

Experimental creation of single Rydberg excitations via adiabatic passageMing-Ti Zhou,^{*} Jian-Long Liu,^{*} Peng-Fei Sun, Zi-Ye An, Jun Li[✉], Xiao-Hui Bao[✉], and Jian-Wei Pan*Hefei National Laboratory for Physical Sciences at Microscale and Department of Modern Physics,**University of Science and Technology of China, Hefei, Anhui 230026, China**and CAS Center for Excellence in Quantum Information and Quantum Physics,**University of Science and Technology of China, Hefei, Anhui 230026, China*

(Received 7 April 2020; accepted 8 June 2020; published 6 July 2020)

In an atomic ensemble, quantum information is typically carried as single collective excitations. It is very advantageous if the creation of single excitations is efficient and robust. A Rydberg blockade enables deterministic creation of single excitations via a collective Rabi oscillation by precisely controlling the pulse area, being sensitive to many experimental parameters. In this paper, we implement the adiabatic rapid passage technique to the Rydberg excitation process in a mesoscopic atomic ensemble. We make use of a two-photon excitation scheme with an intermediate state off-resonant and sweep the laser frequency of one excitation laser. We find the chirped scheme preserves internal phases of the collective Rydberg excitation and is more robust against variance of laser intensity and frequency detuning.

DOI: [10.1103/PhysRevA.102.013706](https://doi.org/10.1103/PhysRevA.102.013706)**I. INTRODUCTION**

Single collective excitations play an essential role in quantum information processing with atomic ensembles [1,2]. In particular, due to the collectively enhanced interaction, the excitations can be efficiently converted to single photons in well-defined modes, thus making them ideally suited for the implementation of quantum light-matter interface [3,4] for quantum repeater [5] and quantum networks [6]. In these applications, it is of great advantage if the excitation creation process is efficient and robust. Following the initial DLCZ protocol [3], spontaneous Raman scattering has become a ubiquitous method of creating single collective excitations. Nevertheless, the creation process is probabilistic, for which the excitation probability has to be kept very low to limit the contribution of high-order excitations. Even though the creation process is heralded by the scattered Raman photon-enabling scalable extension to multiple sources, the low excitation probability will merely result in low-rate applications [1] and impose a demanding requirement on long-lifetime storage [7–11].

To overcome the probabilistic creation issue, one may consider using external deterministic photon sources and storing them in atomic ensembles. In this way, the collective excitations created are deterministic in principle. Nevertheless, the difficulty is shifted to the development of photon sources [12,13] and solving the mismatching issues [14] such as frequency and bandwidth. A more elegant method is to inhibit high-order excitations directly via interactions [15–17]. A Rydberg blockade [18,19] is such a mechanism, which enables creation of a single excitation in a mesoscopic atomic ensemble ($\sim \mu\text{m}$). In this case, an ensemble of atoms behaves as a superatom which undergoes a Rabi oscillation between a ground state and a collective Rydberg state [20].

By fixing the pulse area as π , a single excitation in a Rydberg state can be created deterministically in principle [16,21]. The Rabi frequency is collectively enhanced ($\Omega_N = \sqrt{N}\Omega$, with N for the atom number), leading to difficulty in controlling the pulse area precisely due to the fluctuation of the atom number. Moreover, one also has to control other parameters precisely such as laser intensities and frequency detunings.

Adiabatic passage [22] is a well-known method to realize robust population inversion between two energy levels. For Rydberg atoms, the excitation process typically makes use of a two-photon scheme involving three atomic levels in a ladder configuration [16]. In this case, the technique of stimulated Raman adiabatic passage (STIRAP) [22–24] is very well suited, which achieves population inversion by applying counterintuitive pulses that couple with the top and bottom transitions respectively. This scheme works well for single atoms. For atomic ensembles, it is theoretically found that STIRAP with on-resonance pulses results in a fully dephased collective Rydberg excitation [25] that cannot be retrieved as a single photon, thus it can hardly be useful for the applications that harness collective enhancement. In contrast, the chirped excitation scheme by sweeping the laser frequency provides a promising alternative [26]. In this paper, we experimentally implement this chirped scheme for a mesoscopic atomic ensemble. We find this excitation scheme preserves internal phases of the collective state via observing efficient retrieval of single photons. The chirped scheme is also tested to be more robust than the traditional π -pulse scheme in terms of sensitivity of laser pulse area and frequency detuning.

II. EXPERIMENTAL DESCRIPTION

In our experiment, we make use of laser-cooled rubidium atoms (^{87}Rb), with a temperature of $32 \mu\text{K}$. An ensemble of atoms are confined with an sliced optical dipole trap (1064 nm), with an effective thickness of $6.3 \mu\text{m}$. As shown

^{*}These authors contributed equally to this work.

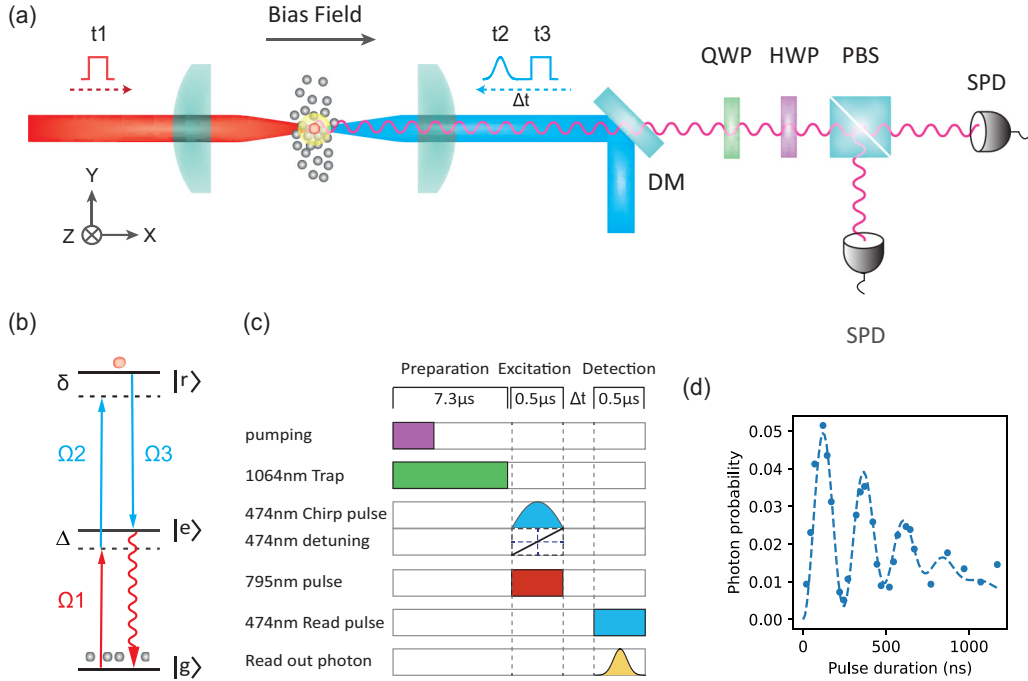


FIG. 1. (a) Experimental layout. The two excitation lasers (Ω_1 and Ω_2) shine the atomic ensemble in opposite directions (x versus $-x$). A read laser Ω_3 shines through the same direction as Ω_2 , leading the retrieval of single photons in the direction of x due to phase matching. A dichroic mirror (DM) is used to couple in the blue lasers and filter the read-out photons. The photons are later rotated with a quarter-wave plate (QWP) and a half-wave plate (HWP) before being split with a polarized beamsplitter (PBS) and detected with single-photon detectors (SPD). (b) Energy levels and laser frequencies. (c) Time sequences of the experiment. (d) Observed collective Rabi oscillations. The sum of detection probabilities is plotted as a function of pulse duration.

in Fig. 1, we make use a two-photon scheme to excite atoms from a ground state ($|g\rangle = |5S_{1/2}, F = 2, m_F = +2\rangle$) to a Rydberg state ($|r\rangle = |81S_{1/2}, m_J = +1/2\rangle$), via an intermediate state ($|e\rangle = |5P_{1/2}, F = 1, m_F = +1\rangle$). One laser at 795 nm couples with the $|g\rangle \leftrightarrow |e\rangle$ transition, with a beam waist of $w_1 = 7.4 \mu\text{m}$ at the atom location and a typical Rabi frequency of $\Omega_1/(2\pi) = 2.3 \text{ MHz}$. A second laser at 474 nm couples with the $|e\rangle \leftrightarrow |r\rangle$ transition, with a beam waist of $w_2 = 8 \mu\text{m}$ and a typical Rabi frequency of $\Omega_2/(2\pi) = 10.7 \text{ MHz}$. The single-photon detuning is $\Delta_1/(2\pi) = -40 \text{ MHz}$ for the 795 nm laser. For the 474 nm excitation laser, the single-photon detuning Δ_2 is close to $-\Delta_1$, being variable. The two-photon excitation has an effective detuning of $\delta = \Delta_1 + \Delta_2$ and an effective single-atom Rabi frequency of $\Omega = \Omega_1\Omega_2/(2\Delta_1)$. The excitation lasers select a region with an effective transversal size of $\sqrt{(\pi/2)(1/w_1^2 + 1/w_2^2)^{-1}} = 6.8 \mu\text{m}$. The collective excitation in the Rydberg state is detected optically by applying a read laser (474 nm) with a Rabi frequency of Ω_3 . The read laser converts atomic excitations into single photons in a well-defined mode through phase matching. By using a beam splitter together with two single-photon detectors, we are able to measure the single-photon probability and perform a Hanbury-Brown-Twiss (HBT) test as well. More details on the experimental setup can be found in our previous publications [27,28].

By applying the two driving lasers, and adiabatically eliminating the intermediate state since $\Omega_1, \Omega_2 \ll \Delta$, we get the

eigenstates of the system as follows (if the blockade is strong enough):

$$\begin{cases} |+\rangle = \sin \theta |G\rangle + \cos \theta |R\rangle \\ |-\rangle = \cos \theta |G\rangle - \sin \theta |R\rangle \end{cases}, \quad (1)$$

where $|G\rangle$ refers to the case that all atoms are in the ground state $|g\rangle$, $|R\rangle = N^{-1/2} \sum_{i=1}^N |r_i\rangle$ refers to a singly excited state with one atom in the Rydberg state $|r\rangle$ and all other atoms remaining in the ground state $|g\rangle$, and $\theta = \arctan(\Omega_N/\delta)/2$ is the mixing angle with Ω_N being the collective Rabi frequency. By setting the two-photon detuning to $\delta = 0$, the system eigenstates are $|\pm\rangle = |G\rangle \pm |R\rangle$, thus the atomic ensemble undergoes collective Rabi oscillations between the state $|G\rangle$ and $|R\rangle$. A typically Rabi oscillation measured is shown in Fig. 1(d), from which we can estimate the effective number of atoms in the region of interest is around 180 via the relation of $N = \Omega_N^2/\Omega^2$ [18,21]. By setting the pulse duration for a pulse area of π , we get the maximal excitation probability. In the ideal case of full blockade with fixed N , δ , and pulse duration, such an excitation process is in principle deterministic.

III. EXPERIMENTAL RESULTS

In the case of chirped excitation [26], the detuning δ sweeps across $\delta = 0$, and the atoms adiabatically evolves from $|G\rangle$ to $|R\rangle$ (via $|-\rangle$ for positive chirping or $|+\rangle$ for negative chirping), and thus deterministic excitation of a single Rydberg atom can be also realized. Such an adiabatic process

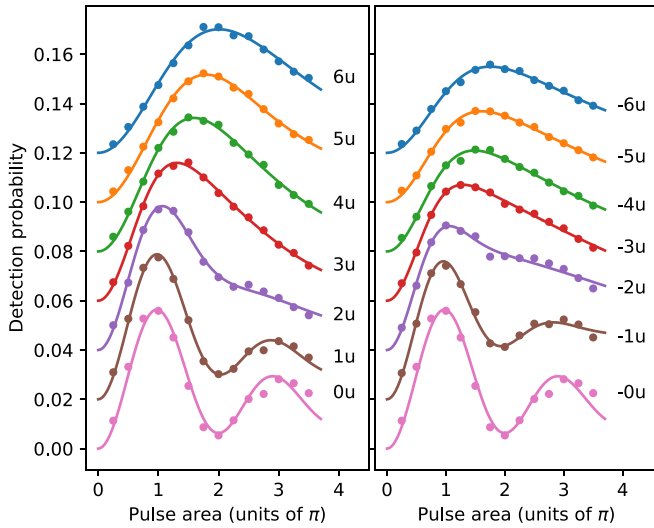


FIG. 2. Measurement of photon detection probability as a function of pulse area for a series of chirp rates. The result of positive chirping is shown in the left panel, and negative chirping is shown in the right panel. The chirp rate varies with a step of $u = 2\pi \times 12$ MHz/ μ s. For clarity, data for each chirp rate are shifted vertically with a step of 0.02 in sequence. Each curve is fitted with a function of $y = a \exp(-bx^2)[1 - \exp(-cx^2) \cos(dx)]$ where a , b , c , d are fitting parameters [21].

is robust, being insensitive to Ω_N , δ , and pulse duration. To ensure that the eigenstate evolution does not introduce orthogonal components, one has to fulfill the adiabatic condition, $|\dot{\Omega}_N \delta - \Omega_N \dot{\delta}| \ll 2(\Omega_N^2 + \delta^2)^{3/2}$ [22]. Thus, it requires a smooth pulse shape. In our experiment, the excitation process has a duration of $2T = 500$ ns. We shape the 474 nm pulse as Gaussian, and shape the 795 nm pulse as squared, which together ensure the smooth evolution of Ω over time. The full width at half maximum (FWHM) is 188 ns for 474 nm and 467 ns for 795 nm, respectively. Frequency chirping is realized via linearly sweeping the 474 nm laser around a central frequency of $\Delta_2 = 40$ MHz with an acousto-optic modulator configured in double pass.

First, we measure the photon detection probability as a function of the pulse area for a series of different chirp rates α by changing the laser power, with the result shown in Fig. 2. As the chirp rate increases, the result shows a variation from damped oscillations to damped plateaus, which is a clear signature of a transition from Rabi oscillations to adiabatic evolutions. The damping is mainly due to inhomogeneous light shifts induced by the contribution of minor double excitations [21] since the Rydberg interaction is not strong enough. It is also very interesting to compare the result of positive chirping with negative chirping. Clearly the positive chirping performs much better, which is due to influence of the intermediate state $|e\rangle$. During the chirping process, the detuning Δ_2 relative to the transition $|e\rangle \leftrightarrow |r\rangle$ evolves from $|\Delta_1| - \alpha T$ to $|\Delta_1| + \alpha T$. Thus $|\Delta_2|$ increases gradually as a function of time for positive chirping, while it decreases for negative chirping. Considering the gradual increase of the Rydberg population, the positive chirping scheme will thus suffer less from the off-resonant coupling and preserves

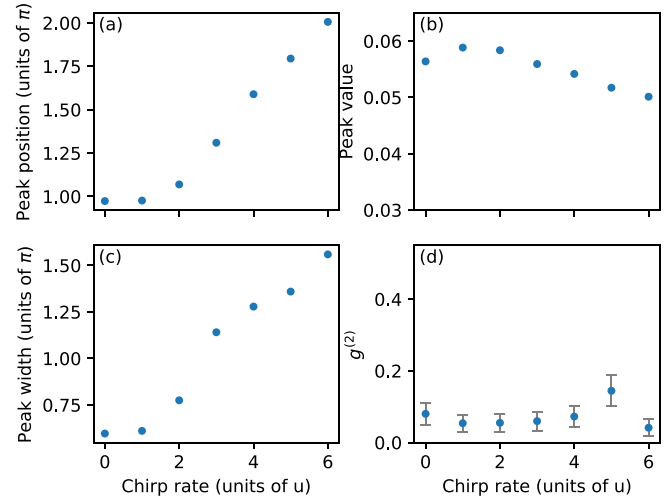


FIG. 3. For all the positive chirping data shown in Fig. 2, we summarize the peak positions in (a), the peak values in (b), and the peak widths (defined the region above 80% of the peak value) in (c). For each chirp rate, in (d) we also give a measured $g^{(2)}$ value which is selected for a point which is closest to the peak position in terms of pulse area.

the Rydberg excitation better. In the following, we focus on positive chirping only.

In order to quantitatively evaluate the performance of chirped excitation, we fit the results in Fig. 2 and get peak values and peak positions, which are summarized in Figs. 3(a) and 3(b). For the evaluation of robustness over the pulse area, we define the peak width as the range where the photon probability is above 80% of the peak value. We derive the peak width from the fitted function for each chirp rate, with results summarized in Fig. 3(c). It is clear to see that the peak width increases significantly as the increase of chirp rate. For a chirp rate of $\alpha = 6u$, the robustness is 2.6 times larger than the π -pulse excitation ($\alpha = 0$). We see that although a higher chirp rate gives rise to better robustness, it requires a larger pulse area than π . Simultaneously, with the increase of the chirp rate, the maximal photon probability achieved gets smaller, which is due to an increased contribution of double excitations. In addition, it is very important to verify that the chirping scheme creates genuine single photons. In our experiment, along with the measurement of the photon probability, we perform HBT test of the retrieved optical field simultaneously. We find that the chirping scheme does not influence the single-photon character in comparison with π -pulse excitation. For reference, we give the measured $g^{(2)}$ values for each chirping rate in Fig. 3(d). For each α , we choose a data point which is closest to the peaks in Fig. 2. We see that the variance is within the range of error bars.

The excitation scheme with chirped pulses is robust not only against the pulse area but also against other parameters, such as the laser frequency detunings. In our experiment, by setting the best pulse area, we deliberately shift Δ_1 and test the robustness of the photon probability against it. The result is shown in Fig. 4, in which we compare the chirping scheme with the π -pulse scheme. It is clear to see that the chirping

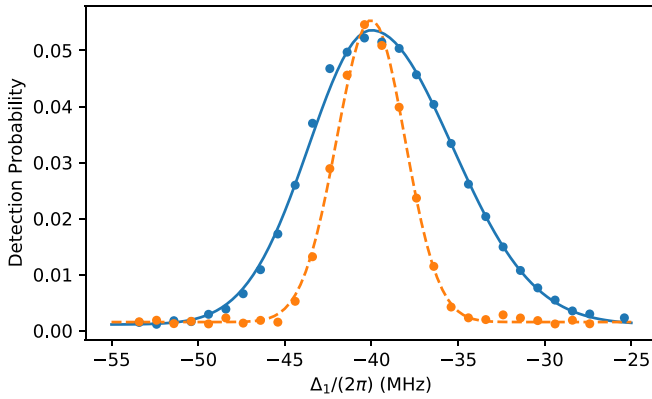


FIG. 4. Measurement of photon detection probability as a function of single-photon detuning Δ_1 . Result of the chirping scheme is shown as solid line in blue, and result of the π -pulse scheme is shown as dashed line in orange. Data points are fitted with asymmetric Gaussian functions. The two results are measured with the same laser powers. The pulse width (FWHM) determined by the 474 nm laser is 188 ns for the chirping scheme and 114 ns for the π -pulse scheme. For the chirping scheme, we set a chirp rate of $\alpha = 4\mu$. The pulse area is optimized to maximize the photon probability at $\Delta_1 = -40$ MHz for the two schemes.

scheme is much robust. Similarly to the case in Fig. 2, we also define a robustness range that the photon probability is higher than 80% of the peak value. We find that the range of robustness for the chirped excitation is 2.1 times larger than the π -pulse excitation. In the case of chirped excitation, we also find obvious asymmetry along the axis $\Delta_1 = -40$ MHz,

which we think is due to the increase of single-atom Rabi frequency $|\Omega|$ with the reduction of $|\Delta_1|$.

IV. CONCLUSION

To summarize, we realize the creation of single Rydberg excitations with chirped pulses via adiabatic rapid passage. The created excitations are verified to be phase coherent via directional retrieval as optical fields. The quantum signature of the excitations is tested via a photonic HBT test, which suggests that genuine single excitations are created and can be further used for quantum information applications. The detected single-photon probability is similar to traditional π -pulse excitation. We also test the robustness against variance of pulse area and two-photon detuning, and both results suggest that the chirping scheme is more robust than π -pulse excitation. Therefore, our work proves the validity of applying the technique of chirped excitation for an ensemble system under Rydberg blockade and may become an elementary tool in future experiments of quantum computing with collective encoding [27,29,30] and a quantum network with Rydberg superatoms [28,31–33].

ACKNOWLEDGMENTS

This work was supported by the National Key R&D Program of China (No. 2017YFA0303902), Anhui Initiative in Quantum Information Technologies, National Natural Science Foundation of China, and the Chinese Academy of Sciences. J.L. acknowledges support from China Postdoctoral Science Foundation (No. 2017M622000).

-
- [1] N. Sangouard, C. Simon, H. de Riedmatten, and N. Gisin, Quantum repeaters based on atomic ensembles and linear optics, *Rev. Mod. Phys.* **83**, 33 (2011).
- [2] S. D. Barrett, P. P. Rohde, and T. M. Stace, Scalable quantum computing with atomic ensembles, *New J. Phys.* **12**, 93032 (2010).
- [3] L.-M. Duan, M. D. Lukin, J. I. Cirac, and P. Zoller, Long-distance quantum communication with atomic ensembles and linear optics, *Nature (London)* **414**, 413 (2001).
- [4] H. J. Kimble, The quantum internet, *Nature (London)* **453**, 1023 (2008).
- [5] H.-J. Briegel, W. Dür, J. I. Cirac, and P. Zoller, Quantum Repeaters: The Role of Imperfect Local Operations in Quantum Communication, *Phys. Rev. Lett.* **81**, 5932 (1998).
- [6] S. Wehner, D. Elkouss, and R. Hanson, Quantum internet: A vision for the road ahead, *Science* **362**, eaam9288 (2018).
- [7] 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, *Nat. Phys.* **6**, 894 (2010).
- [8] 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, *Nat. Phys.* **8**, 517 (2012).
- [9] 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, *Phys. Rev. Lett.* **111**, 240503 (2013).
- [10] 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).
- [11] S.-J. Yang, X.-J. Wang, X.-H. Bao, and J.-W. Pan, An efficient quantum light-matter interface with sub-second lifetime, *Nat. Photonics* **10**, 381 (2016).
- [12] N. Somaschi, V. Giesz, L. De Santis, J. C. Loredo, M. P. Almeida, G. Hornecker, S. L. Portalupi, T. Grange, C. Antón, J. Demory *et al.*, Near-optimal single-photon sources in the solid state, *Nat. Photonics* **10**, 340 (2016).
- [13] H. Wang, H. Hu, T.-H. Chung, J. Qin, X. Yang, J.-P. Li, R.-Z. Liu, H.-S. Zhong, Y.-M. He, X. Ding *et al.*, On-Demand Semiconductor Source of Entangled Photons Which Simultaneously has High Fidelity, Efficiency, and Indistinguishability, *Phys. Rev. Lett.* **122**, 113602 (2019).
- [14] R. Keil, M. Zopf, Y. Chen, B. Höfer, J. Zhang, F. Ding, and O. G. Schmidt, Solid-state ensemble of highly entangled photon sources at rubidium atomic transitions, *Nat. Commun.* **8**, 15501 (2017).
- [15] 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, *Phys. Rev. Lett.* **87**, 037901 (2001).
- [16] M. Saffman and T. G. Walker, Creating single-atom and single-photon sources from entangled atomic ensembles, *Phys. Rev. A* **66**, 065403 (2002).
- [17] L. H. Pedersen and K. Mølmer, Few qubit atom-light interfaces with collective encoding, *Phys. Rev. A* **79**, 012320 (2009).
- [18] M. Saffman, T. G. Walker, and K. Mølmer, Quantum information with Rydberg atoms, *Rev. Mod. Phys.* **82**, 2313 (2010).
- [19] D. Comparat and P. Pillet, Dipole blockade in a cold Rydberg atomic sample, *J. Opt. Soc. Am. B* **27**, A208 (2010).
- [20] Y. Dudin and A. Kuzmich, Strongly interacting Rydberg excitations of a cold atomic gas, *Science* **336**, 887 (2012).
- [21] Y. Dudin, L. Li, F. Bariani, and A. Kuzmich, Observation of coherent many-body Rabi oscillations, *Nat. Phys.* **8**, 790 (2012).
- [22] N. V. Vitanov, T. Halfmann, B. W. Shore, and K. Bergmann, Laser-induced population transfer by adiabatic passage techniques, *Annu. Rev. Phys. Chem.* **52**, 763 (2001).
- [23] K. Bergmann, H. Theuer, and B. W. Shore, Coherent population transfer among quantum states of atoms and molecules, *Rev. Mod. Phys.* **70**, 1003 (1998).
- [24] N. V. Vitanov, A. A. Rangelov, B. W. Shore, and K. Bergmann, Stimulated Raman adiabatic passage in physics, chemistry, and beyond, *Rev. Mod. Phys.* **89**, 015006 (2017).
- [25] D. Petrosyan and K. Mølmer, Stimulated adiabatic passage in a dissipative ensemble of atoms with strong Rydberg-state interactions, *Phys. Rev. A* **87**, 033416 (2013).
- [26] I. I. Beterov, D. B. Tretyakov, V. M. Entin, E. A. Yakshina, I. I. Ryabtsev, C. McCormick, and S. Bergamini, Deterministic single-atom excitation via adiabatic passage and Rydberg blockade, *Phys. Rev. A* **84**, 023413 (2011).
- [27] 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, *Phys. Rev. Lett.* **117**, 180501 (2016).
- [28] 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, *Phys. Rev. Lett.* **123**, 140504 (2019).
- [29] E. Brion, K. Mølmer, and M. Saffman, Quantum Computing with Collective Ensembles of Multilevel Systems, *Phys. Rev. Lett.* **99**, 260501 (2007).
- [30] M. Saffman and K. Mølmer, Scaling the neutral-atom Rydberg gate quantum computer by collective encoding in holmium atoms, *Phys. Rev. A* **78**, 012336 (2008).
- [31] Y. Han, B. He, K. Heshami, C.-Z. Li, and C. Simon, Quantum repeaters based on Rydberg-blockade-coupled atomic ensembles, *Phys. Rev. A* **81**, 052311 (2010).
- [32] B. Zhao, M. Müller, K. Hammerer, and P. Zoller, Efficient quantum repeater based on deterministic Rydberg gates, *Phys. Rev. A* **81**, 052329 (2010).
- [33] L. Li, Y. O. Dudin, and A. Kuzmich, Entanglement between light and an optical atomic excitation, *Nature (London)* **498**, 466 (2013).