Deterministic Time-Bin Entanglement between a Single Photon and an Atomic Ensemble

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Hybrid matter-photon entanglement is the building block for quantum networks. It is very favorable if the entanglement can be prepared with a high probability. In this paper, we report the deterministic creation of entanglement between an atomic ensemble and a single photon by harnessing Rydberg blockade. We design a scheme that creates entanglement between a single photon's temporal modes and the Rydberg levels that host a collective excitation, using a process of cyclical retrieving and patching. The hybrid entanglement is tested via retrieving the atomic excitation as a second photon and performing correlation measurements, which suggest an entanglement fidelity of 87.8%. Our source of matter-photon entanglement will enable the entangling of remote quantum memories with much higher efficiency.

The construction of quantum networks [1–3] over long distance requires matter qubits with an efficient optical interface, since photons are an excellent candidate for long-distance transmission, and matter qubits are necessary to connect different segments. A paradigm for this situation is an entanglement pair between a single photon and a matter qubit. For application in quantum networks, it is necessary that the hybrid entanglement has a very high preparation efficiency and fidelity, at the same time the matter qubit can be preserved for a long duration [4].

It is very promising of harnessing atomic ensembles [4–6] for quantum networks, since an ensemble of atoms are relatively easier to prepare in comparison with single atoms, and it has additional advantage of collective enhancement, which enables efficient interaction with single photons. In the past years, plenty of efforts have been paid to improving the performances of laser-cooled atomic ensembles [7–29]. Notably, very high retrieval efficiency has been achieved either by using large optical depth [16, 22, 24, 27] or with the assistance of a low-finesse cavity [7, 13, 17, 29], sub-second storage has been achieved by confining the atoms with optical lattice and compensating the differential light shift [11, 12, 17, 29], telecom frequency conversion has also been employed to extend distance of remote entanglement [28].

When atomic ensembles are employed as quantum memories, they are typically modeled as non-interacting atoms, and collision between atoms are unfavorable. In this situation, the preparation of entanglement with a single photon is probabilistic, and the preparation probability is kept very low typically to minimize the contribution of high-order events [4, 5]. However, Rydberg interaction provides a promising solution to create entanglement deterministically [30, 31], as a single atom excited in a Rydberg state can block further excitations into the Rydberg state [32, 33]. In recent years, plenty of research have been performed on Rydberg quantum

optics [34–49]. In particular, L. Li *et al* [38] demonstrated entanglement between a Rydberg excitation and a number-state encoded light field. The number-state encoding suffers from being phase sensitive. To avoid this, J. Li *et al* realized entanglement between a Rydberg excitation and a polarization encoded single photon [48]. This scheme nevertheless succeeds merely 50% in principle, limiting its efficiency in entangling remote nodes [28, 50] via entanglement swapping [51].

In this paper, we propose and experimentally realize a scheme that entangles an atomic ensemble with a single photon's time-bin modes deterministically. The detailed scheme is depicted in Fig. 1. We consider a mesoscopic atomic ensemble in the regime of full blockade [33]. All atoms initially stay in a ground state $|g\rangle$. Two Rydberg levels $|r_1\rangle$ and $|r_2\rangle$ are employed to host a collective excitation. Our scheme starts from a Rydberg state preparation phase, in which we first apply a collective $\pi/2$ pulse coupling $|g\rangle$ and $|r_1\rangle$ resulting in a collective state of $(|R_1\rangle + |G\rangle)/\sqrt{2}$, and apply a subsequent π pulse coupling $|g\rangle$ and $|r_2\rangle$ resulting in a superposition state of $(|R_1\rangle + |R_2\rangle)/\sqrt{2}$, where $|G\rangle$ denotes all atoms in $|g\rangle$ and $|R_j\rangle = N^{-1/2} \sum_{i=1}^{N} |g^1 g^2 \cdots r_j^i \cdots g^N\rangle$ denotes a collective excitation in $|r_j\rangle$, with N being number of atoms and i being an atom index. We name the 2nd π pulse as a "patching" pulse, since it creates a full excitation out of a half excitation, harnessing Rydberg blockade between $|r_1\rangle$ and $|r_2\rangle$. In the next phase of our scheme, atom-photon entanglement is generated via a cyclical process of "retrieving and patching". We first retrieve $|R_1\rangle$ to a single photon and patching it back by applying a π pulse coupling $|g\rangle$ and $|r_1\rangle$. Rydberg blockade between $|r_1\rangle$ and $|r_2\rangle$ again plays a key role, ensuring that a $|R_1\rangle$ excitation is recreated only if there was a $|R_1\rangle$ excitation previously. Afterwards, we apply the retrieving and patching process for $|R_2\rangle$, creating a photon in a delayed temporal mode. After these steps, the joint atom-photon
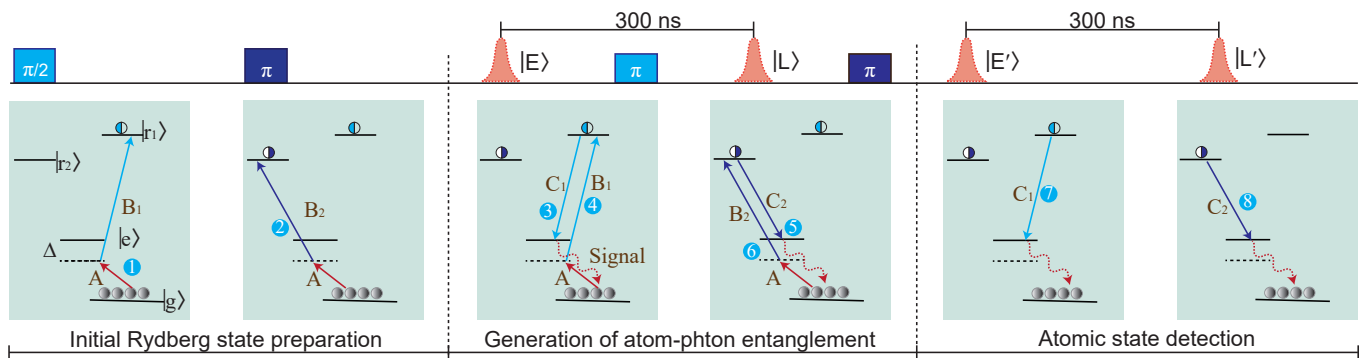
FIG. 1. Scheme of generating deterministic time-bin entanglement. In the first phase, we prepare a Rydberg superposition state $(|R_1\rangle + |R_2\rangle)/\sqrt{2}$. In the second phase, we apply the cyclical "retrieving and patching" process for $|R_1\rangle$ and $|R_2\rangle$ in sequence, which results in retrieval photon whose temporal mode is entangled with the Rydberg levels of the collective excitation. In the last phase, we detect the atomic state via retrieving it to the temporal mode of a second photon. The excitation and patching process involve a two-photon Raman process. Spatial arrangement of the Raman beams is shown in Fig. 2. Numerically filled circles indicate relative time sequences of all the steps.

state can be expressed as

$$|\Psi_{ap}\rangle = (1/\sqrt{2})(|R_1\rangle|E\rangle + |R_2\rangle|L\rangle), \qquad (1)$$

where $|E\rangle$ or $|L\rangle$ denotes a single photon in the temporal early or late mode. We note that all the atomic manipulation steps are deterministic. The retrieving process from a Rydberg excitation to a single photon [52] is also deterministic in principle, as long as the optical depth is high enough. Thus we claim that our scheme is deterministic, which differentiates from the traditional DLCZ scheme [5] that creates entanglement probabilistically through spontaneous Raman scattering. In the last phase, we retrieve the atomic state $|R_1\rangle$ and $|R_2\rangle$ in sequence to a second photon in the temporal mode of $|E'\rangle$ and $|L'\rangle$ to verify the entanglement.

Our experimental setup is illustrated in Fig. 2. An ensemble of ^{87}Rb atoms is captured by a conventional magneto-optical trap (MOT) lasting 30 ms, and a following dark spontaneous force optical trap (dark SPOT) [54, 55] lasting 70 ms. Afterwards, the atoms are loaded into an optical dipole trap formed by a 1064 nm laser propagating along the y direction with a power of 2 W and a waist of 5 μm in the x direction and 25 μm in the z direction, resulting in an optical depth of 1.7 along the x direction and a density of 10^{11} cm^{-3}. The energy levels include a ground state $|g\rangle = |5S_{1/2}, F = 2, m_F = +2\rangle$, an intermediate state $|e\rangle = |5P_{1/2}, F = 1, m_F = +1\rangle$, and two Rydberg states $|r_1\rangle = |81S_{1/2}, m_j = +1/2\rangle$ and $|r_2\rangle = |81S_{1/2}, m_j = -1/2\rangle$. We employ a two-photon Raman process to excite an atom to a Rydberg level, involving a 795 nm laser and a 474 nm laser. Single-photon detuning is set to $\Delta = -40$ MHz. The two Rydberg levels are selected to be close to each other. Thus we are able to share the same 474 nm laser instead of seeking a second blue laser for the other Rydberg state. Determined by the phase-matching condition [52], retrieved

single photons propagate in the x direction, which is the same as the 795 nm Raman laser. At the same time, the single photons also have the same polarization as the Raman laser, thus slight leakage of the Raman laser will spoil the single photons. In our experiment, we solve this problem by using a temporal filter involving an acousto-optic modulator (AOM$_1$ in Fig. 2b)) to deflect the optical field only when retrieving the single photons. Afterwards, the single photon enters an active unbalanced Mach–Zehnder (MZ) interferometer, in which the early mode goes through the long arm while the late mode goes through the short arm. The MZ interferometer is employed not only to measure the first photon but also the second photon retrieved for atomic state detection, which is enabled with an optical switch (AOM$_2$ in Fig. 2b). Finally, the photons are measured with single-photon detectors (SPDs) and analyzed in polarization.

Before characterizing the atom-photon entanglement, it is crucial to verify that the "patching" process is coherent and a Rydberg excitation is genuinely created in a superposition state. Therefore, we first perform an experiment that retrieves the atomic excitation immediately after its creation. In other words, we skip the second phase in Fig. 1. Thus the initial atomic state $(|R_1\rangle + |R_2\rangle)/\sqrt{2}$ is directly converted to $(|E'\rangle + e^{i\phi}|L'\rangle)/\sqrt{2}$, with an internal phase ϕ being dependent on the relative phases of B$_1$, B$_2$, C$_1$, and C$_2$. In order to get a stable phase, we make use of an arbitrary function generator (AFG) with dual outputs to control the AOMs for B$_1$/C$_1$ and B$_2$/C$_2$ respectively as shown in the inset of Fig. 2b, which sets a common phase for all the 4 pulses. We perform measurement in the basis of $|\pm\rangle = (|E'\rangle \pm |L'\rangle)/\sqrt{2}$, with the result shown in Fig. 3. We vary the phase of C$_1$ and measure the single photon counts. As expected, the relative counts show as complementary sinusoidal oscillations as a function of phase. By fitting the result, we get
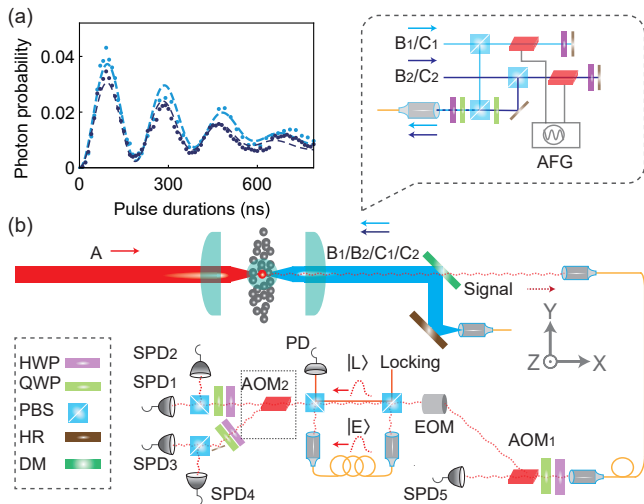
FIG. 2. Experimental setup. (a) Observed collective Rabi oscillations. Light blue dots represent the oscillation between $|G\rangle$ and $|R_1\rangle$ and dark blue dots represent the oscillation between $|G\rangle$ and $|R_2\rangle$. Dashed lines are the fitting results, indicating a π pulse duration of 92.95 ns and 92.18 ns for $|R_1\rangle$ and $|R_2\rangle$ respectively. (b) Layout of the experiment. The 795 nm beam counterpropagates with the 474 nm beams, all of which are tightly focused to address a small region of the atomic cloud to define a mesoscopic ensemble with full blockade (more details available in [53]). Single photons are retrieved in the x direction, afterwards subjected to fiber coupling, temporal filtering, interfering through an unbalanced MZ interferometer, switching, rotating in polarization and final detecting sequentially. The fibers used in the two arms of the the MZ interferometer are 65 m and 5 m which give a relative photon delay of about 300 ns. The interferometer is actively stabilized by injecting a locking beam into the idle port. Our experiment involves four blue beams ($B_1/B_2/C_1/C_2$), which differentiate each other in polarization , frequency, or both. We make use of a multiplexing scheme shown as inset of Fig. 2b to adjust the frequencies dynamically and combine the beams into a single-mode fiber. AOM_2 is merely used for the Bell measurement. Some abbreviations: halfwave plate (HWP), quarterwave plate (QWP), polarizing beamsplitter (PBS), high reflection mirror (HR), dichroic mirror (DM), photodiode (PD), electro-optic modulator (EOM).

FIG. 3. Verification of the initial Rydberg state preparation. $(|R_1\rangle+|R_2\rangle)/\sqrt{2}$ is converted to $(|E'\rangle+e^{i\phi}|L'\rangle)/\sqrt{2}$ and measured in the basis of $(|E'\rangle\pm|L'\rangle)/\sqrt{2}$. Dependence on the internal phase ϕ is plotted. Dashed lines are the fitting results.
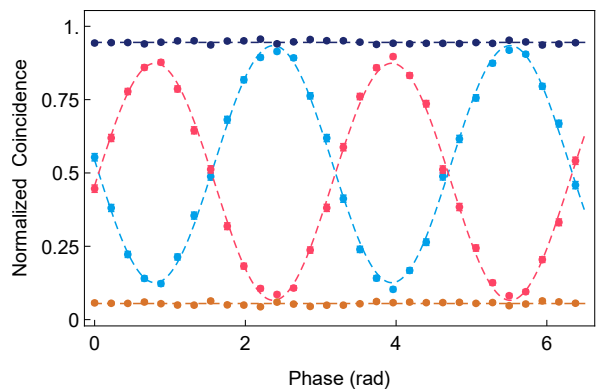
FIG. 4. Characterization of the entanglement $|\Psi_{pp}\rangle$ via correlation measurement. In the eigenbasis, the sum of $|E\rangle|E'\rangle$ and $|L\rangle|L'\rangle$ coincidences is shown in dark blue, while sum of $|E\rangle|L'\rangle$ and $|L\rangle|E'\rangle$ coincidences is shown in orange. In the superposition basis, The sum of $|+\rangle|+'\rangle$ and $|-\rangle|-'\rangle$ coincidences is shown in red, while sum of $|+\rangle|-'\rangle$ and $|-\rangle|+'\rangle$ is shown in light blue. The dash lines are the fitting results.

an interference visibility of $V_{+/-} = 0.909 \pm 0.006$, which is mainly limited by inhomogeneity of the two temporal modes, optical misalignment, and detector afterpulse. The overall measured photon efficiency is about 1.7%, which includes preparation efficiency of a Rydberg excitation ($\sim$90%), retrieval efficiency ($\sim$13%), transmission and fiber coupling efficiency ($\sim$69%), AOM deflecting efficiency ($\sim$77%), transmission efficiency through unbalanced MZ interferometer ($\sim$47%), and detector efficiency ($\sim$60%). We note that by harnessing enhancement with a low-finesse ring cavity [13], the retrieval efficiency may be improved significantly.

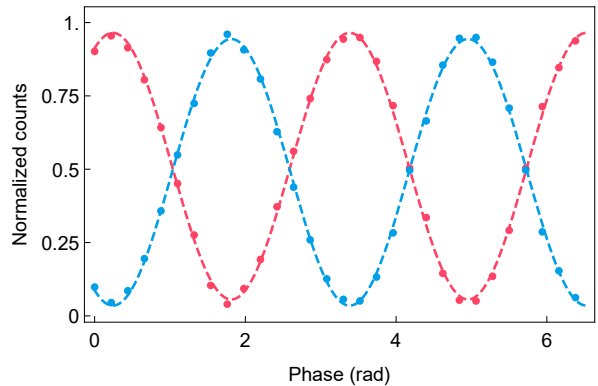Next, we implement the whole scheme shown in Fig. 1. We add back the phase of atom-photon entanglement

generation, by applying the retrieving and patching process for $|R_1\rangle$ and $|R_2\rangle$ in sequence, which creates a first photon that is entangled with atomic excitation in the form of $|\Psi_{ap}\rangle$. To verify the entanglement, we retrieve the atomic excitation as a second photon that entangles with the first photon in the form of $|\Psi_{pp}\rangle = (1/\sqrt{2})(|E\rangle|E'\rangle + e^{i\psi}|L\rangle|L'\rangle)$, with an internal phase ψ dependent on the Raman and retrieving fields. To verify the entanglement, we perform correlation measurement both in the eigenbasis of $|E\rangle/|L\rangle$, and in a superposition basis of $|+\rangle/|-\rangle$. The detectors SPD_1 and SPD_2 are employed for this measurement through temporal multiplexing, and measurement settings are configured by rotating the waveplates. Experimental results are shown in Fig. 4. As expected, when we vary the

internal phase ψ linearly, coincidences in the eigenbasis show no dependence, while coincidences in the superposition basis show as sinusoidal oscillations. By fitting the results, we get a visibility of $V_1 = 0.890 \pm 0.010$ for the eigenbasis, and $V_2 = 0.811 \pm 0.008$ for the superposition basis, which together give an entanglement fidelity as $F \approx (1 + V_1 + 2V_2)/4 = 0.878 \pm 0.005$ [56]. A moderate visibility in the superposition basis contributes most for the entanglement infidelity. Feasible measures for improvement includes using faster switches, using detectors with a lower probability of afterpulse, and using two Rydberg levels of different principal quantum number to reduce crosstalk.

TABLE I. Measurement results for Bell-CHSH test.

α, β	$22.5, 45$	$22.5, 0$	$67.5, 45$	$67.5, 0$
$E(\alpha, \beta)$	-0.415	-0.531	-0.568	0.659
σ_E	0.029	0.028	0.026	0.025

Finally, we verify the entanglement directly via testing the Bell-CHSH inequality [57, 58]. In this test, the two photons need to be measured in different settings, thus the previous detector multiplexing scheme does not work. We make use of an active switcher ($\mathrm{AOM_2}$) to solve this problem. The AOM lets the first photon pass through and directs it to $\mathrm{SPD_1}$ and $\mathrm{SPD_2}$, while deflects the second photon and directs it to $\mathrm{SPD_3}$ and $\mathrm{SPD_4}$. To perform the Bell test, we need to measure the S parameter that is defined as $S = |E(\alpha, \beta) + E(\alpha^*, \beta) + E(\alpha, \beta^*) - E(\alpha^*, \beta^*)|$, with $E(\alpha, \beta)$ being the joint expectation value when one photon is measured in the α setting and the other photon is measured in the β setting. A setting θ refers to a measurement in the basis of $|\theta\rangle = \cos(\theta)|E\rangle + \sin(\theta)|L\rangle$ and $|\theta^\perp\rangle = \sin(\theta)|E\rangle - \cos(\theta)|L\rangle$. $E(\alpha, \beta)$ takes a maximal value 1 for perfect parallel correlation, and -1 for perfect anticorrelation. For perfect entanglement and optimized settings, the S parameter can take a maximal value of $2\sqrt{2}$. The threshold to certify entanglement is $S > 2$. We set the internal phase ψ of $|\Psi_{pp}\rangle$ to 0 and perform the measurements. The results are shown in Tab. I, and we get $S = 2.173 \pm 0.055$, which violates the inequality by 3.16 standard deviations.

In summary, we propose and experimentally realize a scheme of deterministic time-bin entanglement between a single photon and a mesoscopic atomic ensemble. We harness two Rydberg levels to host a collective atomic qubit, and employ a cyclical "retrieving and patching" process to create atom-photon entanglement. Self-blockade in each Rydberg level, and cross blockade between the two levels play key roles for the scheme. Our experiment fully implements the scheme. Via performing correlation measurements, we estimate an entanglement fidelity of 87.8%. We also perform a Bell inequality test to verify the entanglement directly. Further improvements include using a low-finesse cavity to improve the retrieval efficiency [7, 13], transferring the atomic qubit from Rydberg levels to ground state levels [41, 43], and using optical lattice to achieve long-lived storage [17, 29] etc. With these developments, the atom-photon entanglement created in our work may become a strong candidate as a fundamental element for constructing large-scale quantum networks [1–4].

[1] H. J. Kimble, The quantum internet, Nature **453**, 1023 (2008).

[2] C. Simon, Towards a global quantum network, Nature Photonics **11**, 678 (2017).

[3] S. Wehner, D. Elkouss, and R. Hanson, Quantum internet: A vision for the road ahead, Science **362**, eaam9288 (2018).

[4] N. Sangouard, C. Simon, H. de Riedmatten, and N. Gisin, Quantum repeaters based on atomic ensembles and linear optics, Reviews of Modern Physics **83**, 33 (2011).

[5] L.-M. Duan, M. Lukin, J. I. Cirac, and P. Zoller, Long-distance quantum communication with atomic ensembles and linear optics, Nature **414**, 413 (2001).

[6] K. Hammerer, A. S. Sørensen, and E. S. Polzik, Quantum interface between light and atomic ensembles, Reviews of Modern Physics **82**, 1041 (2010).

[7] J. Simon, H. Tanji, J. K. Thompson, and V. Vuletić, Interfacing Collective Atomic Excitations and Single Photons, Physical Review Letters **98**, 183601 (2007).

[8] B. Zhao, Y.-A. Chen, X.-H. Bao, T. Strassel, C.-S. Chuu, X.-M. Jin, J. Schmiedmayer, Z.-S. Yuan, S. Chen, and J.-W. Pan, A millisecond quantum memory for scalable quantum networks, Nature Physics **5**, 95 (2009).

[9] R. Zhao, Y. Dudin, S. Jenkins, C. Campbell, D. Matsukevich, T. Kennedy, and A. Kuzmich, Long-lived quantum memory, Nature Physics **5**, 100 (2009).

[10] S.-Y. Lan, A. G. Radnaev, O. A. Collins, D. N. Matsukevich, T. A. Kennedy, and A. Kuzmich, A multiplexed quantum memory, Optics Express **17**, 13639 (2009).

[11] Y. O. Dudin, A. G. Radnaev, R. Zhao, J. Z. Blumoff, T. A. B. Kennedy, and A. Kuzmich, Entanglement of Light-Shift Compensated Atomic Spin Waves with Telecom Light, Physical Review Letters **105**, 260502 (2010).

[12] A. G. Radnaev, Y. O. Dudin, R. Zhao, H. H. Jen, S. D. Jenkins, A. Kuzmich, and T. A. B. Kennedy, A quantum memory with telecom-wavelength conversion, Nature Physics **6**, 894 (2010).

[13] X.-H. Bao, A. Reingruber, P. Dietrich, J. Rui, A. Dück, T. Strassel, L. Li, N.-L. Liu, B. Zhao, and J.-W. Pan, Efficient and long-lived quantum memory with cold atoms inside a ring cavity, Nature Physics **8**, 517 (2012).

[14] Y. Dudin, L. Li, and A. Kuzmich, Light storage on the time scale of a minute, Physical Review A **87**, 31801 (2013).

[15] Z. Xu, Y. Wu, L. Tian, L. Chen, Z. Zhang, Z. Yan, S. Li, H. Wang, C. Xie, and K. Peng, Long Lifetime

and High-Fidelity Quantum Memory of Photonic Polarization Qubit by Lifting Zeeman Degeneracy, Physical Review Letters **111**, 240503 (2013).

[16] Y.-W. Cho, G. T. Campbell, J. L. Everett, J. Bernu, D. B. Higginbottom, M. T. Cao, J. Geng, N. P. Robins, P. K. Lam, and B. C. Buchler, Highly efficient optical quantum memory with long coherence time in cold atoms, Optica **3**, 100 (2016).

[17] S.-J. Yang, X.-J. Wang, X.-H. Bao, and J.-W. Pan, An efficient quantum light–matter interface with sub-second lifetime, Nature Photonics **10**, 381 (2016).

[18] Y.-F. Pu, N. Jiang, W. Chang, H.-X. Yang, C. Li, and L.-M. Duan, Experimental realization of a multiplexed quantum memory with 225 individually accessible memory cells, Nature Communications **8**, 15359 (2017).

[19] L. Tian, Z. Xu, L. Chen, W. Ge, H. Yuan, Y. Wen, S. Wang, S. Li, and H. Wang, Spatial Multiplexing of Atom-Photon Entanglement Sources using Feedforward Control and Switching Networks, Physical Review Letters **119**, 130505 (2017).

[20] R. Chrapkiewicz, M. Dabrowski, and W. Wasilewski, High-capacity angularly multiplexed holographic memory operating at the single-photon level, Physical Review Letters **118**, 063603 (2017).

[21] B.-S. Shi, D.-S. Ding, and W. Zhang, Quantum storage of orbital angular momentum entanglement in cold atomic ensembles, Journal of Physics B: Atomic, Molecular and Optical Physics **51**, 032004 (2018).

[22] Y.-F. Hsiao, P.-J. Tsai, H.-S. Chen, S.-X. Lin, C.-C. Hung, C.-H. Lee, Y.-H. Chen, Y.-F. Chen, I. A. Yu, and Y.-C. Chen, Highly Efficient Coherent Optical Memory Based on Electromagnetically Induced Transparency, Physical Review Letters **120**, 183602 (2018).

[23] P. Vernaz-Gris, K. Huang, M. Cao, A. S. Sheremet, and J. Laurat, Highly-efficient quantum memory for polarization qubits in a spatially-multiplexed cold atomic ensemble, Nature Communications **9**, 363 (2018).

[24] Y. Wang, J. Li, S. Zhang, K. Su, Y. Zhou, K. Liao, S. Du, H. Yan, and S.-L. Zhu, Efficient quantum memory for single-photon polarization qubits, Nature Photonics , 22 (2019).

[25] W. Chang, C. Li, Y.-K. Wu, N. Jiang, S. Zhang, Y.-F. Pu, X.-Y. Chang, and L.-M. Duan, Long-Distance Entanglement between a Multiplexed Quantum Memory and a Telecom Photon, Physical Review X **9**, 041033 (2019).

[26] B. Jing, X.-J. Wang, Y. Yu, P.-F. Sun, Y. Jiang, S.-J. Yang, W.-H. Jiang, X.-Y. Luo, J. Zhang, X. Jiang, X.-H. Bao, and J.-W. Pan, Entanglement of three quantum memories via interference of three single photons, Nature Photonics **13**, 210 (2019).

[27] M. Cao, F. Hoffet, S. Qiu, A. S. Sheremet, and J. Laurat, Efficient reversible entanglement transfer between light and quantum memories, Optica **7**, 1440 (2020).

[28] Y. Yu, F. Ma, X.-Y. Luo, B. Jing, P.-F. Sun, R.-Z. Fang, C.-W. Yang, H. Liu, M.-Y. Zheng, X.-P. Xie, W.-J. Zhang, L.-X. You, Z. Wang, T.-Y. Chen, Q. Zhang, X.-H. Bao, and J.-W. Pan, Entanglement of two quantum memories via fibres over dozens of kilometres, Nature **578**, 240 (2020).

[29] X.-J. Wang, S.-J. Yang, P.-F. Sun, B. Jing, J. Li, M.-T. Zhou, X.-H. Bao, and J.-W. Pan, Cavity-Enhanced Atom-Photon Entanglement with Subsecond Lifetime, Physical Review Letters **126**, 090501 (2021).

[30] B. Zhao, M. Müller, K. Hammerer, P. Zoller, and

M. Muller, Efficient quantum repeater based on deterministic Rydberg gates, Physical Review A **81**, 052329 (2010).

[31] Y. Han, B. He, K. Heshami, C.-Z. Li, and C. Simon, Quantum repeaters based on Rydberg-blockade-coupled atomic ensembles, Physical Review A **81**, 052311 (2010).

[32] M. D. Lukin, M. Fleischhauer, R. Cote, L. M. Duan, D. Jaksch, J. I. Cirac, and P. Zoller, Dipole Blockade and Quantum Information Processing in Mesoscopic Atomic Ensembles, Physical Review Letters **87**, 037901 (2001).

[33] M. Saffman, T. G. Walker, and K. Mølmer, Quantum information with Rydberg atoms, Reviews of Modern Physics **82**, 2313 (2010).

[34] T. Peyronel, O. Firstenberg, Q.-Y. Liang, S. Hofferberth, A. V. Gorshkov, T. Pohl, M. D. Lukin, and V. Vuletić, Quantum nonlinear optics with single photons enabled by strongly interacting atoms, Nature **488**, 57 (2012).

[35] Y. O. Dudin, L. Li, F. Bariani, and A. Kuzmich, Observation of coherent many-body Rabi oscillations, Nature Physics **8**, 790 (2012).

[36] Y. O. Dudin and A. Kuzmich, Strongly Interacting Rydberg Excitations of a Cold Atomic Gas, Science **336**, 887 (2012).

[37] O. Firstenberg, T. Peyronel, Q.-Y. Liang, A. V. Gorshkov, M. D. Lukin, and V. Vuletić, Attractive photons in a quantum nonlinear medium, Nature **502**, 71 (2013).

[38] L. Li, Y. O. Dudin, and A. Kuzmich, Entanglement between light and an optical atomic excitation, Nature **498**, 466 (2013).

[39] D. Tiarks, S. Baur, K. Schneider, S. Dürr, and G. Rempe, Single-Photon Transistor Using a Forster Resonance, Physical Review Letters **113**, 053602 (2014).

[40] H. Gorniaczyk, C. Tresp, J. Schmidt, H. Fedder, and S. Hofferberth, Single-Photon Transistor Mediated by Interstate Rydberg Interactions, Physical Review Letters **113**, 053601 (2014).

[41] M. Ebert, A. Gill, M. Gibbons, X. Zhang, M. Saffman, and T. G. Walker, Atomic Fock State Preparation Using Rydberg Blockade, Physical Review Letters **112**, 043602 (2014).

[42] M. Ebert, M. Kwon, T. Walker, and M. Saffman, Coherence and Rydberg Blockade of Atomic Ensemble Qubits, Physical Review Letters **115**, 093601 (2015).

[43] J. Li, M.-T. Zhou, B. Jing, X.-J. Wang, S.-J. Yang, X. Jiang, K. Mølmer, X.-H. Bao, and J.-W. Pan, Hong-Ou-Mandel Interference between Two Deterministic Collective Excitations in an Atomic Ensemble, Physical Review Letters **117**, 180501 (2016).

[44] H. Gorniaczyk, C. Tresp, P. Bienias, A. Paris-Mandoki, W. Li, I. Mirgorodskiy, H. P. Büchler, I. Lesanovsky, and S. Hofferberth, Enhancement of Rydberg-mediated single-photon nonlinearities by electrically tuned Förster resonances, Nature Communications **7**, 12480 (2016).

[45] J. D. Thompson, T. L. Nicholson, Q.-Y. Liang, S. H. Cantu, A. V. Venkatramani, S. Choi, I. A. Fedorov, D. Viscor, T. Pohl, M. D. Lukin, and V. Vuletić, Symmetry-protected collisions between strongly interacting photons, Nature **542**, 206 (2017).

[46] H. Busche, P. Huillery, S. W. Ball, T. Ilieva, M. P. A. Jones, and C. S. Adams, Contactless nonlinear optics mediated by long-range Rydberg interactions, Nature Physics **13**, 655 (2017).

[47] D. Tiarks, S. Schmidt-Eberle, T. Stolz, G. Rempe, and S. Dürr, A photon–photon quantum gate based on Ryd-

berg interactions, Nature Physics **15**, 124 (2019).

[48] J. Li, M.-T. Zhou, C.-W. Yang, P.-F. Sun, J.-L. Liu, X.-H. Bao, and J.-W. Pan, Semideterministic Entanglement between a Single Photon and an Atomic Ensemble, Physical Review Letters **123**, 140504 (2019).

[49] S. H. Cantu, A. V. Venkatramani, W. Xu, L. Zhou, B. Jelenković, M. D. Lukin, and V. Vuletić, Repulsive photons in a quantum nonlinear medium, Nature Physics **16**, 921 (2020).

[50] Z.-S. Yuan, Y.-A. Chen, B. Zhao, S. Chen, J. Schmiedmayer, and J.-W. Pan, Experimental demonstration of a bdcz quantum repeater node, Nature **454**, 1098 (2008).

[51] J.-W. Pan, D. Bouwmeester, H. Weinfurter, and A. Zeilinger, Experimental Entanglement Swapping: Entangling Photons That Never Interacted, Physical Review Letters **80**, 3891 (1998).

[52] M. Saffman and T. G. Walker, Creating single-atom and single-photon sources from entangled atomic ensembles, Physical Review A **66**, 065403 (2002).

[53] M.-T. Zhou, J.-L. Liu, P.-F. Sun, Z.-Y. An, J. Li, X.-H. Bao, and J.-W. Pan, Experimental creation of single rydberg excitations via adiabatic passage, Physical Review A **102**, 013706 (2020).

[54] W. Ketterle, K. B. Davis, M. A. Joffe, A. Martin, and D. E. Pritchard, High densities of cold atoms in a dark spontaneous-force optical trap, Physical Review Letters **70**, 2253 (1993).

[55] C. Townsend, N. Edwards, K. Zetie, C. Cooper, J. Rink, and C. Foot, High-density trapping of cesium atoms in a dark magneto-optical trap, Physical Review A **53**, 1702 (1996).

[56] O. Gühne and G. Tóth, Entanglement detection, Physics Reports **474**, 1 (2009).

[57] J. S. Bell, On the Problem of Hidden Variables in Quantum Mechanics, Reviews of Modern Physics **38**, 447 (1966).

[58] J. F. Clauser, M. A. Horne, A. Shimony, and R. A. Holt, Proposed Experiment to Test Local Hidden-Variable Theories, Physical Review Letters **23**, 880 (1969).