Ultrafast energy exchange between two single Rydberg atoms on the nanosecond timescale

Y. Chew,1,2 T. Tomita,1 T. P. Mahesh,1,2 S. Sugawa,1,2 S. de Léséleuc,1,2,∗ and K. Ohmori1,2,†

1Institute for Molecular Science, National Institutes of Natural Sciences, Okazaki 444-8585, Japan
2SOKENDAI (The Graduate University for Advanced Studies), Okazaki 444-8585, Japan

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Rydberg atoms, with their giant electronic orbitals, exhibit dipole-dipole interaction reaching the GHz range at a distance of a micron, making them a prominent contender for realizing quantum operations well within their coherence time. However, such strong interactions have never been harnessed so far, mainly because of the stringent requirements on the fluctuation of the atom positions and the necessary excitation strength. Here, using atoms trapped in the motional ground-state of optical tweezers and excited to a Rydberg state with picosecond pulsed lasers, we observe an interaction-driven energy exchange, i.e., a Förster oscillation, occurring in a timescale of nanoseconds, two orders of magnitude faster than in any previous work with Rydberg atoms. This ultrafast coherent dynamics gives rise to a conditional phase which is the key resource for an ultrafast controlled-Z gate. This opens the path for quantum simulation and computation operating at the speed-limit set by dipole-dipole interactions with this ultrafast Rydberg platform.

I. INTRODUCTION

Progress in the field of quantum simulation and computation is fueled by efforts made on a variety of platforms (e.g., superconducting (SC) qubits, quantum dots, trapped ions, neutral atoms, ...) to reach a critical fidelity of quantum operations. This requires to operate entanglement between two neutral atoms separated by a microscopic distance (i.e., a Förster oscillation, occurring in a timescale of nanoseconds), two orders of magnitude faster than in any previous work with Rydberg atoms. This ultrafast coherent dynamics gives rise to a conditional phase which is the key resource for an ultrafast controlled-Z gate. This opens the path for quantum simulation and computation operating at the speed-limit set by dipole-dipole interactions with this ultrafast Rydberg platform.

Arrays of Rydberg atoms in optical tweezers are one of the most exciting systems for quantum simulation [24–27] and computation [28–30]. At its core, the dipole-dipole interaction $\hat{H}_{\text{dip}} \sim d_{1}d_{2}/4\pi\varepsilon_{0}R^{3}$ [31, 32] is used to operate entanglement between two neutral atoms separated by a microscopic distance $R$ thanks to the large matrix elements of the dipole operator $\hat{d}$ ($\sim 1000 e\alpha_{0}$). For two Rydberg atoms with principal quantum number $n = 40$, distant by $R \sim 1 \mu\text{m}$ (to trap them in independent tweezers), the coupling strength between pairs of orbitals reaches $J = \langle r'r''|\hat{H}_{\text{dip}}|rr\rangle \sim 2\pi \times 1 \text{GHz}$, setting the speed limit of entanglement at $t_{J} \sim 1 \text{ns}$, five orders of magnitude faster than the $100 \mu\text{s}$ radiative lifetime of Rydberg states, as shown in Fig. 1(a). Currently, the best entangling protocols of Rydberg atoms [28–30] are performed deeply in the adiabatic regime compared to the available interaction strength, with a typical duration of $0.5 \mu\text{s}$, as they rely on Rydberg blockade [33]. In this scenario, the interaction strength, either the dipole-dipole coupling $J$, or more often the weaker second-order van der Waals shift $V$, is larger than the coupling $\Omega_{\text{cw}}$ from ground to Rydberg states with continuous-wave (cw) lasers, such that excitation of two atoms is prohibited. This gives rise to entanglement in a timescale $\pi/\Omega_{\text{cw}} \gg t_{J}$, with a gate speed technically limited by the available power of cw lasers. This approach is motivated by the suppression of population of the strongly interacting state $|rr\rangle$, thus avoiding (i) leakage into other Rydberg states $|r'r''\rangle$, and (ii) motional coupling to the atoms’ external d.o.f. $(\hat{x}, \hat{p})$ caused by the distance-dependent dipole-dipole interaction. Interestingly, other communities have learned to minimize exactly these effects (e.g., leakage to non-computational states in SC qubits [20], motional coupling in ion traps [15]), while approaching the entanglement speed limit [10, 17, 18, 21–23]. On the other hand, the “slow” blockade gate requires to spend a relatively long time in Rydberg states affected by: finite lifetime, black-body radiation [34], Doppler effect, laser phase noise [35, 36], electric field fluctuation, and motional coupling due to different trapping potentials of Rydberg atoms [37, 38].

Here, we take a first step into achieving an entanglement protocol with Rydberg atoms at the speed limit set by the dipole-dipole interaction. Two single $^{87}\text{Rb}$ atoms separated by a distance as small as $1.5 \mu\text{m}$ are both excited to a $nD$ Rydberg state by using pulsed lasers with a duration of tens of picoseconds [39, 40]. This is faster than the timescale of interaction $J$ but slow enough to resolve the Rydberg orbitals whose splitting

* sylvain@ims.ac.jp
† ohmori@ims.ac.jp
each experiment. We post-select the runs where one or more Rb atoms are randomly loaded in the traps and detected by taking fluorescence images at the beginning and end of each trap. The traps were randomly distributed within 15% of the diffraction theory limit. Single atomic distance $R_{\text{cw}}$ coupling $\Omega$ is then focused with a high-NA objective lens shown in Fig. 1(b). The array consists of pairs of traps with an adjustable spacing ranging from 1.5 $\mu$m to 5 $\mu$m [Fig. 1(c,d)], obtained by summing two holograms: one computed by the weighted Gerchberg-Saxton algorithm [Fig. 1(c,d)], obtained by summing two holograms: one computed by the weighted Gerchberg-Saxton algorithm to generate a regular 2D array and a binary phase grating with tunable period to diffract each single trap into a pair. The measured trap depth is 0.62 mK and the trap lifetime is then focused with a high-NA objective lens shown in Fig. 1(b). A first 780 nm laser pulse (duration: 2 ps, bandwidth: 700 GHz) drives the Rydberg transition with $\Omega R/\hbar = 2 \pi \times (147, 117, 35)$ kHz. From these values, we calculate a tweezers waist of 0.53/0.62 $\mu$m [43] and a Rayleigh length of 1.56 $\mu$m, within 15% of the diffraction theory limit. Single atoms are randomly loaded in the traps and detected by taking 10 ms fluorescence images at the beginning and end of each experiment. We post-select the runs where one or two traps of a given pair are loaded when necessary.

By operating in a regime where the dynamics is driven by the dipole-dipole interaction $J = C_3/R^3$ (in contrast to the blockade regime where it is driven by the laser coupling $\Omega_{\text{cw}}$), it becomes crucial to control the interatomic distance $R$ and to minimize its uncertainty $\Delta R$ as it translates into an interaction noise $\Delta J / J = 3 \Delta R / R$. Usually, experiments with Rydberg atoms in tweezers are performed with “hot” atoms ($T \sim 30 \mu$K) whose position fluctuates by $\sqrt{k_B T / \hbar} \sim 100$ nm (500 nm along $z$), either because they rely on Rydberg blockade or because large distances ($R \sim 10 \mu$m) are used [24, 44, 45]. For us, these thermal fluctuations give an unacceptable noise $\Delta R_{\text{th}} / R \sim 10\%$. Here, we successfully suppress them by applying Raman sideband cooling to bring the atoms into the motional ground-state of the tweezers [46, 47].

From the sideband spectra shown in Fig. 1(e), we extract the mean motional quanta $\bar{n} = (0.11, 0.11, 0.56)$ giving a position spread $\sqrt{\bar{n} + 1/2} \sqrt{\theta / \hbar / \omega} = (22, 25, 60)$ nm. This translates into an uncertainty $\Delta R_{\text{qu}} = 35$ nm ($\Delta R_{\text{qu}} / R < 2\%$) dominated at 90% by zero-point quantum fluctuations. We emphasize that, in this limit, the coupling with the external d.o.f. is mostly coherent.

We now briefly describe the ultrafast ($\sim 100$ ps) coherent excitation of Rydberg atoms [39, 40] with two single-photon Rabi $\pi$-pulses from the ground state $|g\rangle = |S_{1/2}\rangle$ to the intermediate state $|e\rangle = |P_{3/2}\rangle$, and then up to the Rydberg state $|d\rangle = |4D_{5/2}\rangle$, as depicted in Fig. 1(b). A first 780 nm laser pulse (duration: 2 ps, bandwidth: 700 GHz) drives the $g \leftrightarrow e$ transition with its energy set to achieve a Rabi $\pi$-pulse, thus fully transferring the valence electron to the intermediate level. After tens of picoseconds, a 480 nm pulse drives the $e \leftrightarrow d$ transition. To account for the finite energy splitting of the Rydberg series [see Fig. 2(a)], we cut the 480 nm bandwidth down to 100 GHz (14 ps) to avoid exciting the nearby $42D$ and $44D$ levels, as checked by numerical simulations. To achieve sufficient driving strength we focus

![FIG. 1. Ultrafast Rydberg platform.](image_url)
the 480 nm beam to a diameter of 20 μm; consequently only a single column of the array, shown in Fig. 1(c), experiences the maximum intensity. We succeed in driving more than 2π cycles on the e ↔ d transition with a single pulse, or two π-pulses in a pump-probe configuration. Large pulse-to-pulse energy fluctuation of the 480 nm laser currently limits the π-pulse fidelity to 75%.

The excitation is performed in ~ 100 ps (dominated by the delay between the two pulses), fast enough to neglect spontaneous emission from the 5P orbital (25 ns), as well as interaction between Rydberg states. We also ensure that the electronic \( m_S \) and nuclear \( m_I \) spin degrees of freedom do not mix with the electronic orbital \( m_L \) after the excitation. To this end, we initially prepare the atoms in the spin-polarized state \(|5S_{1/2}, F = 2, m_F = 2\rangle = |5S, m_L = 0\rangle |m_S = 1/2, m_I = 3/2\rangle \) and adjust the two excitation lasers to be \( \sigma^+ \)-polarized to prepare the state \(|d\rangle = |43D, m_L = 2\rangle |m_S = 1/2, m_I = 3/2\rangle \) (this also forbids the excitation of S-orbitals). From now on, we drop the spin degrees of freedom as they will not mix with the Rydberg orbital dynamics.

### III. ULTRAFAST FÖRSTER OSCILLATION

Following the excitation, a pair of atoms in \(|dd\rangle\) experiences the effect of the dipole-dipole Hamiltonian \( \hat{H}_{\text{dip}} \), which is dominated by the coupling to the pair state \(|pf\rangle = |45P; 41F\rangle\), as shown in Fig. 2. The detuning \( \Delta E \) of this channel is accidentally small in \(^{87}\text{Rb}\) (−8 MHz), giving rise to the so-called Förster oscillation between \(|dd\rangle\) and the quasi-resonant symmetric state \(|\bar{p}f\rangle = (|pf\rangle + |fp\rangle)/\sqrt{2} \) with a coupling \( J = \sqrt{2}C_3/R^3 \). With increasing interaction time \( t \), the atoms evolve into \( |\Psi(t)\rangle = \cos(Jt) |dd\rangle - i \sin(Jt) |\bar{p}f\rangle \) and at \( t = \pi/J \), they are back into \(-|dd\rangle\) having acquired a π-phase shift. A refined description of the system requires to account for the finite size \( \Delta R_{\text{au}} \) of the wavefunction \( \psi(R) \) describing the relative position of the two atoms, leading to the following entangled state of internal and external d.o.f.:

\[
|\Psi(t)\rangle = \int dR \psi(R) \left[ \cos \left( \frac{\sqrt{2}C_3}{R^3} t \right) |R; dd\rangle - i \sin \left( \frac{\sqrt{2}C_3}{R^3} t \right) |R; \bar{p}f\rangle \right]
\]

Other channels or degrees of freedom could perturb this ideal dynamics. We find that \(|44P; 42F\rangle\) is the next relevant channel, however for \( R > 1.5 \mu m \) the coupling remains weaker than the energy difference, only giving a weak non-resonant perturbation, i.e., a van der Waals shift \( V \) [Fig. 2(c)]. Other dipole-dipole channels, as well as higher-order ones (e.g., dipole-quadrupole) are further negligible: they are taken into account in numerical simulations but are left aside of the discussion. We now consider the projection of orbital angular momentum \( m_L \), whose dynamics is in general non-trivial as \( \hat{H}_{\text{dip}} \) contains terms changing each atom angular momentum [42] by \( \Delta m_L = 0, \pm 1 \). As shown in Fig. 2(b), with our choice of initial state and by aligning the pair of atoms with the quantization axis, the resonant coupling gives a splitting \( J = \sqrt{2}C_3/R^3 \) between the two eigenstates (solid lines), while the off-resonant channel adds a van der Waals shift \( V = - (\sqrt{2}C_3/R^3)^2 / \Delta E \) (dashed lines). The probability distribution of inter-atomic distance \( |\psi(R)|^2 \) is shown for the measured \( \Delta R_{\text{au}} = 35 \text{ nm} \).

![FIG. 2. Rydberg levels and dipole-dipole couplings. (a,b) Single-atom spectrum showing dipole matrix elements \( d \) from state 43D (a), and the different \( m_L \) sublevels (b), highlighting the selection rule for a pair of atoms aligned with the quantization axis. Atomic orbitals for \( m_L = L \) are represented. (c) Two-atom spectrum obtained by diagonalizing \( \hat{H}_{\text{dip}} \). The inset shows the coupling \( |C_3| = d_1d_2/4\pi\epsilon_0 \) and energy difference \( \Delta E \) between the relevant pair-states. The resonant coupling gives a splitting \( J = \sqrt{2}C_3/R^3 \) between the two eigenstates (solid lines), while the off-resonant channel adds a van der Waals shift \( V = - (\sqrt{2}C_3/R^3)^2 / \Delta E \) (dashed lines). The probability distribution of inter-atomic distance \( |\psi(R)|^2 \) is shown for the measured \( \Delta R_{\text{au}} = 35 \text{ nm} \).](image-url)
FIG. 3. Ultrastar Förster oscillation. (a) Pump-probe sequence. (b–e) Probability $P_d$ to be in state $|d⟩$ after a delay $t = 2.29$ ns (b,c) or $t = 6.51$ ns (c,e). The oscillation is shown as a function of the interatomic distance $R$ (b,d), or the interaction area $J_t = \sqrt{2C_3t/R^3}$ (c,f). The black curves are simulations of the ideal Förster oscillation rescaled by the SPAM errors (dotted), including the effect of quantum uncertainty in position (dashed), and perturbing interaction channels (solid). In (e), we show the data of all 16 pairs of atoms, while this is averaged in (b–d). For each distance, we repeat the experiment 2000 times, out of which we select the $\sim 25\%$ shots where both atoms of a pair are loaded, resulting in a quantum projection noise $\lesssim 3\%$ for each point in (e), and $\lesssim 1\%$ in (b–d). Display the oscillation as a function of the interaction area $J_t = \sqrt{2C_3t/R^3}$ [Fig. 3(c,e)]. Satisfyingly, the period matches the ab-initio theory (dotted curves) for the two datasets, clearly demonstrating that the dynamics is coherently driven by the Förster channel.

We now discuss the finite contrast and damping of the oscillation. The 40 $\%$ contrast is caused by state preparation and measurement (SPAM) errors, i.e., imperfect $\pi$-pulse in the excitation and de-excitation processes, and is not related to the Förster oscillation. These errors are independently measured and used to rescale the contrast of all simulations, giving good agreement with the experimental data. Concerning the damping, a first source is the quantum uncertainty of the atoms’ position in their traps. Calculations (dashed curves), consisting in tracing over the external d.o.f. in Eq. (1), demonstrate that this effect is indeed responsible for most of the damping for large values of $J_t$, as clearly seen in Fig. 3(e). The effect of perturbing channels (solid lines) effectively gives an additional damping, which is more pronounced in Fig. 3(c), where we use smaller distances increasing the perturbation. By plotting data points for each pair of trap, we also notice a horizontal scatter increasing with $J_t$. These errors in the measured distance $R$ are most likely caused by our assumption that atoms in a pair are exactly in the same $z$-plane [48]. Eventually, there remains a slight discrepancy between the measured damping and our simulations. We attribute this to imperfect polarization of the excitation lasers leading to a coupling of the ideal Förster dynamics with the $m_L$ and $m_S$ degrees of freedom. These last two points could be addressed in future experiments by devising independent in-situ measurements.

IV. CONDITIONAL PHASE

We then investigate the conditional phase $\phi$ imprinted on a pair of atoms. To this end, we now consider atoms initialized in a coherent superposition $|e⟩ + |d⟩$ of the in-
intermediate and Rydberg state. The interaction drives the state of atom 1 of a pair into $|e⟩ + |e^{i\phi}⟩|d⟩$ when atom 2 in state $|d⟩$. Ideally, $|e^{i\phi}⟩ = \cos(Jt)$ would realize a controlled-Z gate at $Jt = \pi$. Having previously measured the population $|c|^2$, we now gain sensitivity to the phase $\phi$ by performing a Ramsey pump-probe experiment [39, 49]. As depicted in Fig. 4(a), a first $\pi/2$-pulse prepares the coherent state, which is then probed after an interaction time $t = 6.51$ ns with a second $\pi/2$-pulse whose arrival time is scanned with attosecond precision over $3$ fs to realize a variable phase $\theta$. The Ramsey signal given by atom 1, conditioned on the state of atom 2, could be observed with single-atom addressing techniques [9, 29]. Here, Ramsey fringes are recorded irrespective of the state of atom 2 [45, 50], thus giving a signal oscillating as $\cos(\theta) + |c⟩\cos(\theta + \phi) = C_R \cos(\theta + \theta_R)$ with a relative contrast $C_R$ and phase shift $\theta_R$ from fringe fringes obtained with a single atom. In the ideal case, the interaction produces a maximally-entangled state at $Jt = \pi$ ($C_R = 0$), and returns the atoms to a pure product state at $Jt = 2\pi$ ($C_R = 1$, $\theta_R = 0$). While the measured fringes shown in Fig. 4(b) approximately follow this simple scenario, there are clear deviations: the contrast is imperfect and there are phase shifts.

To capture these, we need to account for the van der Waals shift $V$ caused by the off-resonant channels, and the natural energy defect $\Delta E$ of the resonance. In the limit $V$, $\Delta E \ll J$, we can derive $|e^{i\phi}⟩ \approx \cos(Jt)e^{-i(V+\Delta E)t/2}$, which is represented on the Bloch sphere. We then show, in Fig. 4(c), the expected Ramsey contrast and phase shift as a function of the interaction area $Jt$ as two atoms are brought closer. We now obtain an excellent agreement with the measurements, supporting that we accurately capture the evolution of the conditional phase $\phi$. For our experimental parameters, we calculate that $\phi = 1.17 \pi$, thus overshooting the ideal $\pi$-phase shift. This could be corrected by tuning the energy of the resonant $|pf⟩$ state to $\Delta E = -V$, for example with a weak electric field ($< 1 \text{ V/cm}$) applying a Stark shift [41]. Finally, we point that a related interaction-driven procedure was used by Jo et al. [45] to obtain a conditional phase between Rydberg atoms at a distance $R \sim 10 \text{ \mu m}$, using a pure van der Waals coupling, in a time $\pi/V = 2.6 \text{ \mu s}$. Here, we demonstrate a 400 times faster dynamics thanks to the use of ultrafast pulsed-laser technology to excite Rydberg states in tens of picoseconds, and much closer distances bringing the interaction strength towards the GHz range.

V. DISCUSSION AND CONCLUSION

The next step towards an ultrafast $C_Z$ gate would be to improve the fidelity of the pulsed excitation; and to map a long-lived hyperfine qubit to a ground-Rydberg superposition, which can also be achieved in an ultrafast timescale set by the 6.8 GHz ground-state hyperfine splitting [6]. Errors originating from perturbing channels, such as leaks in other pair-states $|r^i r^j⟩$ or dynamical phases, can be eliminated by fine-tuning the energy levels using electric fields (dc-Stark shift) or non-resonant laser or microwave fields (ac-Stark shift), to achieve synchronization of maximal entanglement and minima of leaks, as performed with SC qubits [10, 21–23]. The fundamental source of infidelity for an interaction-driven gate is entanglement with the external d.o.f. of the atoms, with the quantum uncertainty in distance leading here to a finite probability for the atoms to remain in the resonant $|pf⟩$ state of $|\sin(Jt)|^2 \approx (3\pi\Delta R/R)^2 = 1.8\%$. We envision that advanced coherent control techniques can further suppress such errors. The motional coupling can be reduced by an order of magnitude by preparing squeezed states of motions [51, 52], and even further by using a decoupling echo-like sequence [16]. One could also dynamically adjust the Förster resonance defect $\Delta E$ to perform robust fast adiabatic gates [20].

In summary, we have observed an ultrafast quasi-resonant energy exchange between two single Rydberg atoms trapped in individual tweezers, with an interstate control with a precision limited by quantum fluctuations. The interaction-driven dynamics gives rise to a calculated conditional phase $\phi = 1.17 \pi$, in good agreement with observations. This is the key resource for an ultrafast two-qubit gate operating at the fundamental speed-limit set by the dipole-dipole Hamiltonian, and not by technical limitations set by the available power of cw lasers. Such a gate would be two orders of magnitude faster than with state-of-the-art protocols based on the blockade mechanism, thus evading decoherence mechanisms affecting Rydberg atoms on a timescale of tens of microseconds. Furthermore, our experimental platform could be used to simulate the dynamics of interacting spin-1 models by bringing more than two atoms at close distance, to investigate Rydberg interactions when orbitals of neighbor atoms overlap [40], or the use of Rydberg wavepackets instead of single Rydberg levels.

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AUTHOR CONTRIBUTIONS

Y.C., T.T., T.P.M. and S.d.L. designed and carried out the experiments. Y.C. and S.d.L. performed data analysis, theory and simulations, and wrote the manuscript with contributions from all authors. S.d.L. and K.O. supervised and guided this work.

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