion and $\mathrm{C} = e^2/4\pi\epsilon_0$ is the Coulomb constant with e the elementary charge, ϵ_0 the vacuum permittivity, γ the trap field gradient and r_j the relative coordinate of the outer electron of each ion. We have introduced the interaction between the excited electron of a Rydberg state with the ion core by the last term. For small vibrations z_j of the ionic core around the equilibrium position $\bar{Z}_j$, we expand the potential

$$\begin{aligned}
V^{\alpha\beta} &\approx z_1 \left[m \left(\omega^\alpha\right)^2 \bar{Z}_1 - \frac{\mathrm{C}}{(\bar{Z}_1 - \bar{Z}_2)^2} \right] \\
&\quad + z_2 \left[m \left(\omega^\beta\right)^2 \bar{Z}_2 + \frac{\mathrm{C}}{(\bar{Z}_1 - \bar{Z}_2)^2} \right] \\
&\quad + V_{\mathrm{trap}} + V_{\mathrm{DC}} + V_0^{\alpha\beta} + \mathcal{O}(z_1^n z_2^l), \\
V_{\mathrm{trap}} &= z_1^2 \left[\frac{m \left(\omega^\alpha\right)^2}{2} + \frac{\mathrm{C}}{|\bar{Z}_1 - \bar{Z}_2|^3} \right] \\
&\quad + z_2^2 \left[\frac{m \left(\omega^\beta\right)^2}{2} + \frac{\mathrm{C}}{|\bar{Z}_1 - \bar{Z}_2|^3} \right] - \frac{2\mathrm{C} z_1 z_2}{|\bar{Z}_1 - \bar{Z}_2|^3} \\
V_{\mathrm{DC}} &= z_1 \left[-4e\gamma r_1 + \frac{2\mathrm{C}(r_1 - r_2)}{|\bar{Z}_1 - \bar{Z}_2|^3} \right] \\
&\quad + z_2 \left[-4e\gamma r_2 - \frac{2\mathrm{C}(r_1 - r_2)}{|\bar{Z}_1 - \bar{Z}_2|^3} \right]
\end{aligned}$$

where we kept up to quadratic expansions in terms of small quantities r_j and z_j. $V_0^{\alpha\beta}$ gives a static potential, which depends on the equilibrium positions,

$$V_0^{\alpha\beta} = \frac{m \left(\omega^\alpha\right)^2 \bar{Z}_1^2}{2} + \frac{m \left(\omega^\beta\right)^2 \bar{Z}_2^2}{2} + \frac{\mathrm{C}}{|\bar{Z}_1 - \bar{Z}_2|} \quad (2)$$

As the change in equilibrium positions by the additional static potential V_{DC} is typically small, we calculate the ion distances between two ground state ions:

$$\frac{\mathrm{C}}{|\bar{Z}_1 - \bar{Z}_2|^3} = 2e\gamma$$

and obtain:

$$V_{\mathrm{DC}} \approx -4e\gamma (r_2 z_1 + r_1 z_2) \quad (3)$$

We identify this term as a cross-coupling between the negatively singly charged electron of one ion with the positively double charged core of the other ion. For two ions in the Rydberg state both ions see an additional trap potential. If only one ion is in the Rydberg state, the neighboring ground state ion will see a modified trap potential. The term V_{DC} describes the electron dynamics, which is typically much faster than the ion core dynamics, and can be treated via second order perturbation. Thereby, we obtain modified trap frequencies for Rydberg ions compared to ground state ions (see main text). From the force balance condition (linear orders) the state-dependent equilibrium positions are obtained

$$\bar{Z}_1^{\alpha\beta} = \left[\frac{\mathrm{C} \left(\omega^\beta\right)^4}{m \left(\omega^\alpha\right)^2 \left(\left(\omega^\alpha\right)^2 + \left(\omega^\beta\right)^2 \right)} \right]^{\frac{1}{3}}, \quad (4)$$

$$\bar{Z}_2^{\alpha\beta} = - \left[\frac{\mathrm{C} \left(\omega^\alpha\right)^4}{m \left(\omega^\beta\right)^2 \left(\left(\omega^\alpha\right)^2 + \left(\omega^\beta\right)^2 \right)} \right]^{\frac{1}{3}}, \quad (5)$$

and we find the geometric center of the two ions:

$$\begin{aligned}
Z_{\mathrm{c}}^{\alpha\beta} &= \bar{Z}_1^{\alpha\beta} + \bar{Z}_2^{\alpha\beta} \\
&= \frac{\mathrm{C}^{1/3} \left(\left(\omega^\beta\right)^2 - \left(\omega^\alpha\right)^2 \right)}{\left[m \left(\omega^\beta\right)^2 \left(\omega^\alpha\right)^2 \left(\left(\omega^\alpha\right)^2 + \left(\omega^\beta\right)^2 \right)^2 \right]^{1/3}}. \quad (6)
\end{aligned}$$

With the equilibrium positions, we can calculate the static potential

$$V_0^{\alpha\beta} = \frac{3}{2} \left[\frac{m \mathrm{C}^2 \left(\omega^\alpha\right)^2 \left(\omega^\beta\right)^2}{\left(\omega^\alpha\right)^2 + \left(\omega^\beta\right)^2} \right]^{\frac{1}{3}} \quad (7)$$

We should note that the state-dependent difference of the static potential $V_0^{\alpha\beta}$ will be canceled by proper laser detuning. This means that the laser frequency for excitation of a single ion and two ions to the Rydberg state will be different.

Phonon modes

Using the equilibrium positions, we obtain

$$\begin{aligned}
V^{\alpha\beta} &= \frac{m z_1^2}{2} \left[\left(\omega^\alpha\right)^2 + \left(J^{\alpha\beta}\right)^2 \right] \\
&\quad + \frac{m z_2^2}{2} \left[\left(\omega^\beta\right)^2 + \left(J^{\alpha\beta}\right)^2 \right] \\
&\quad - m \left(J^{\alpha\beta}\right)^2 z_1 z_2 + V_0^{\alpha\beta}, \quad (8)
\end{aligned}$$

with

$$\left(J^{\alpha\beta}\right)^2 = \frac{2 \left(\omega^\alpha \omega^\beta\right)^2}{\left(\omega^\alpha\right)^2 + \left(\omega^\beta\right)^2} \quad (9)$$

We introduce a transformation, which mixes position coordinates by an angle θ:

$$\begin{pmatrix} z_1 \\ z_2 \end{pmatrix} = \begin{pmatrix} \cos\theta & -\sin\theta \\ \sin\theta & \cos\theta \end{pmatrix} \begin{pmatrix} q_1 \\ q_2 \end{pmatrix}, \quad (10)$$

and the potential becomes

$$V = \frac{m}{2}\left(\left(\omega_1^{\alpha\beta}\right)^2 q_1^2 + \left(\omega_2^{\alpha\beta}\right)^2 q_2^2\right) + V_0^{\alpha\beta} \tag{11}$$
$$+ \frac{m}{2}\left[\left((\omega^\alpha)^2 - \left(\omega^\beta\right)^2\right)\sin 2\theta - 2\left(J^{\alpha\beta}\right)^2 \cos 2\theta\right] q_1 q_2$$

with

$$\left(\omega_j^{\alpha\beta}\right)^2 = \omega^\alpha\omega^\beta\left[\left(\frac{\omega^\beta}{\omega^\alpha}\right)^{(-1)^j}\cos^2\theta^{\alpha\beta} + \left(\frac{\omega^\alpha}{\omega^\beta}\right)^{(-1)^j}\sin^2\theta^{\alpha\beta}\right]$$
$$+ (J^{\alpha\beta})^2\left[1 + (-1)^j\sin(2\theta^{\alpha\beta})\right]. \tag{12}$$

The potential is diagonal when the mixing angle becomes

$$\theta^{\alpha\beta} = \frac{\pi}{4} - \frac{1}{2}\arctan\frac{(\omega^\alpha)^2 - \left(\omega^\beta\right)^2}{2\left(J^{\alpha\beta}\right)^2}. \tag{13}$$

At low temperatures, the vibrations are described by vibrational quanta acting on the collective coordinates $q_j = l_j^{\alpha\beta}\left(\tilde{a}_j^\dagger + \tilde{a}_j\right)$ with the oscillator length $l_j^{\alpha\beta} = \sqrt{\hbar/\left(2m\omega_j^{\alpha\beta}\right)}$ and the phonon Hamiltonian is expressed in terms of the state-depending creation $\tilde{a}_j^\dagger = (a_j^{\alpha\beta})^\dagger$ and annihilation $\tilde{a}_j = a_j^{\alpha\beta}$ operators ($\hbar = 1$).

$$H^{\alpha\beta} = \sum_{j=1}^{2}\omega_j^{\alpha\beta}\tilde{a}_j^\dagger\tilde{a}_j + V_0^{\alpha\beta}. \tag{14}$$

Summing up all basis states we obtain the phonon Hamiltonian of the full system:

$$H_{\rm p} = \sum_{\alpha\beta=\uparrow,\downarrow}\left(H^{\alpha\beta}\right)\Pi^{\alpha\beta} \tag{15}$$

with the state projection operator

$$\Pi^{\alpha\beta} = |\alpha\rangle_1\langle\alpha|_1 \otimes |\beta\rangle_2\langle\beta|_2 \tag{16}$$

Electric kick

The Hamiltonian for the fast electric pulse driving the ions is given by

$$H_{\rm d} = f(t)(Z_1 + Z_2) \tag{17}$$
$$= f(t)(z_1 + z_2) + f(t)(\bar{Z}_1^{\alpha\beta} + \bar{Z}_2^{\alpha\beta}) \tag{18}$$
$$= f(t)(\cos\theta^{\alpha\beta} + \sin\theta^{\alpha\beta})q_1$$
$$+ f(t)(-\sin\theta^{\alpha\beta} + \cos\theta^{\alpha\beta})q_2 + f(t)Z_{\rm c} \tag{19}$$
$$= f(t)l_1^{\alpha\beta}(\cos\theta^{\alpha\beta} + \sin\theta^{\alpha\beta})\left(\tilde{a}_1^\dagger + \tilde{a}_1\right)$$
$$+ f(t)l_2^{\alpha\beta}(\cos\theta^{\alpha\beta} - \sin\theta^{\alpha\beta})\left(\tilde{a}_2^\dagger + \tilde{a}_2\right)$$
$$+ f(t)Z_{\rm c}^{\alpha\beta} \tag{20}$$
$$= \sum_{j=1}^{2}F_j^{\alpha\beta}(t)\left(\tilde{a}_j^\dagger + \tilde{a}_j\right) + f(t)Z_{\rm c}^{\alpha\beta}. \tag{21}$$

The driving Hamiltonian is given by:

$$H_{\rm d}(t) = \sum_{\alpha\beta}\left[\sum_{j=1}^{2}(F_j^{\alpha\beta}(t)\,\tilde{a}_j + \text{h.c.}) + f(t)\,Z_{\rm c}^{\alpha\beta}\right]\Pi^{\alpha\beta}. \tag{22}$$

Time evolution operator

We obtain the interaction Hamiltonian by:

$$H_{\rm I} = e^{iH_{\rm p}t}\,H_{\rm d}\,e^{-iH_{\rm p}t}$$
$$H_{\rm I} = \sum_{\alpha\beta}\left(\sum_{j=1}^{2}\left(F_j^{\alpha\beta}(t)e^{i\omega_j^{\alpha\beta}t}\tilde{a}_j + \text{h.c.}\right)\right.$$
$$\left. + f(t)Z_{\rm c}^{\alpha\beta} + V_0^{\alpha\beta}\right)\Pi^{\alpha\beta}. \tag{23}$$

As the Hamiltonian in the interaction picture is time dependent we use a Magnus expansion for time ordered systems:

$$U_{\rm I}(t) = \exp\left[-i\int_{t_0}^{t}d\tau H_{\rm I}(\tau)\right.$$
$$- \frac{1}{2}\int_{t_0}^{t}d\tau'\int_{t_0}^{\tau'}d\tau\,[H_{\rm I}(\tau'), H_{\rm I}(\tau)]$$
$$+ \frac{i}{6}\int_{t_0}^{t}d\tau''\int_{t_0}^{\tau''}d\tau'\int_{t_0}^{\tau'}d\tau$$
$$\times ([H_{\rm I}(\tau''), [H_{\rm I}(\tau'), H_{\rm I}(\tau)]]$$
$$\left. + [H_{\rm I}(\tau), [H_{\rm I}(\tau'), H_{\rm I}(\tau'')]]) + \cdots\right] \tag{24}$$

Using the commutator relations $\left[\tilde{a}_i, \tilde{a}_j^\dagger\right] = \delta_{ij}$ and $[\tilde{a}_i, \tilde{a}_j] = \left[\tilde{a}_i^\dagger, \tilde{a}_j^\dagger\right] = 0$ of creation and annihilation operators we find

$$[H_{\rm I}(\tau'), H_{\rm I}(\tau)] = \sum_{\alpha\beta} \tag{25}$$
$$\left(-2i\sum_{j=1}^{2}F_j^{\alpha\beta}(\tau')F_j^{\alpha\beta}(\tau)\sin\left[\omega_j^{\alpha\beta}(\tau' - \tau)\right]\right)\Pi^{\alpha\beta}$$
$$[H_{\rm I}(\tau''), [H_I(\tau'), H_I(\tau)]] = 0 \tag{26}$$

The complete time evolution operator is thereby:

$$U_{\rm I}(t) = \sum_{\alpha\beta}\prod_{j=1}^{2}\left[\mathcal{D}\left(A_j^{\alpha\beta}(t)\right)\right]$$
$$\times \exp\left[i\sum_{j=1}^{2}\varphi_j^{\alpha\beta}(t) - i\Phi_{\rm e}^{\alpha\beta}(t)\right]\Pi^{\alpha\beta} \tag{27}$$

with

$$A_j^{\alpha\beta}\left(f(t),\omega_j^{\alpha\beta},t\right) = \int_{t_0}^{t} d\tau F_j^{\alpha\beta}(\tau) e^{i\omega_j^{\alpha\beta}\tau}, \tag{28}$$

$$\Phi_{\mathrm{e}}^{\alpha\beta}\left(f(t),\omega_j^{\alpha\beta},t\right) = \int_{t_0}^{t} d\tau f(t) Z_{\mathrm{c}}^{\alpha\beta} + V_0^{\alpha\beta}, \tag{29}$$

$$\varphi_j^{\alpha\beta}\left(f(t),\omega_j^{\alpha\beta},t\right) = \int_{t_0}^{t} d\tau' \int_{t_0}^{\tau'} d\tau \left[F_j^{\alpha\beta}(\tau') F_j^{\alpha\beta}(\tau) \times \sin\left(\omega_j^{\alpha\beta}(\tau'-\tau)\right)\right]. \tag{30}$$

Gate fidelity

We analyze the time evolution of the electronic basis states $|\alpha\beta\rangle = \{|\downarrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\uparrow\uparrow\rangle\}$. As the states $|\downarrow\uparrow\rangle$ and $|\uparrow\downarrow\rangle$ are symmetric, we only consider state $|\uparrow\downarrow\rangle$. An ideal controlled phase gate has the evolution operator

$$U_{\mathrm{CP}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}. \tag{31}$$

The state fidelity without lifetime limit is defined as the overlap $F = |\langle\Psi(0)| U_{\mathrm{CP}} U_I(T) |\Psi(0)\rangle|^2$, which we evaluate here explicitly for the two ion superposition state $|\Psi(0)\rangle = 1/2\left[(|\downarrow\rangle + |\uparrow\rangle) \otimes (|\downarrow\rangle + |\uparrow\rangle)\right]$ with the ion crystal initially in the motional ground state. For a constant driving field we obtain:

$$F = \frac{1}{16}\left| \exp\left[-\frac{\left|A_1^{\downarrow\downarrow}\right|^2}{2} + i\left(\varphi_1^{\downarrow\downarrow} + \Phi_{\mathrm{e}}^{\downarrow\downarrow}\right)\right] + 2\exp\left[-\frac{\left|A_1^{\uparrow\downarrow}\right|^2}{2} - \frac{\left|A_2^{\uparrow\downarrow}\right|^2}{2} + i\left(\varphi_1^{\uparrow\downarrow} + \varphi_2^{\uparrow\downarrow} + \Phi_{\mathrm{e}}^{\uparrow\downarrow}\right)\right] - \exp\left[-\frac{\left|A_1^{\uparrow\uparrow}\right|^2}{2} + i\left(\varphi_1^{\uparrow\uparrow} + \Phi_{\mathrm{e}}^{\uparrow\uparrow}\right)\right]\right|^2. \tag{32}$$