




Increasing the decoherence rate of Rydberg polaritons due to accumulating dark Rydberg atoms

Ko-Tang Chen,¹ Bongjune Kim ^{1,*} Chia-Chen Su,¹ Shih-Si Hsiao,¹ Shou-Jou Huang,²
Wen-Te Liao ^{2,3,4} and Ite A. Yu ^{1,4,†}

¹Department of Physics, National Tsing Hua University, Hsinchu 30013, Taiwan

²Department of Physics, National Central University, Taoyuan City 320317, Taiwan

³Physics Division, National Center for Theoretical Sciences, Taipei 10617, Taiwan

⁴Center for Quantum Technology, Hsinchu 30013, Taiwan



(Received 13 October 2021; accepted 16 March 2022; published 8 April 2022)

We experimentally observed an accumulative type of nonlinear attenuation and distortion of slow light, i.e., Rydberg polaritons, with the Rydberg state $|32D_{5/2}\rangle$ in the weak-interaction regime. The present effect of attenuation and distortion cannot be explained by considering only the dipole-dipole interaction (DDI) between Rydberg atoms in $|32D_{5/2}\rangle$. Our observation can be attributed to the atoms in the dark Rydberg states rather than those in the bright Rydberg state, i.e., $|32D_{5/2}\rangle$, driven by the coupling field. The dark Rydberg states are all the possible states in which the population decaying from $|32D_{5/2}\rangle$ accumulated over time, and they were not driven by the coupling field. Consequently, the DDI between the dark and bright Rydberg atoms increased the decoherence rate of the Rydberg polaritons. We performed three different experiments to verify the above hypothesis, to confirm the existence of the dark Rydberg states, and to measure the decay rate from the bright to the dark Rydberg states. In the theoretical model, we included the decay process from the bright to the dark Rydberg states and the DDI effect induced by both the bright and dark Rydberg atoms. All the experimental data of slow light taken at various probe Rabi frequencies were in good agreement with the theoretical predictions based on the model. This study points out an additional decoherence rate in the Rydberg-state electromagnetically induced transparency effect and provides a better understanding of the Rydberg-polariton system.

DOI: [10.1103/PhysRevResearch.4.023024](https://doi.org/10.1103/PhysRevResearch.4.023024)

I. INTRODUCTION

Atoms in the Rydberg states possess a strong electric dipole-dipole interaction (DDI) among themselves. The blockade effect arising from the DDI is a versatile mechanism for quantum information processing [1–4]. The combination of the DDI and the effect of electromagnetically induced transparency (EIT) can mediate strong photon-photon interactions [5–12]. Thus Rydberg atoms have led to applications such as quantum logic gates [5,13–15], single-photon generation [16–18], single-photon transistors [6,7], and single-photon switches [8]. Furthermore, the system of Rydberg atoms in a vapor or an array is also a platform for the study of many-body physics [19–26].

Most of the studies on Rydberg atoms have been performed in a strong-interaction regime. In contrast, the DDI-induced effect in the weak-interaction regime based on EIT was studied theoretically [27] and experimentally [28] with a low principal quantum number of $n = 32$ and a low Rydberg-

atom density equal to or less than $2.0 \times 10^9 \text{ cm}^{-3}$. The weak-interaction regime in those studies was $(r_b/r_a)^3 < 0.06$, where r_a and r_b are the half mean distance between the Rydberg atoms and the blockade radius, respectively. Due to the EIT effect [29], the propagation delay time or photon-matter interaction time in a high-optical-density (high-OD) medium was a couple of microseconds. With the help of a sufficient collision rate and interaction time in a high-OD EIT medium of weakly interacting Rydberg atoms, it was observed that there was a smaller width of the transverse momentum distribution of Rydberg polaritons at the exit point of the system as compared with that at the entrance [28].

The observation in Ref. [28] is a demonstration of the thermalization process of Rydberg polaritons and indicates the possibility of Rydberg polariton Bose-Einstein condensation (BEC). It was also suggested to make Rydberg polaritons stationary and cool down the temperature in both transverse and longitudinal directions for the realization of BEC. To achieve Rydberg polariton BEC, one needs to gather rather many particles of weakly interacting polaritons with a long interaction time in the system. However, the loss of the polaritons due to an extra attenuation, which is discussed in this paper, hinders the particles from achieving Rydberg polariton BEC.

In this paper, we systematically studied the transmission of a probe field propagating through a Rydberg-EIT system with experimental conditions similar to those in Ref. [28]. We observed distorted output pulses which were inconsistent

*upfe11@gmail.com

†yu@phys.nthu.edu.tw

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

with the theoretical prediction considering the DDI-induced decoherence rate [27,28]. Based on the measurements, we found evidence of the presence of dark Rydberg states which were not driven by the coupling field. The population can be transferred from a bright Rydberg state, which was excited by the coupling field, to the dark Rydberg states. Since the dark Rydberg atoms were not interacting with the coupling field, they accumulated over time. On the other hand, due to the DDI between the dark and bright Rydberg atoms, the DDI-induced decoherence rate increased with time.

Following the scenario described in the previous paragraph, we proposed a theoretical model based on the experimental data. Using a theoretical model to take into account the effect that the population accumulates in some dark Rydberg states and the dark Rydberg atoms cause an extra DDI-induced decoherence rate, we can successfully explain the experimental data. We believe the effect is universal. There are several previous papers that studied decay or transition from the bright to the dark Rydberg states and the phenomena caused by the dark Rydberg atoms. In this paper, however, the phenomenon of the increasing DDI-induced decoherence rate due to atoms accumulating in dark Rydberg states is systematically studied. This work can provide a better understanding for the creation of dark Rydberg atoms and their influence on Rydberg polaritons in the Rydberg-EIT system. This work also points out an obstacle in the realization of BEC with weakly interacting Rydberg polaritons.

This paper is organized as follows. In Sec. II, we present the distorted output probe pulse shapes, which we cannot explain with the theoretical model as it only considered the DDI between the bright Rydberg states under the EIT condition. In addition, we show the necessity of the consideration of the dark Rydberg states by measurements of the decay time constant, the steady-state attenuation coefficient of the square input pulse, and the lifetime of the dark Rydberg state. In Sec. III, we introduce a modified theoretical model to describe the increasing attenuation coefficient over time depending on the density of the Rydberg atom. We also report the method for estimating the decay rate from the bright Rydberg state to the dark Rydberg state and compare the experimental data with the theoretical prediction. In Sec. IV, we discuss the possible transfer process from the bright to the dark Rydberg states and the interaction strength between a bright Rydberg atom and a dark Rydberg atom. Finally, we summarize the results in Sec. V.

II. OBSERVATION OF THE ACCUMULATION EFFECT

A. Experiment setup

The experiment was performed with a cigar-shaped cold ^{87}Rb atom cloud produced by a magneto-optical trap. The dimension of the cloud was $1.5 \times 1.5 \times 5.0 \text{ mm}^3$ [30], and the atom temperature was about $350 \text{ } \mu\text{K}$ [28,31]. The transition scheme formed by the probe and coupling fields is shown in Fig. 1(a). The probe field drove the transition between the ground state $|1\rangle$ and the intermediate state $|3\rangle$, and the coupling field drove the transition between state $|3\rangle$ and the Rydberg state $|2\rangle$. States $|1\rangle$, $|2\rangle$, and $|3\rangle$ corresponded to the ground state $|5S_{1/2}, F=2, m_F=2\rangle$, the

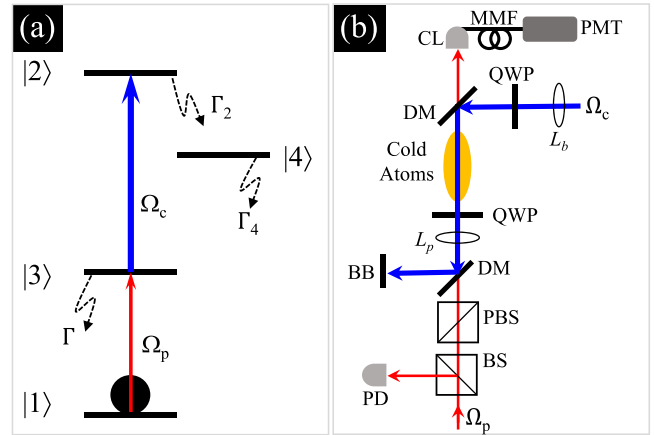


FIG. 1. (a) Relevant energy levels and transitions of the laser fields. $|1\rangle$ and $|3\rangle$ are the ground and intermediate excited states. $|2\rangle$ is the Rydberg state driven by the coupling field, and $|4\rangle$ represents all the nearby Rydberg states of $|2\rangle$ that involve the DDI effect. $|2\rangle$ and $|4\rangle$ are the bright and dark Rydberg states, respectively. Γ , Γ_2 , and Γ_4 represent the spontaneous decay rates of $|3\rangle$, $|2\rangle$, and $|4\rangle$. Ω_p and Ω_c denote the Rabi frequencies of the probe and coupling fields. The two fields' frequencies were stabilized to maintain the two-photon resonance. As compared with Γ , the magnitude of the one-photon detuning was negligible. (b) Sketch of the experiment setup. BB, beam blocker; BS, beam splitter; PBS, polarizing beam splitter; PD, photodetector; DM, dichroic mirror; L_p and L_b , lenses; QWP, quarter-wave plate; CL, collimation lens; MMF, multimode optical fiber; PMT, photomultiplier tube. Red and blue arrows indicate the optical paths of the probe and coupling beams, respectively.

Rydberg state $|32D_{5/2}, m_J = 5/2\rangle$, and the excited state $|5P_{3/2}, F=3, m_F=3\rangle$ of ^{87}Rb atoms, respectively. We prepared all of the population in a single Zeeman state $|5S_{1/2}, F=2, m_F=2\rangle$ by optical pumping [32]. In the experiment, the σ_+ -polarized probe and coupling fields were used. Owing to the optical pumping and polarization of the laser fields, the relevant energy levels $|1\rangle$, $|2\rangle$, and $|3\rangle$ were considered as a single Zeeman state. The spontaneous decay rate of $|3\rangle$ is $\Gamma = 2\pi \times 6.07 \text{ MHz}$, and that of $|2\rangle$ is $\Gamma_2 = 2\pi \times 7.9 \text{ kHz}$ or $1.3 \times 10^{-3}\Gamma$ [33,34]. As the van der Waals interaction energy is denoted as $\hbar C_6/r^6$, the Rydberg atoms in $|32D_{5/2}, m_J = 5/2\rangle$ have $C_6 = -2\pi \times 130 \text{ MHz } \mu\text{m}^6$ [35].

The experimental scheme is shown in Fig. 1(b). Frequency-stabilized laser systems generated the probe and coupling fields. The details of the stabilization method are described in Refs. [28,31]. The probe and coupling fields were the first-order beams of the acousto-optic modulators (AOMs, not drawn in the figure). The AOMs were used to control the time sequence, shape the probe pulse, and adjust the frequency and the amplitude of the fields precisely. Each field was sent to the atom cloud by the polarization-maintaining optical fiber (PMF) after the AOM. To minimize the Doppler effect, we used the counterpropagating scheme of the probe and coupling fields. At the center of the cloud, the e^{-1} full widths of the probe and coupling beams were 130 and $250 \text{ } \mu\text{m}$, respectively. The pulse shape and amplitude of the input probe field were monitored by a photodetector (PD) before entering the atom cloud as shown in Fig. 1(b). The probe field was

detected by a photomultiplier tube (PMT) after passing through the atom cloud. A digital oscilloscope (Agilent MSO6014A) acquired the signal from the PMT and produced the raw data.

B. Theoretical model without the accumulation effect

The Rydberg atoms inside the cloud were considered randomly distributed particles, similar to ideal gases, due to the assumption that the condition of a weakly interacting many-body system of Rydberg polaritons was satisfied. Therefore the mean-field model developed in Ref. [27] was adopted to describe the DDI-induced behaviors in this paper.

We initially considered only the states $|1\rangle$, $|2\rangle$, and $|3\rangle$ shown by Fig. 1(a) in the theoretical model. The DDI-induced decoherence rate γ_{DDI} and frequency shift δ_{DDI} , which are caused by the population in the Rydberg state $|2\rangle$, were taken into account. The optical Bloch equations (OBEs) of the density matrix operator and the Maxwell-Schrödinger equation (MSE) of the probe field are given below:

$$\frac{\partial}{\partial t} \rho_{21} = \frac{i}{2} \Omega_c \rho_{31} + i(\delta_{\text{DDI}} + \delta) \rho_{21} - \left(\gamma_{\text{DDI}} + \gamma_0 + \frac{\Gamma_2}{2} \right) \rho_{21}, \quad (1)$$

$$\frac{\partial}{\partial t} \rho_{31} = \frac{i}{2} \Omega_p (\rho_{11} - \rho_{33}) + \frac{i}{2} \Omega_c \rho_{21} + i \Delta_p \rho_{31} - \frac{\Gamma}{2} \rho_{31}, \quad (2)$$

$$\frac{\partial}{\partial t} \rho_{32} = \frac{i}{2} \Omega_p \rho_{21}^* + \frac{i}{2} \Omega_c (\rho_{22} - \rho_{33}) - i \Delta_c \rho_{32} - \frac{\Gamma}{2} \rho_{32}, \quad (3)$$

$$\frac{\partial}{\partial t} \rho_{22} = \frac{i}{2} \Omega_c \rho_{32} - \frac{i}{2} \Omega_c \rho_{32}^* - \Gamma_2 \rho_{22},$$

$$\frac{\partial}{\partial t} \rho_{33} = -\frac{i}{2} \Omega_p^* \rho_{31} + \frac{i}{2} \Omega_p \rho_{31}^* - \frac{i}{2} \Omega_c \rho_{32} \quad (4)$$

$$+ \frac{i}{2} \Omega_c \rho_{32}^* - \Gamma \rho_{33}, \quad (5)$$

$$\frac{\partial}{\partial t} \rho_{11} = \frac{i}{2} \Omega_p^* \rho_{31} - \frac{i}{2} \Omega_p \rho_{31}^* + \Gamma \rho_{33}, \quad (6)$$

$$\frac{1}{c} \frac{\partial}{\partial t} \Omega_p + \frac{\partial}{\partial z} \Omega_p = i \frac{\alpha \Gamma}{2L} \rho_{31}, \quad (7)$$

where ρ_{ij} ($i, j = 1, 2, 3$) represents a matrix element of the density matrix operator, Ω_p and Ω_c are the Rabi frequencies of the probe and coupling fields, δ is the two-photon detuning, γ_0 is the intrinsic decoherence rate in the experimental system, Δ_p and Δ_c are the one-photon detunings of the probe and coupling transitions, and α and L are the optical depth and length of the medium.

The values of γ_{DDI} and δ_{DDI} were obtained by the analytic formulas of the attenuation coefficient $\Delta\beta$ and the phase shift $\Delta\phi$, respectively, induced by the DDI in the steady-state condition. The formulas were derived in Ref. [27], utilizing a mean-field model based on the nearest-neighbor distribution, and they were experimentally verified in Ref. [28]. At the

condition of $\delta = 0$ and $\Delta_c \ll \Gamma$, the formulas of $\Delta\beta$ and $\Delta\phi$ are given by

$$\Delta\beta \left(\equiv \frac{2\alpha\Gamma\gamma_{\text{DDI}}}{\Omega_c^2} \right) = \frac{2\pi^2\alpha\sqrt{|C_6|\Gamma}}{3\Omega_c} n_R, \quad (8)$$

$$\Delta\phi \left(\equiv \frac{\alpha\Gamma\delta_{\text{DDI}}}{\Omega_c^2} \right) = \frac{\pi^2\alpha\sqrt{|C_6|\Gamma}}{3\Omega_c} n_R, \quad (9)$$

where n_R is the density of the Rydberg-state atoms. Using the relation $n_R = n_a \rho_{22}$ between n_R and the density of all atoms n_a , we introduced the coefficient A , which is written as

$$A = \frac{\pi^2\Omega_c\sqrt{|C_6|\Gamma}}{3\sqrt{\Gamma}} n_a. \quad (10)$$

The coefficient A represents the DDI-induced decoherence rate or frequency shift per unit ρ_{22} . From Eqs. (8) and (10), we obtained

$$\gamma_{\text{DDI}} = A\rho_{22}. \quad (11)$$

Similarly, from Eqs. (9) and (10), we also obtained

$$\delta_{\text{DDI}} = A\rho_{22}. \quad (12)$$

Thus γ_{DDI} and δ_{DDI} were replaced by $A\rho_{22}$ in the OBEs. According to the values of Ω_c , C_6 , and n_a used in the experiment, it was determined that $A = 0.76\Gamma$. We set and fixed A to 0.76Γ in all the theoretical predictions of this work, and the values of γ_{DDI} and δ_{DDI} are linearly proportional to the population of the Rydberg state, $\rho_{22}(z, t)$.

Furthermore, the effect of the DDI, i.e., γ_{DDI} and δ_{DDI} , was ignored to obtain Eq. (3) due to the assumption of $A\rho_{22}$, $\Gamma_2 \ll \Gamma$, which was the case in the experiment. Throughout this study, the two-photon resonance condition of δ ($\equiv \Delta_p + \Delta_c$) = 0 was always maintained. Please note that the term $i(\delta_{\text{DDI}} + \delta)\rho_{21}$ in Eq. (1), i.e., $i(A\rho_{22} + \delta)\rho_{21}$, played a negligible role in the output probe transmission, since $\delta = 0$ and $4(A\rho_{22})^2\Gamma^2 \ll \Omega_c^4$ in all the cases of this work. To obtain Eqs. (5) and (6), we also considered that the population in $|2\rangle$ rarely decayed to $|1\rangle$ and $|3\rangle$. When we made all of the population in $|2\rangle$ decay to $|1\rangle$ by adding the term $+\Gamma_2\rho_{22}$ to the right-hand side of Eq. (6), the calculation result changed very little because $\Omega_p^2 \ll \Omega_c^2$ in this paper.

C. Distorted output pulses of slow light

Before the measurement, the values of the one-photon detuning and two-photon detuning were set to zero and experimentally verified. We determined the experimental parameters in the order of $\Omega_c \rightarrow \gamma_0 \rightarrow \text{OD}$. The parameters were confirmed again after the measurement in the reverse order of that before measurement. The determined values of Ω_c , γ_0 , and OD were 1.0Γ , $9.0 \times 10^{-3}\Gamma$, and 70, respectively. Details can be found in Appendix A and Ref. [31].

Following the definition of the half mean distance between Rydberg polaritons, $r_a = (4\pi n_R/3)^{-1/3}$, the estimated smallest r_a in this experiment was $5.0 \mu\text{m}$ when Ω_{p0} was 0.2Γ [27]. The blockade radius r_B was $1.9 \mu\text{m}$ according to the formula of $r_B = (2|C_6|\Gamma/\Omega_c^2)^{1/6}$ [11]. $\Omega_{p0} = 0.2\Gamma$ was the largest input probe Rabi frequency, and $\Omega_c = 1.0\Gamma$ was kept throughout the EIT experiment. Thus it was considered a weakly interacting many-body system of Rydberg polaritons, which satisfied the condition of $(r_B/r_a)^3 \ll 1$.

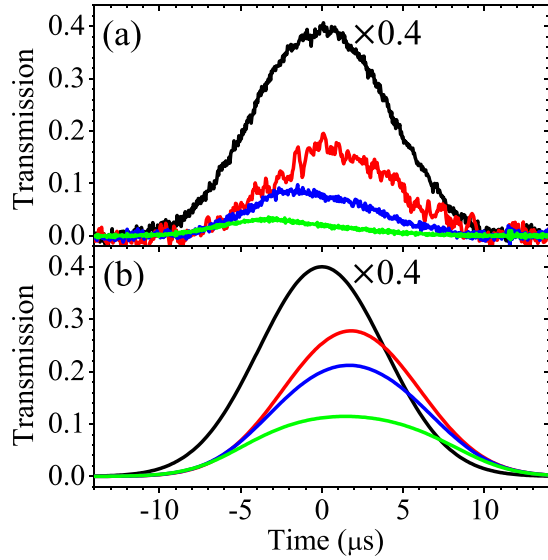


FIG. 2. Experimental observation that the slow light of a Gaussian pulse was distorted and a larger value of Ω_{p0} made the distortion more severe. (a) Experimental data of the output pulses vs time are shown by red, blue, and green curves, and their input Rabi frequencies Ω_{p0} of 0.05Γ , 0.1Γ , and 0.2Γ , respectively. Since the shape of the three input pulses is very similar, we only plot the one with the Ω_{p0} of 0.1Γ as shown by the black curve, which is scaled down by a factor of 0.4. In the measurements, α (optical depth) = 70 , $\Omega_c = 1.0\Gamma$, and $\gamma_0 = 9.0 \times 10^{-3}\Gamma$, which were determined experimentally [31]. (b) Theoretical predictions by solving Eqs. (1)–(7) with the above values of Ω_{p0} , α , Ω_c , and γ_0 , and with the coefficient $A = 0.76\Gamma$ given by Eq. (10).

Using the above experimental conditions, we measured the slow light data as shown in Fig. 2(a). An input probe pulse with a Gaussian e^{-1} full width of $11.5 \mu\text{s}$ was used in this measurement. This input pulse was far longer than that used in determining the OD. Considering the values of OD and Ω_c , the delay time τ_d was expected to be $\sim 1.8 \mu\text{s}$ as the result of a short input Gaussian pulse with the e^{-1} full width of $0.66 \mu\text{s}$. However, the peak position of the output pulse with $\Omega_{p0} = 0.05\Gamma$ of the long Gaussian input probe pulse was nearly the same as the peak position of the input pulse (i.e., the delay time was nearly zero). Furthermore, a stronger input pulse had an output peak position that preceded the input peak position and showed a more distorted shape of the output pulse.

The degree of distortion of the output pulse shape depends on Ω_{p0} , or equivalently the Rydberg atom density. One might guess that the DDI effect could explain the distortion, and the experimental data of Fig. 2(a) could be predicted by Eqs. (1)–(7) with the introduction of γ_{DDI} and δ_{DDI} . To test this hypothesis, we calculated the theoretical predictions as shown in Fig. 2(b) by solving Eqs. (1)–(7), which included the effect of DDI. We used the experimentally determined values of Ω_{p0} , α , Ω_c , and γ_0 in the calculation. Owing to the DDI effect, i.e., the term γ_{DDI} or $A\rho_{22}$ in Eq. (1), we did observe a lower transmission with a higher value of Ω_{p0} in the theoretical prediction. In each case of Ω_{p0} , the theoretical prediction gave a very similar delay time ($\sim 1.8 \mu\text{s}$), which resulted in a higher

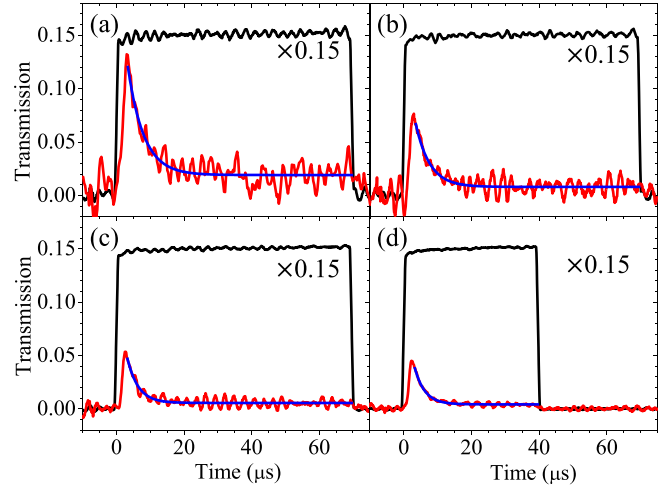


FIG. 3. Experimental observation of the accumulation effect, in which the DDI-induced attenuation increased with time. In each panel, the black curve scaled down by a factor of 0.15 is the input pulse, the red curve represents the output pulse, and the blue curve is the best fit of the tail part of the red curve. The best fit is an exponential-decay function. The values of Ω_{p0} were (a) 0.08Γ , (b) 0.1Γ , (c) 0.15Γ , and (d) 0.2Γ , respectively. In the measurements, α (optical depth) = 70 , $\Omega_c = 1.0\Gamma$, and $\gamma_0 = 1.1 \times 10^{-2}\Gamma$, which were determined experimentally [31].

peak transmission than the experimental value. The theoretical predictions did not agree with the experimental data.

D. Phenomenon of the DDI-induced attenuation increasing with time

To further study the phenomenon, we applied the long square probe pulse with the input Rabi frequency Ω_{p0} of 0.08Γ , 0.10Γ , 0.15Γ , or 0.2Γ under the constant presence of the coupling field, as shown in Fig. 3. The experimental parameters α (OD) = 70 , $\Omega_c = 1.0\Gamma$, $\Delta = 0$, and $\delta = 0$ were kept the same as in the measurements with the Gaussian input, but γ_0 was changed to $1.1 \times 10^{-2}\Gamma$ in the measurement.

Regarding the behavior of the output probe field in Fig. 3, we observed that the transmission decreased (or the attenuation increases) with time, after it reached the peak value in each of the measurements. The peak value of the output probe transmission decreased with Ω_{p0} . This was expected from the DDI effect, i.e., the term γ_{DDI} or $A\rho_{22}$ in Eq. (1), where $\rho_{22} \approx \Omega_p^2/\Omega_c^2$. However, neither the theoretical model nor Eqs. (1)–(7) described in Sec. II B were able to explain the phenomenon of the transmission decay or the increasing attenuation over time. We fitted the decayed part of the experimental data with an exponential-decay function and determined the decay time constant and the steady-state attenuation coefficient. The blue curves shown in Fig. 3 are the best fits.

Based on the best fits in Fig. 3, the decay time constant τ_{sq} and steady-state attenuation coefficient β_{ss} as functions of Ω_{p0}^2 are shown in Figs. 4(a) and 4(b), respectively. The value of β_{ss} was given by the absolute value of the logarithm of the transmission at the steady state. Figures 4(a) and 4(b) clearly show that a larger value of Ω_{p0}^2 resulted in a faster decay and a larger steady-state attenuation.

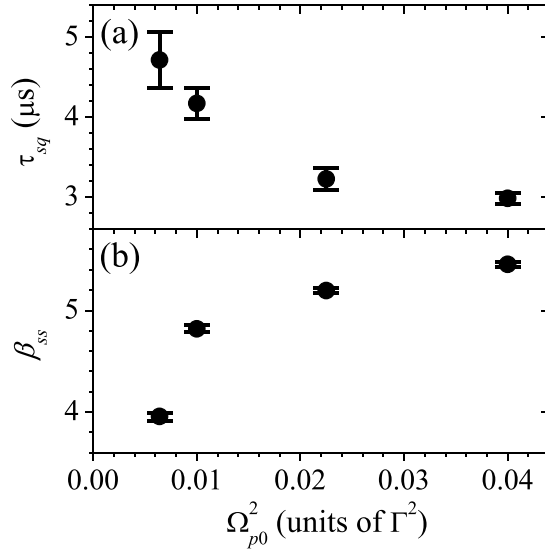


FIG. 4. Experimental observation of a larger Ω_{p0} resulting in a faster accumulation of DDI-induced attenuation. (a) Decay time constant τ_{sq} as a function of Ω_{p0}^2 . The values of τ_{sq} were determined by the best fits shown in Fig. 3. (b) The steady-state attenuation coefficient β_{ss} as a function of Ω_{p0}^2 . $\beta_{ss} = -\ln(T_{ss})$, where T_{ss} is the steady-state transmission. The values of T_{ss} were determined by the vertical offsets of the blue curves shown in Fig. 3.

A higher Rydberg polariton density leads to a larger DDI-induced attenuation. Thus the data in Figs. 3 and 4 inferred that the Rydberg polariton density increased with time. One can guess that some of the Rydberg population in $|2\rangle$ was transferred to other states (denoted as $|4\rangle$) and that the atoms in $|4\rangle$ did not interact with the coupling field but were able to have a DDI with the atoms in $|2\rangle$. Due to the existence of the DDI, $|4\rangle$ had to represent some nearby Rydberg states of $|2\rangle$. The population in $|4\rangle$, which was not driven by the coupling field, should have accumulated. As more population accumulated in $|4\rangle$, the DDI-induced attenuation of $|2\rangle$ became large. Thus the probe output transmission decreased with time, exhibiting the accumulation phenomenon of the DDI effect.

In Fig. 1(a), we introduce $|4\rangle$ to represent all possible nearby Rydberg states of $|2\rangle$. The spontaneous or collisional decay process could cause the transfer of the population from $|2\rangle$ to $|4\rangle$, because no additional field was applied in the experiment. Since $|2\rangle$ was driven by the coupling field, it was called the bright Rydberg state. On the other hand, $|4\rangle$ did not interact with any applied field and was called the dark Rydberg state.

E. Evidence of dark Rydberg states

To verify the hypothesis based on the dark Rydberg state $|4\rangle$, we measured the existing time of the DDI effect, after nearly all the atoms in the bright Rydberg state $|2\rangle$ had been deexcited. The basic idea of this measurement was as follows. Since the atoms in $|4\rangle$ did not interact with any applied field, they should decay by themselves and have a decay rate of Γ_4 . Furthermore, $|4\rangle$ represented a number of Rydberg states, so Γ_4^{-1} must be close to the Rydberg-state lifetime. Conse-

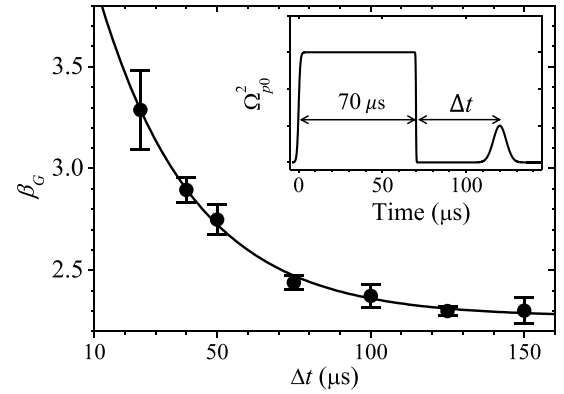


FIG. 5. Experimental observation of the lifetime of the dark Rydberg states by the measurement of DDI-induced attenuation β_G as a function of Δt . Inset: the time sequence of the measurement. The coupling field was continuously on during the measurement. Circles are the experimental data, and the black curve is the best fit of an exponential-decay function. The decay time constant of the best fit was $31 \mu\text{s}$, which is similar to the lifetimes of dark Rydberg states with a principal quantum number n of around 32.

quently, after the atoms in $|2\rangle$ had been removed, the DDI effect should still exist in the system and gradually decay with the time constant of Γ_4^{-1} . Then, we explored the DDI-induced attenuation as a function of time in the system by employing a very weak Gaussian probe pulse and the coupling field. The probe pulse was weak enough to cause a negligible DDI effect by itself.

The sequence of the input probe field is depicted in the inset of Fig. 5. A $70\text{-}\mu\text{s}$ square probe pulse of $\Omega_{p0} = 0.1\Gamma$ was first employed. The pulse duration was sufficiently long compared with the lifetime of $|2\rangle$, which was approximately $20 \mu\text{s}$ at room temperature. The OD was 70, and the coupling field of $\Omega_c = 1.0\Gamma$ was continuously kept on during the measurement. After the square probe pulse was switched off, the coupling field quickly deexcited the remaining population in $|2\rangle$. The deexcitation time was estimated to be about 27 ns. We waited for a certain time Δt after the square pulse was turned off and then applied a weak Gaussian probe pulse to measure the DDI-induced attenuation β_G as a function of Δt . The value of β_G was equal to the absolute value of the logarithm of the peak transmission of the output Gaussian pulse. The input Gaussian pulse had a peak Ω_{p0} of 0.05Γ and an e^{-1} full width of $10 \mu\text{s}$. Compared with the $70\text{-}\mu\text{s}$ square probe pulse, the weaker and much shorter Gaussian probe pulse induced a negligible DDI effect.

We observed the exponential-decay behavior of β_G versus Δt as shown in Fig. 5, in which the circles are the experimental data and the black curve is the best fit of an exponential-decay function. The decay time constant of the best fit was $31 \mu\text{s}$, which is about the lifetime of a Rydberg state with a principal quantum number between 32 and 38. The atoms in $|2\rangle$ were quickly deexcited by the coupling field, after the square probe pulse was switched off. If the atoms in the dark Rydberg state $|4\rangle$ could not exist in the system, the value of β_G would quickly drop to its steady-state value due to the absence of the atoms in the bright Rydberg state $|2\rangle$. The slow decay of β_G demonstrated the existence of the atoms in

$|4\rangle$, which did not interact with the coupling field, but gave the DDI effect and decayed slowly.

III. THEORY OF THE ACCUMULATION EFFECT AND EXPERIMENTAL VERIFICATION

A. Theoretical model with the accumulation effect

In the previous section, we discussed the necessity of the consideration of dark Rydberg states in the system. To simulate the accumulative DDI effect observed in the experiment, we improved the theoretical model described in Sec. II B by including the dark Rydberg state $|4\rangle$ and the decay process from $|2\rangle$ to $|4\rangle$. The DDI effect between one atom in $|2\rangle$ and one atom in $|4\rangle$ was also taken into account. Thus the OBEs can be given by

$$\frac{\partial}{\partial t}\rho_{21} = \frac{i}{2}\Omega_c\rho_{31} + i(A\rho_{22} + B'\rho_{44} + \delta)\rho_{21} - \left(A\rho_{22} + B\rho_{44} + \gamma_0 + \frac{\Gamma_{24}}{2}\right)\rho_{21}, \quad (13)$$

$$\frac{\partial}{\partial t}\rho_{31} = \frac{i}{2}\Omega_p(\rho_{11} - \rho_{33}) + \frac{i}{2}\Omega_c\rho_{21} + i\Delta_p\rho_{31} - \frac{\Gamma}{2}\rho_{31}, \quad (14)$$

$$\frac{\partial}{\partial t}\rho_{32} = \frac{i}{2}\Omega_p\rho_{21}^* + \frac{i}{2}\Omega_c(\rho_{22} - \rho_{33}) - i\Delta_c\rho_{32} - \frac{\Gamma}{2}\rho_{32}, \quad (15)$$

$$\frac{\partial}{\partial t}\rho_{22} = \frac{i}{2}\Omega_c\rho_{32} - \frac{i}{2}\Omega_c\rho_{32}^* - \Gamma_{24}\rho_{22}, \quad (16)$$

$$\frac{\partial}{\partial t}\rho_{33} = -\frac{i}{2}\Omega_p^*\rho_{31} + \frac{i}{2}\Omega_p\rho_{31}^* - \frac{i}{2}\Omega_c\rho_{32} + \frac{i}{2}\Omega_c\rho_{32}^* - \Gamma\rho_{33}, \quad (17)$$

$$\frac{\partial}{\partial t}\rho_{44} = \Gamma_{24}\rho_{22} - \Gamma_4\rho_{44}, \quad (18)$$

$$\frac{\partial}{\partial t}\rho_{11} = \frac{i}{2}\Omega_p^*\rho_{31} - \frac{i}{2}\Omega_p\rho_{31}^* + \Gamma\rho_{33}, \quad (19)$$

where the coefficient A represents the DDI-induced decoherence rate or frequency shift per unit ρ_{22} with a value given by Eq. (10), the coefficient B (or B') is similar to the coefficient A and represents the DDI-induced decoherence rate (or frequency shift) per unit ρ_{44} , Γ_{24} is the decay rate from $|2\rangle$ to $|4\rangle$, and Γ_4 is the decay rate of $|4\rangle$.

The terms $A\rho_{22} + B'\rho_{44}$ and $A\rho_{22} + B\rho_{44}$ in Eq. (13) represent δ_{DDI} and γ_{DDI} , respectively. The values of γ_{DDI} and δ_{DDI} varied with space and time due to the populations of $\rho_{22}(z, t)$ and $\rho_{44}(z, t)$ during the propagation of the probe light. The decay rate Γ_{24} was parametrized as

$$\Gamma_{24} \equiv C\rho_{22} + D, \quad (20)$$

where the coefficient C is the two-body decay rate per unit ρ_{22} and the coefficient D is the one-body decay rate. The atomic density n_a of the system is a part of the coefficient C . Further discussion regarding the transfer mechanism can be found in Sec. IV.

In Eq. (15), the DDI effect, i.e., γ_{DDI} and δ_{DDI} , and the decay rates, i.e., Γ_{24} , and Γ_2 , were ignored due to the experimental condition of $A\rho_{22}, B\rho_{44}, \Gamma_{24}, \Gamma_2 \ll \Gamma$. Please note that the frequency shift induced by the population in each of the dark Rydberg states might be positive or negative, and thus the net frequency shift resulted in $|B'| \leq B$. Furthermore, the term $i(A\rho_{22} + B'\rho_{44} + \delta)\rho_{21}$ in Eq. (13) played a negligible role in the output probe transmission, since $\delta = 0$ and $4|A\rho_{22} + B\rho_{44}|^2\Gamma^2 \ll \Omega_c^4$ in all the cases of this work.

The value of A given by Eq. (10) was derived from Ref. [27] and experimentally verified in Ref. [28]. We first measured the coefficients C and D as described in Sec. III B. Using the known values of A , C , and D , we then determined B using the experimental data of Fig. 3 as illustrated in Sec. III C. Finally, we compared the experimental data of the slow light shown in Fig. 2 with the theoretical predictions calculated using the experimentally determined values of A , B , C , and D in Sec. III C. The comparison was used to test the validity of the theoretical model introduced here.

B. Determination of the decay rate from bright to dark Rydberg states

We designed an experiment to determine the coefficients C and D in Eq. (20). The decay rate, Γ_{24} , of the atoms in $|2\rangle$ was measured against the atomic density of $|2\rangle$, n_R . The experiment details can be found in Appendix B. The procedure of the measurement of Γ_{24} was as follows. We first prepared a given number of atoms in the ground state $|1\rangle$ and excited them to $|2\rangle$ with a two-photon transition (TPT) pulse. The TPT had a large one-photon detuning Δ . After the TPT pulse, no light field was applied, and the atoms in $|2\rangle$ decayed. Then, we waited for a certain time Δt and also depleted the residual atoms in $|1\rangle$. At the end of Δt , the remaining atoms in $|2\rangle$ were brought back to $|1\rangle$ with another TPT pulse. Finally, after the second TPT pulse we determined the number of returned atoms in $|1\rangle$ or, equivalently, measured the absorption coefficient $\Delta\beta$ of the weak probe field. The result of $\Delta\beta$ was proportional to the remaining atoms in $|2\rangle$ after the given Δt . By varying Δt and measuring $\Delta\beta$, we obtained the data points of $\Delta\beta$ as a function of Δt and fitted the data with an exponential function. The best fit gave the value of Γ_{24} .

The circles in Fig. 6 are the experimental data of Γ_{24} as a function of n_R . Each circle represents the result of a series of measurements similar to those shown in Appendix C. We fitted the circles in Fig. 6 with the fitting function of a straight line. The interception of the best fit determined the coefficient D defined in Eq. (20), which was $2.0 \times 10^{-3}\Gamma$. The determined value of D was comparable to the spontaneous decay rate of $32D_{5/2}$ at room temperature, which is $1.3 \times 10^{-3}\Gamma$ [33,34]. Furthermore, the slope, k , of the best fit determined the coefficient C defined in Eq. (20) in the following way: Since $kn_R = C\rho_{22}$ and $n_R = \rho_{22}n_a = \rho_{22}\alpha/(\sigma_0L)$, the coefficient C is given by

$$C = k\alpha/(\sigma_0L), \quad (21)$$

where α is the OD used in the EIT experiment and σ_0 is the absorption cross section of the resonant probe transition from $|5S_{1/2}, F=2, m_F=2\rangle$ to $|5P_{3/2}, F=3, m_F=3\rangle$. During the measurements of the data shown in Figs. 2(a) and

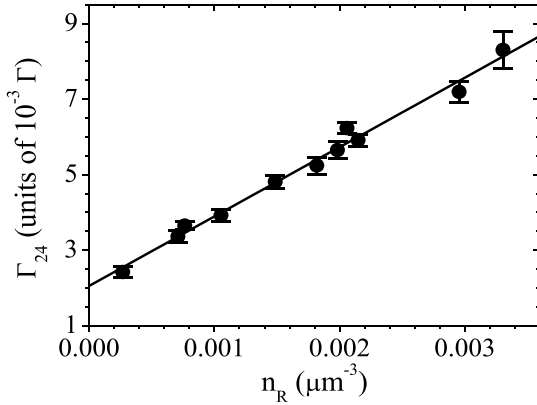


FIG. 6. The decay rate Γ_{24} as a function of the density, n_R , of the atoms in $|2\rangle$. Circles are the experimental data. Each data point was determined by the result of a series of measurements similar to those in Appendix C. The black line is the best fit of a straight line.

3(a)–3(d), the optical depth α was 70, and we used the above equation to estimate that the value of C was $9.1 \times 10^{-2} \Gamma$ at $\alpha = 70$. Possible processes of the population transfer from the bright to the dark Rydberg states are discussed in Sec. IV.

C. Comparison between theoretical predictions and experimental results

After determining the values of C and D in the decay rate Γ_{24} used in Eqs. (13), (16), and (18), we next utilized the experimental data of Fig. 3 to determine the value of coefficient B used in Eq. (13). Since the measured value of Γ_{24} was the total decay rate of $|2\rangle$ but not merely the decay rate from $|2\rangle$ to $|4\rangle$, the determined value of B might account for the discrepancy. Figure 7 shows the comparison between the experimental data of the long square probe pulses shown in Figs. 3(a)–3(d) and the theoretical predictions calculated with the modified model described in Sec. III A. In the calculation, we used the experimentally determined values of the optical depth, the coupling Rabi frequency, and the intrinsic decoherence rate γ_0 in the system [31]. We set A (the DDI-induced decoherence rate per unit ρ_{22}) to the value given by Eq. (10) and used the values of C and D derived from the result of Fig. 6. The only adjustable parameter in the calculation of the predictions was B (the DDI-induced decoherence rate per unit ρ_{44}). Note that we set $B' = B$ in the term $i(A\rho_{22} + B'\rho_{44} + \delta)\rho_{21}$ in Eq. (13), but this term played a negligible role in the calculation result of the output probe transmission. A good agreement between the theoretical predictions and experimental data was obtained at the coefficient $B = 7.7\Gamma$.

A single optimum value of B was able to explain all the experimental data taken at different input probe Rabi frequencies. This demonstrated that the theoretical model presented by Eqs. (13)–(20) was qualitatively valid. Furthermore, we compared the experimental data of the Gaussian input probe pulses as shown in Fig. 2 with the theoretical predictions. The values of the parameters used in the calculation of the predictions were the same as those in Fig. 7 except for the value of γ_0 , which had a day-to-day fluctuation of $1.5 \times 10^{-3} \Gamma$.

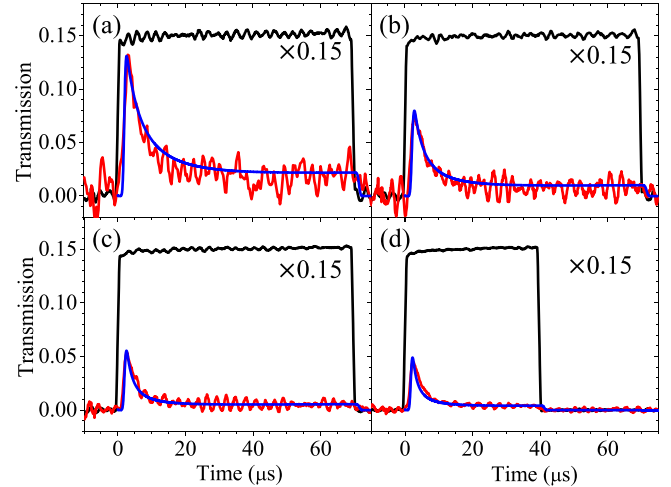


FIG. 7. Comparison between the experimental data of the long square probe pulses and the theoretical predictions. Black curves represent the input probe pulses and are scaled down by a factor of 0.15. Red curves represent the output probe pulses under the constant presence of the coupling field. The experimental data are the same as those shown in Fig. 3, where the input Rabi frequencies Ω_{p0} are (a) 0.08Γ , (b) 0.1Γ , (c) 0.15Γ , and (d) 0.2Γ . Blue curves are the predictions calculated using Eqs. (7) and (13)–(20). In the calculation, we set α (optical depth) = 70, $\Omega_c = 1.0\Gamma$, and $\gamma_0 = 1.1 \times 10^{-2} \Gamma$, which were determined experimentally, the coefficients $C = 9.1 \times 10^{-2} \Gamma$ and $D = 1.3 \times 10^{-3} \Gamma$, which were deduced from the result of Fig. 6, and $A = 0.76\Gamma$ given by Eq. (10). B was adjusted to make the predictions fit the data, and its optimum value is 7.7Γ .

Figures 8(a)–8(c) show that the experimental data were all in good agreement with the theoretical predictions, manifesting that the theoretical model was also quantitatively successful.

IV. DISCUSSION

We discuss some possible mechanisms of the population transfer to the dark Rydberg state in this section. Experimental observations led by the existence of the dark Rydberg state which was transferred from a bright Rydberg state have been reported in several papers [36–42]. The underlying mechanisms of such transfers can be classified into four categories as follows: (i) transitions driven by a microwave field [36], (ii) the spontaneous decay [37–39], (iii) the DDI-induced antiblockade excitation and state-change collision assisted by radiation trapping [40,43], and (iv) the super-radiance of the transition with a long wavelength induced by the blackbody radiation (BBR) [41,42]. Please see Appendix D for further discussion of these papers.

However, mechanisms (i), (ii), and (iii) cannot explain our observations. The corresponding reasons are the following. (a) No additional microwave field was applied for population transfer to another Rydberg state. (b) The spontaneous decay was a one-body process, and its rate should not depend on the Rydberg-atom density. However, the observed decay rate of the population in the bright Rydberg state depended on the Rydberg-atom density, and it is much higher than the spontaneous decay at room temperature. (c) According to the experimental condition, we made an estimation, illustrated in

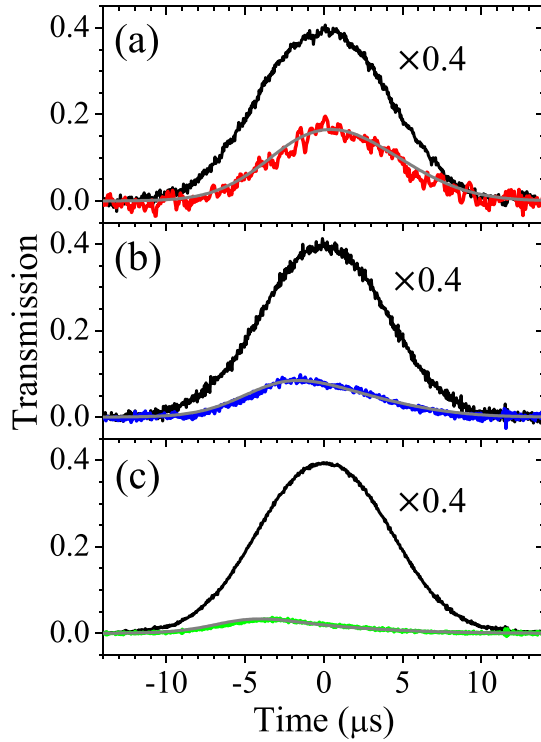


FIG. 8. Comparison between the experimental data of the Gaussian input probe pulses and the theoretical predictions. Black curves represent the input probe pulses and are scaled down by a factor of 0.4. Red, blue, and green curves represent the output probe pulses under the constant presence of the coupling field. The experimental data are the same as those shown in Fig. 2, where the input Rabi frequencies Ω_{p0} are (a) 0.05Γ , (b) 0.1Γ , and (c) 0.2Γ . Gray curves are the theoretical predictions calculated with Eqs. (7) and (13)–(20). All the parameters used in the calculation here are the same as those used in Fig. 7, except for $\gamma_0 = 9.0 \times 10^{-3}\Gamma$.

the next paragraph, to show that the state-changing Rydberg collision rate [44–47] is too low to be responsible for the population transfer from the bright to the dark Rydberg states. Furthermore, it is difficult for the antiblockade excitation [48,49] to occur in a weak-interaction regime (such as that in our experiment) due to large energy differences between the bright Rydberg state and its nearby states.

We can rule out mechanisms (i) and (ii) for the corresponding quite obvious reasons (a) and (b). However, before ruling out the third case, we estimated the DDI-induced state-exchange collision or the Rydberg state-changing collision rate compared with that in Ref. [40]. The decay rate due to state-changing collisions is proportional to $n_R \times (\text{principal quantum number})^{12}$ [44]. Comparing the experimental conditions of the present study ($n_R \sim 10^9 \text{ cm}^{-3}$, principal quantum number = 32) with Ref. [40] ($n_R \sim 10^9 \text{ cm}^{-3}$, principal quantum number = 111), the state-changing collision rate in our experiment should be on the order of 0.1 Hz based on the rate presented in Ref. [40]. The estimated value is far smaller than the determined value of Γ_{24} shown in Fig. 6. Therefore the state-changing collision is not responsible for the observed two-body decay.

TABLE I. The four dark Rydberg states, which the population in $|32D_{5/2}\rangle$ predominately decays to, due to the BBR at room temperature. Γ_{BBR} is the transition rate induced by the BBR at $T = 300 \text{ K}$. λ_0 and $\Delta E/h$ are the transition wavelength and frequency. τ_{ds} is the lifetime of each state at $T = 300 \text{ K}$. All the values were calculated on the basis of Ref. [50].

State $ nL_J\rangle$	Γ_{BBR} (KHz)	λ_0 (mm)	$\Delta E/h$ (GHz)	τ_{ds} (μs)
$ 30F_{7/2}\rangle$	22.5	1.9	158.4	15.0
$ 31F_{7/2}\rangle$	15.1	4.0	74.0	16.4
$ 34P_{3/2}\rangle$	14.8	1.9	155.4	34.7
$ 33P_{3/2}\rangle$	9.1	4.4	68.63	32.2

As a possible mechanism of the population transfer from the bright to the dark Rydberg states, we considered the super-radiance of transitions induced by the BBR. According to the transition or decay rate from $|32D_{5/2}\rangle$ to the nearby states induced by the BBR at $T = 300 \text{ K}$ and the corresponding wavelength, we found the four most probable dark Rydberg states. Their information is listed in Table I. To explain the measured two-body decay rate by the super-radiance of the transitions induced by the BBR at the room temperature, we made the estimation the decay rate from $|32D_{5/2}\rangle$ to $|33P_{3/2}\rangle$ as an example. In Fig. 6, the decay rate at $n_R = 0.003 \mu\text{m}^{-3}$ was about five times faster than the spontaneous decay rate of $|32D_{5/2}\rangle$ at the room temperature. To estimate the number of particles participating in the super-radiance, i.e., the enhancement factor of the decay rate, we considered an effective range for the cooperative interaction to be $\sim 1/100$ of the transition wavelength λ_0 [41,42]. The number of Rydberg atoms inside the effective volume of $(\lambda_0/100)^3$ was 260 according to the value of λ_0 in Table I and $n_R = 0.003 \mu\text{m}^{-3}$. Hence the super-radiance of the BBR-induced transitions can likely be the mechanism that transfers the population dark Rydberg states faster.

The values of A and B were 0.76Γ and 7.7Γ , respectively, indicating that the DDI strength between a bright Rydberg atom and a dark Rydberg atom is much larger than that between two bright Rydberg atoms. To explain this, we considered that the DDI between a bright Rydberg atom and a dark Rydberg atom is a dipole-dipole interaction of the collision process of $|32D_{5/2}\rangle + |nL_J\rangle \rightarrow |nL_J\rangle + |32D_{5/2}\rangle$, where $|nL_J\rangle$ is one of the dark Rydberg states in Table I. The collision process is resonant, because the two Rydberg atoms just exchange their quantum states. To estimate the strength of each resonant DDI process between a bright Rydberg atom and a dark Rydberg atom in Table I, we followed steps similar to the estimation of A described in Sec. II B and Ref. [27]. The estimated value of B is denoted as \tilde{B} . The relation between \tilde{B} and the dipolar coupling coefficient C_3 is given by

$$\tilde{B} = \frac{2\pi^2 \overline{C_3}}{3} n_a, \quad (22)$$

where $\overline{C_3}$ is the average value of C_3 by considering the branch ratio as a weight value. The branch ratio is the ratio between the decay or excitation rates of the $\Delta m_J = \pm 1, 0$ transitions

TABLE II. Estimation of the strength of the resonant DDI between an atom in state $|32D_{5/2}\rangle$ and another in state $|nL_J\rangle$. \tilde{B} represents the estimated value of the coefficient B according to Eq. (22), \overline{C}_3 is the dipolar coupling coefficient, and \overline{C}_3 is the average value of C_3 weighted by the branch ratio, which is the ratio between the decay or excitation rates of the transitions of $\Delta m_J = \pm 1, 0$ from $|32D_{5/2}\rangle$ to $|nL_J\rangle$. The values of the branch ratio and C_3 were calculated on the basis of Ref. [50].

State $ nL_J\rangle$	m_J	Branch ratio	$C_3/(2\pi)$ (MHz μm^3)	$\overline{C}_3/(2\pi)$ (MHz μm^3)	\tilde{B} (Γ)
$ 30F_{7/2}\rangle$	7/2	0.58	220	146	7.9
	5/2	0.30	63		
	3/2	0.12	10.5		
$ 31F_{7/2}\rangle$	7/2	0.57	687	456	24.7
	5/2	0.31	196		
	3/2	0.12	33		
$ 34P_{3/2}\rangle$	3/2	1	206	206	11.2
$ 33P_{3/2}\rangle$	3/2	1	636	636	34.4

from $|32D_{5/2}\rangle$ to $|nL_J\rangle$. The estimated value of \tilde{B} with \overline{C}_3 of each possible dark Rydberg state is listed in Table II.

Although the simple derivation of using Eq. (22) may cause overestimation of \tilde{B} , a resonant dipole-dipole interaction between the bright and dark Rydberg states can provide an intuitive understanding of the behavior of the system. The resonant DDI strength arises from C_3 , and \overline{C}_3 is the average value of C_3 among the Zeeman states of the same $|nL_J\rangle$. The strength of the nonresonant DDI between two bright Rydberg atoms arises from C_6 . According to Eqs. (10) and (22), we can obtain the ratio of $A/\tilde{B} = (\Omega_c \sqrt{|C_6|/\Gamma}) / (2\overline{C}_3)$. Since \overline{C}_3 in Table II is much larger than $(\Omega_c \sqrt{|C_6|/\Gamma}) / 2 = 2\pi \times 14$ MHz μm^3 in the experiment, \tilde{B} or the estimated value of B is much larger than A . Therefore the experimentally determined value of B indicated that the interaction between a bright Rydberg atom and a dark Rydberg atom is likely a resonant DDI. The lifetime of each of the four possible dark Rydberg states is also listed in Table I. The lifetimes of $|30F_{7/2}\rangle$ and $|31F_{7/2}\rangle$ are significantly shorter than those of $|33P_{3/2}\rangle$ and $|34P_{3/2}\rangle$. The population accumulated in the former is far less than that in the latter. Therefore we think that the dark Rydberg states of $|33P_{3/2}\rangle$ and $|34P_{3/2}\rangle$ contribute to the coefficient B much more than $|30F_{7/2}\rangle$ and $|31F_{7/2}\rangle$ do. The decay time constant, i.e., 31 μs , of the best fit in Fig. 5 is close to the lifetimes of $|33P_{3/2}\rangle$ and $|34P_{3/2}\rangle$ in Sec. II E.

V. CONCLUSION

In this paper, we systematically studied the transmission of a probe field propagating through a Rydberg-EIT system, in which the DDI strength was in the weak-interaction regime. We observed the distorted output of a long Gaussian input probe pulse. Such a phenomenon was unable to be predicted by the theoretical model, which only considered the DDI between the bright Rydberg atoms. According to further measurements, we explained that the distortion was due to the extra attenuation. This phenomenon was caused by a much larger DDI-induced decoherence rate due to the atoms accumulating in the dark Rydberg states. The population in the

dark Rydberg states was transferred from the bright Rydberg state at an unexpectedly high rate. We attribute the high transfer rate to the super-radiance of transitions induced by the blackbody radiation. Using a theoretical model to take into account the effect that population accumulates in some dark Rydberg states and the dark Rydberg atoms cause an extra DDI-induced decoherence rate, we can successfully explain the experimental data.

In conclusion, this study provides a better understanding of the creation of dark Rydberg atoms and their influence on Rydberg polaritons in the Rydberg-EIT system under a long interaction time [51–53], and we believe the effect is universal. This work also points out an obstacle in the realization of BEC with weakly interacting Rydberg polaritons [28,54].

ACKNOWLEDGMENTS

We thank the referees for their comments, which stimulated the discussions in Sec. IV. This work was supported by Grants No. 109-2639-M-007-001-ASP and No. 110-2639-M-007-001-ASP of the Ministry of Science and Technology, Taiwan.

APPENDIX A: EXPERIMENTAL DETAILS FOR THE DETERMINATION OF PARAMETERS

Before the measurement, we determined the experimental parameters in the order of the coupling Rabi frequency $\Omega_c \rightarrow$ the intrinsic decoherence rate $\gamma_0 \rightarrow$ the optical depth or OD α . The parameters were confirmed again after the measurement in the reverse order of that before measurement. The method for the determination of experimental parameters is the same as described in Ref. [28].

We determined Ω_c by the Autler-Townes splitting in the EIT spectrum. To measure the frequency separation between two minima in the spectrum, a low OD value between 1 and 2 was used. The Autler-Townes splitting was obtained by sweeping the probe field frequency using an acousto-optic modulator (AOM). The AOM is not shown in Fig. 1(b). The double-path scheme of the AOM can provide the stable input probe power during the frequency sweeping. The degree of asymmetry of the EIT spectrum enabled us to determine the zero coupling detuning condition [31].

γ_0 was determined by the peak transmission of the long Gaussian probe pulse at the resonance. The input Gaussian pulse e^{-1} full width was 7 μs . According to the non-DDI EIT theory, the peak transmission at the resonance condition is given by

$$T = \exp\left(-\frac{2\alpha\Gamma}{\Omega_c^2}\gamma_0\right). \quad (\text{A1})$$

To avoid the decoherence effect induced by the dipole-dipole interaction (DDI), we used low OD (~ 15 – 20) and probe Rabi frequency Ω_p ($\sim 0.05\Gamma$) for the measurement of γ_0 .

After Ω_c and γ_0 were determined with low OD, we set the OD to the experimental value. We varied the OD by adjusting the repumping laser power during the dark magneto-optical trap (MOT) period and the duration of the period. To have the highest OD used in the experiment, we optimized the repumping laser power and the duration. To have a low value

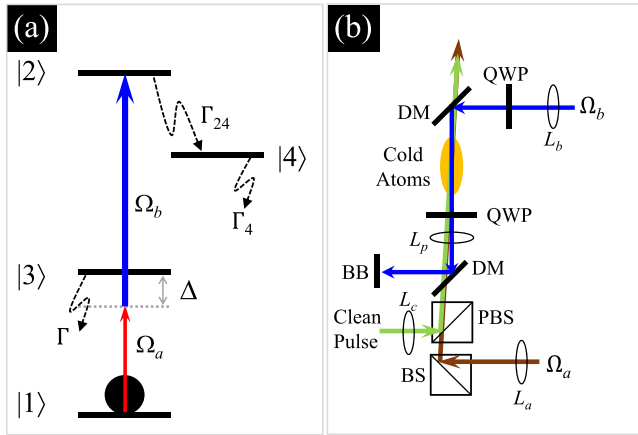


FIG. 9. (a) Scheme of the two-photon transition, which moved the population from $|1\rangle$ to $|2\rangle$. The energy levels here are the same as those in Fig. 1. Please note that $|1\rangle \rightarrow |3\rangle$ is a cycling transition and thus the population of any undesired transition to $|3\rangle$ quickly decayed back only to $|1\rangle$. Ω_a and Ω_b denote the Rabi frequencies of the two square pulses of the laser fields. We kept the frequencies of Ω_a and Ω_b to the two-photon resonance and applied a one-photon detuning Δ of 4Γ to Ω_b or of -4Γ to Ω_a . Γ and Γ_4 are the spontaneous decay rates of $|3\rangle$ and $|4\rangle$, respectively. Γ_{24} is the decay rate from $|2\rangle$ to $|4\rangle$. (b) Optical paths of Ω_a , Ω_b , and a clean pulse in the experiment. Ω_b was the same as that of the coupling field, but had a different frequency. The clean pulse was employed to wipe out the population in $|1\rangle$.

of the OD, we changed the repumping power and the duration of the dark MOT from their optimum values.

We measured the delay time τ_d of the short Gaussian probe pulse to determine the value of OD, i.e., α . The e^{-1} full width of the short input Gaussian pulse was $0.66 \mu\text{s}$. According to the non-DDI EIT theory, τ_d is given by

$$\tau_d = \frac{\alpha\Gamma}{\Omega_c^2}. \quad (\text{A2})$$

APPENDIX B: EXPERIMENTAL DETAILS FOR MEASURING THE DECAY RATE OF THE BRIGHT RYDBERG STATE

We designed an experiment to determine the coefficients C and D in Eq. (20). The decay rate, Γ_{24} , of the atoms in $|2\rangle$ was measured against the atomic density of $|2\rangle$, n_R . We employed the two-photon transition (TPT) scheme rather than the EIT scheme to place the population in $|2\rangle$, because the TPT scheme is able to move a large portion of the atoms in $|1\rangle$ to $|2\rangle$. In the measurement of Γ_{24} , we added the pulses of Ω_a and Ω_b , which formed the TPT pulse, and the clean pulse as shown in Fig. 9(b). The coupling field Ω_c in the EIT experiment was utilized as Ω_b , but we changed its frequency to set the one-photon detuning Δ to 4.0Γ for the TPT as depicted in Fig. 9(a). The frequency of Ω_a was tuned such that the TPT satisfied the two-photon resonance. A weak and resonant probe pulse, denoted as $\Omega_{p'}$, was used in the measurement. The pulse of $\Omega_{p'}$ had the same optical path as the field of Ω_p shown in Fig. 1(b). The absorption coefficient of $\Omega_{p'}$ indicated the number of atoms in $|1\rangle$.

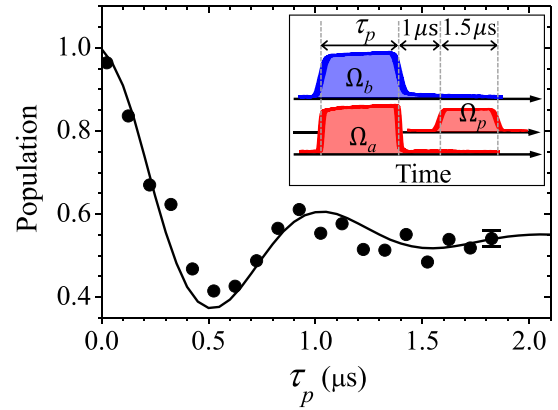


FIG. 10. Experimental demonstration of the two-photon transition (TPT) shown in Fig. 9(a). In the main figure, we plot the remaining population in state $|1\rangle$ after the TPT pulse as a function of the TPT pulse width τ_p . In the inset, we show the timing sequence of the pulses of Ω_a and Ω_b (to drive the TPT) and $\Omega_{p'}$ (to measure the population in $|1\rangle$), as well as their pulse shapes. We set $\Delta = 4.0\Gamma$, α (optical depth) = 0.5, $\Omega_a = \Omega_b = 1.2\Gamma$, and $\Omega_{p'} = 0.08\Gamma$ in the measurement. Circles are the experimental data, which have similar error bars. Only the last data point shows an error bar, which represents the typical value. The black curve is the theoretical prediction of a simple three-level cascade system under the two-photon resonance. The prediction was calculated with the decoherence rate of $6.5 \times 10^{-2}\Gamma$ and the above values of Δ , Ω_a , and Ω_b .

The polarization, propagation direction, and beam size of Ω_b (or $\Omega_{p'}$) were the same as those of Ω_c (or Ω_p) in the EIT experiment. The field of Ω_a was generated by a diode laser, which was injection locked by the same laser that generated the field of Ω_p in the EIT experiment. Hence the two-photon frequency of Ω_a and Ω_b was stable. The pulse of Ω_a was sent to the atom cloud by a PMF after passing through an AOM. The clean pulse was used to remove the population in $|1\rangle$. After passing through the AOM, the clean pulse had a frequency that was resonant to the transition of $|5S_{1/2}, F = 2\rangle \rightarrow |5P_{3/2}, F = 2\rangle$. The polarization of Ω_a and the clean pulse were σ_+ and σ_- , respectively. There was a separation angle of 0.36° (or 0.40°) between the propagation directions of Ω_a (or the clean pulse) and Ω_b . Lens L_a (or L_c) was used together with L_p to make Ω_a (or the clean pulse) a collimated beam with the e^{-1} full width of 3.6 mm (or 4.5 mm). The two beam sizes of Ω_a and the clean pulse were sufficiently large to cover the entire atom cloud.

To study the efficiency of the TPT pulse, i.e., the simultaneous pulses of Ω_a and Ω_b , we varied the TPT pulse duration τ_p and measured the population or atom number right after the TPT pulse as shown in Fig. 10. The population in $|1\rangle$ was determined by the absorption coefficient of $\Omega_{p'}$. The timing sequence of Ω_a , Ω_b , and $\Omega_{p'}$ and their pulse shapes are depicted in the inset. In the main plot, the circles are the experimental data, and the curve is the theoretical prediction of a simple three-level cascade system under the two-photon resonance, i.e., Eqs. (1)–(6) are utilized with $\delta = 0$, $\delta_{\text{DDI}} = 0$, $\gamma_{\text{DDI}} = 0$, $\Omega_p \rightarrow \Omega_a$, and $\Omega_c \rightarrow \Omega_b$. To calculate the prediction, we set Δ , Ω_a , and Ω_b to the experimental values and

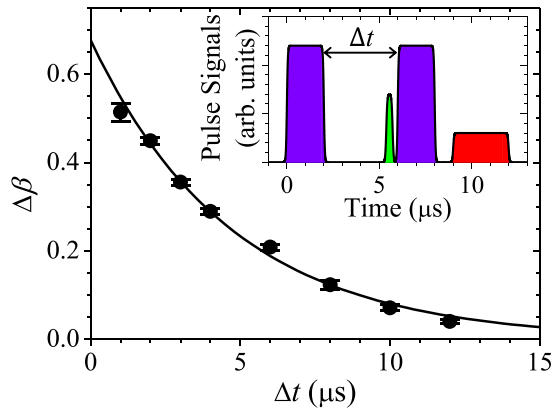


FIG. 11. Determination of the decay rate of the bright Rydberg state $|2\rangle$. The inset shows the time sequence of the measurement. The purple, green, and red areas represent the two-photon transition (TPT) pulses, the clean pulse, and the probe pulse, respectively. In the main plot, circles are the representative data of the difference between the absorption coefficients with and without the second TPT pulse, $\Delta\beta$, as a function of Δt . The black curve is the best fit of an exponential-decay function, which determined the decay rate. In the measurement of these data points, $n_R = 0.002 \mu\text{m}^{-3}$, and the values of Δ , Ω_a , Ω_b , and $\Omega_{p'}$ are the same as those shown in Fig. 10.

adjusted γ_0 to match the experimental data. The fair agreement between the data and the prediction verified the effect of the TPT pulse on the population. According to the study shown in Fig. 10, we decided to use a $1.8\text{-}\mu\text{s}$ TPT pulse with $\Delta = 4.0\Gamma$ and $\Omega_a = \Omega_b = 1.2\Gamma$ in the measurement of Γ_{24} because the transition probability after $\tau_p = 1.8 \mu\text{s}$ was insensitive to the pulse width. Please note that the reduced population in $|1\rangle$ was used to determine the population in $|2\rangle$, but during the TPT pulse the excitation to the intermediate state $|3\rangle$ was not negligible. Nevertheless, since the population in $|3\rangle$ decayed only to $|1\rangle$ in a rather short length of time right after the TPT pulse, the excitation of the population to $|3\rangle$ did not affect the determination of the population in $|2\rangle$.

APPENDIX C: DETAILS ON DETERMINING THE DECAY RATE FROM BRIGHT TO DARK RYDBERG STATES

To obtain the decay rate, Γ_{24} , of the atoms in $|2\rangle$ as a function of the density n_R of the atoms in $|2\rangle$, we performed the measurement described in the first paragraph of Sec. III B. The time sequence of the measurement is depicted in the inset of Fig. 11. We kept the time difference between the peak of the clean pulse and the rising edge of the second TPT pulse to about 400 ns , and that between the falling edge of the second TPT pulse and the rising edge of the probe pulse to about $1 \mu\text{s}$. Each of the two TPT pulses had a duration of $1.8 \mu\text{s}$. The TPT pulse was able to move a fixed fraction (about half) of the atoms from the initial state to the final state, i.e., $|1\rangle \rightarrow |2\rangle$ or $|2\rangle \rightarrow |1\rangle$. The residual atoms either were removed from the system or did not affect the measurement of the decay rate. We varied Δt and measured the absorption coefficient $\Delta\beta$ of the weak probe pulse $\Omega_{p'}$. The representative data of the difference between the values of $\Delta\beta$ with and without the second TPT pulse as a function of Δt are plotted in

Fig. 11. Since the atoms in $|1\rangle$ right before the second TPT were all depleted by the clean pulse, $\Delta\beta$ corresponded to the remaining atoms in $|2\rangle$ after a decay time of Δt .

The data points in Fig. 11 were all taken at $\Delta\alpha = 2.9$, where $\Delta\alpha$ is defined as the difference of the values of OD before and after the first TPT pulse. The two values of OD were about 5.2 and 2.3 in this case. We were able to estimate the density of the atoms in $|2\rangle$ after the first TPT pulse from $\Delta\alpha$ using the following relation:

$$\Delta\alpha = \sigma_0 n_1 L = \sigma_0 n_R L, \quad (\text{C1})$$

where σ_0 is the absorption cross section of the resonant probe transition from $|5S_{1/2}, F = 2, m_F = 2\rangle$ to $|5P_{3/2}, F = 3, m_F = 3\rangle$ and n_1 is the density of the atoms in $|1\rangle$ that were moved to $|2\rangle$ by the first TPT pulse. Consequently, n_1 was equal to the initial Rydberg-atom density n_R , which participated in the decay process of $|2\rangle$. As shown in Fig. 11, we fitted the experimental data taken at a given n_R with an exponential-decay function. The decay time constant of the best fit gave the value of Γ_{24} of the given n_R .

APPENDIX D: PREVIOUS STUDIES ON THE POPULATION TRANSFER TO DARK RYDBERG STATES

Experimental observations of population transfer from one Rydberg state to another, i.e., from a bright Rydberg state to the dark Rydberg state, have been reported in several papers [36–42]. The underlying mechanisms of such transfers can be transitions driven by a microwave field [36], the spontaneous decay induced by blackbody radiation and vacuum fluctuations [37–39], the DDI-induced antiblockade excitation and state-exchange collision assisted by radiation trapping [40], and the super-radiance induced by blackbody radiation (BBR) [41,42]. We discuss these papers and compare their results and experimental conditions with our observations and experimental condition in the following paragraphs.

The authors of Ref. [36] measured the linewidth of the transition from $|45D_{5/2}\rangle$ to $|46D_{5/2}\rangle$, while the entire Rydberg population was initially prepared in $|45D_{5/2}\rangle$. As they moved half of the Rydberg population to $|46P_{3/2}\rangle$ by applying a microwave before the linewidth measurement, the measured linewidth was broadened. The authors explained that the DDI between a $|45D_{5/2}\rangle$ atom and a $|46P_{3/2}\rangle$ atom was stronger than that between two $|45D_{5/2}\rangle$ atoms, resulting in the linewidth broadening. The population transfer from the bright ($|45D_{5/2}\rangle$) to dark ($|46P_{3/2}\rangle$) Rydberg state was intentionally driven by a microwave field, and no accumulative DDI effect was reported in Ref. [36]. Since we did not apply any additional field to move the population from the bright to the dark Rydberg states, the physical mechanism in Ref. [36] is unable to explain the accumulative DDI effect observed in our study.

On the other hand, the population in the dark Rydberg state could be produced via the spontaneous decay from the bright Rydberg state induced by the blackbody radiation and vacuum fluctuation. In Ref. [37], the authors measured the spectrum of the two-photon transition from a ground state to the Rydberg state $|18S_{1/2}\rangle$ with the ^{87}Rb atoms trapped in a three-dimensional (3D) optical lattice. They observed that the measured linewidth was about two orders of magnitude

larger than the expected linewidth due to the DDI between two $|18S_{1/2}\rangle$ atoms. Such a large linewidth was explained by the DDI between an atom in $|18S_{1/2}\rangle$ and another in a nearby Rydberg state, $|17P\rangle$ or $|18P\rangle$. The existence of the population in $|17P\rangle$ and $|18P\rangle$ due to the spontaneous decay from $|18S_{1/2}\rangle$ was further verified by Ref. [38]. In Ref. [39], the authors reported a similar phenomenon with the bright Rydberg state of $|28D_{5/2}\rangle$ and the dark Rydberg states of $|26F_{7/2}\rangle$, $|27F_{7/2}\rangle$, $|29P_{3/2}\rangle$, and $|30P_{3/2}\rangle$. Nevertheless, the spontaneous decay was a one-body process and did not depend on the Rydberg-atom density. As shown in Fig. 6, the observed decay rate linearly depended on the atomic density of the bright Rydberg state. Furthermore, it was also larger than the spontaneous decay rate of $|32D_{5/2}\rangle$ used in our experiment, which was $2\pi \times 7.9$ kHz or $1.3 \times 10^{-3}\Gamma$. Thus the decay from the bright to the dark Rydberg states observed in our study is unable to be explained by the spontaneous decay.

Population transfer from the bright to the dark Rydberg states can also be induced by direct antiblockade excitation [48,49] and state-changing Rydberg collisions [44–47]. In Ref. [40], the authors drove the Rydberg-EIT transition from a ground state to $|111S_{1/2}\rangle$ (the bright Rydberg state) and detected the ions coming from a number of dark Rydberg states (nearby states other than $|111S_{1/2}\rangle$) after an ionization pulse. Compared with the experiment in this paper, the experiment in Ref. [40] was carried out under a high atomic density of 5×10^{12} cm⁻³ and in the strong-interaction regime of $(r_B/r_a)^3 > 1$. Under such an atomic density, the authors of Ref. [40] explained that the effect of radiation trapping [43] was prominent, producing more atoms in the bright Rydberg state. The atoms produced by the radiation trapping had all possible angular momentum

angles. Then, the strong-interaction regime enabled the DDI-induced antiblockade excitation and state-changing collisions to generate dark Rydberg atoms from the bright Rydberg atoms. These dark Rydberg atoms produced ions after an ionization pulse, while the ions were clearly not able to come from the bright Rydberg state $|111S_{1/2}\rangle$. The dark Rydberg atom also resulted in the unexpected reduction of the output probe photon number. The authors stated that the reduction is unable to be explained by the prediction of the blockade effect with only the bright Rydberg state but no dark Rydberg states.

A faster population transfer from the bright to the dark Rydberg state than the spontaneous decay rate can be explained by the super-radiance induced by BBR [41,42]. In Ref. [42], the authors observed the decay of the population from $|nD_{5/2}\rangle$ to $|(n+1)P_{3/2}\rangle$ in laser-cooled cesium Rydberg atoms where the principal quantum number n is 60, 63, and 70. The size of the atom cloud was 550 μ m in diameter. The bright Rydberg state, $|nD_{5/2}\rangle$, is driven by two-photon excitation. $|(n+1)P_{3/2}\rangle$ is one of the dark Rydberg states which is the energetically closest state. The population of the bright and the dark Rydberg state was measured by the state-selective ionization method with a multichannel plate. The authors showed that the decay rate from $|nD_{5/2}\rangle$ to $|(n+1)P_{3/2}\rangle$ became faster with a larger number of bright Rydberg state atoms. For example, when they prepared 2.2×10^4 atoms in $|60D_{5/2}\rangle$, they observed an ~ 2 MHz decay rate which was a nearly 660-fold faster decay rate than the decay rate at room temperature. In this case, the effective interaction volume for the cooperative interaction was on the order of $\sim (\lambda_0/550)^3$, where λ_0 is the transition wavelength of the $|60D_{5/2}\rangle \rightarrow |61P_{3/2}\rangle$ transition, 92.9 mm.

-
- [1] 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).
- [2] D. Comparat, and P. Pillet, Dipole blockade in a cold Rydberg atomic sample, *J. Opt. Soc. Am. B* **27**, A208 (2010).
- [3] M. Saffman, T. G. Walker, and K. Mølmer, Quantum information with Rydberg atoms, *Rev. Mod. Phys.* **82**, 2313 (2010).
- [4] T. Baluktian, B. Huber, R. Löw, and T. Pfau, Evidence for Strong van der Waals Type Rydberg-Rydberg Interaction in a Thermal Vapor, *Phys. Rev. Lett.* **110**, 123001 (2013).
- [5] D. Tiarks, S. Schmidt-Eberle, T. Stolz, G. Rempe, and S. Dürr, A photon-photon quantum gate based on Rydberg interactions, *Nat. Phys.* **15**, 124 (2019).
- [6] H. Gorniaczyk, C. Tresp, J. Schmidt, H. Fedder, and S. Hofferberth, Single-Photon Transistor Mediated by Interstate Rydberg Interactions, *Phys. Rev. Lett.* **113**, 053601 (2014).
- [7] D. Tiarks, S. Baur, K. Schneider, S. Dürr, and G. Rempe, Single-Photon Transistor Using a Förster Resonance, *Phys. Rev. Lett.* **113**, 053602 (2014).
- [8] S. Baur, D. Tiarks, G. Rempe, and S. Dürr, Single-Photon Switch Based on Rydberg Blockade, *Phys. Rev. Lett.* **112**, 073901 (2014).
- [9] J. D. Pritchard, D. Maxwell, A. Gauguet, K. J. Weatherill, M. P. A. Jones, and C. S. Adams, Cooperative Atom-Light Interaction in a Blockaded Rydberg Ensemble, *Phys. Rev. Lett.* **105**, 193603 (2010).
- [10] D. Petrosyan, J. Otterbach, and M. Fleischhauer, Electromagnetically Induced Transparency with Rydberg Atoms, *Phys. Rev. Lett.* **107**, 213601 (2011).
- [11] 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 (London)* **488**, 57 (2012).
- [12] E. Zeuthen, M. J. Gullans, M. F. Maghrebi, and A. V. Gorshkov, Correlated Photon Dynamics in Dissipative Rydberg Media, *Phys. Rev. Lett.* **119**, 043602 (2017).
- [13] D. Jaksch, J. I. Cirac, P. Zoller, S. L. Rolston, R. Côté, and M. D. Lukin, Fast Quantum Gates for Neutral Atoms, *Phys. Rev. Lett.* **85**, 2208 (2000).
- [14] D. S. Weiss, and M. Saffman, Quantum computing with neutral atoms, *Phys. Today* **70**(7), 44 (2017).
- [15] H. Levine, A. Keesling, G. Semeghini, A. Omran, T. T. Wang, S. Ebadi, H. Bernien, M. Greiner, V. Vuletić, H. Pichler, and M. D. Lukin, Parallel Implementation of High-Fidelity Multi-qubit Gates with Neutral Atoms, *Phys. Rev. Lett.* **123**, 170503 (2019).

- [16] Y. O. Dudin and A. Kuzmich, Strongly interacting Rydberg excitations of a cold atomic gas, *Science* **336**, 887 (2012).
- [17] F. Ripka, H. Kübler, R. Löw, and T. Pfau, A room-temperature single-photon source based on strongly interacting Rydberg atoms, *Science* **362**, 446 (2018).
- [18] D. P. Ornelas-Huerta, A. N. Craddock, E. A. Goldschmidt, A. J. Hachtel, Y. Wang, P. Bienias, A. V. Gorshkov, S. L. Rolston, and J. V. Porto, On-demand indistinguishable single photons from an efficient and pure source based on a Rydberg ensemble, *Optica* **7**, 813 (2020).
- [19] Peter Schauß, M. Cheneau, M. Endres, T. Fukuhara, S. Hild, A. Omran, T. Pohl, C. Gross, S. Kuhr, and I. Bloch, Observation of spatially ordered structures in a two-dimensional Rydberg gas, *Nature (London)* **491**, 87 (2012).
- [20] Y.-Y. Jau, A. M. Hankin, T. Keating, I. H. Deutsch, and G. W. Biedermann, Entangling atomic spins with a Rydberg-dressed spin-flip blockade, *Nat. Phys.* **12**, 71 (2016).
- [21] J. Zeiher, R. van Bijnen, P. Schauß, S. Hild, J.-y. Choi, T. Pohl, I. Bloch, and C. Gross, Many-body interferometry of a Rydberg-dressed spin lattice, *Nat. Phys.* **12**, 1095 (2016).
- [22] H. Labuhn, D. Barredo, S. Ravets, S. de Léséleuc, T. Macrì, T. Lahaye, and A. Browaeys, Tunable two-dimensional arrays of single Rydberg atoms for realizing quantum Ising models, *Nature (London)* **534**, 667 (2016).
- [23] A. Omran, H. Levine, A. Keesling, G. Semeghini, T. T. Wang, S. Ebadi, H. Bernien, A. S. Zibrov, H. Pichler, S. Choi, J. Cui, M. Rossignolo, P. Rembold, S. Montangero, T. Calarco, M. Endres, M. Greiner, V. Vuletić, and M. D. Lukin, Generation and manipulation of Schrödinger cat states in Rydberg atom arrays, *Science* **365**, 570 (2019).
- [24] V. Borish, O. Marković, J. A. Hines, S. V. Rajagopal, and M. Schleier-Smith, Transverse-Field Ising Dynamics in a Rydberg-Dressed Atomic Gas, *Phys. Rev. Lett.* **124**, 063601 (2020).
- [25] D. Bluvstein, A. Omran, H. Levine, A. Keesling, G. Semeghini, S. Ebadi, T. T. Wang, A. A. Michailidis, N. Maskara, W. W. Ho, S. Choi, M. Serbyn, M. Greiner, V. Vuletić, and M. D. Lukin, Controlling quantum many-body dynamics in driven Rydberg atom arrays, *Science* **371**, 1355 (2021).
- [26] H. Bernien, S. Schwartz, A. Keesling, H. Levine, A. Omran, H. Pichler, S. Choi, A. S. Zibrov, M. Endres, M. Greiner, V. Vuletić, and M. D. Lukin, Probing many-body dynamics on a 51-atom quantum simulator, *Nature (London)* **551**, 579 (2017).
- [27] S.-S. Hsiao, K.-T. Chen, and I. A. Yu, Mean field theory of weakly-interacting Rydberg polaritons in the EIT system based on the nearest-neighbor distribution, *Opt. Express* **28**, 28414 (2020).
- [28] B. Kim, K.-T. Chen, S.-S. Hsiao, S.-Y. Wang, K.-B. Li, J. Ruseckas, G. Juzeliūnas, T. Kirova, M. Auzinsh, Y.-C. Chen, Y.-F. Chen, and I. A. Yu, A weakly-interacting many-body system of Rydberg polaritons based on electromagnetically induced transparency, *Commun. Phys.* **4**, 101 (2021).
- [29] M. Fleischhauer, A. Imamoglu, and J. P. Marangos, Electromagnetically induced transparency: Optics in coherent media, *Rev. Mod. Phys.* **77**, 633 (2005).
- [30] Y.-W. Lin, H.-C. Chou, P. P. Dwivedi, Y.-C. Chen, and I. A. Yu, Using a pair of rectangular coils in the MOT for the production of cold atom clouds with large optical density, *Opt. Express* **16**, 3753 (2008).
- [31] B. Kim, K.-T. Chen, C.-Y. Hsu, S.-S. Hsiao, Y.-C. Tseng, C.-Y. Lee, S.-L. Liang, Y.-H. Lai, J. Ruseckas, G. Juzeliūnas, and I. A. Yu, Effect of laser-frequency fluctuation on the decay rate of Rydberg coherence, *Phys. Rev. A* **100**, 013815 (2019).
- [32] M.-J. Lee, J. Ruseckas, C.-Y. Lee, V. Kudriašov, K.-F. Chang, H.-W. Cho, G. Juzeliūnas, and I. A. Yu, Experimental demonstration of spinor slow light, *Nat. Commun.* **5**, 5542 (2014).
- [33] I. I. Beterov, I. I. Ryabtsev, D. B. Tretyakov, and V. M. Entin, Quasiclassical calculations of blackbody-radiation-induced depopulation rates and effective lifetimes of Rydberg nS , nP , and nD alkali-metal atoms with $n \leq 80$, *Phys. Rev. A* **79**, 052504 (2009); **80**, 059902(E) (2009).
- [34] D. A. Steck, Rubidium 87 D Line Data, available online at <https://steck.us/alkalidata/> (version 2.2.2, last revised 9 July 2021).
- [35] T. G. Walker and M. Saffman, Consequences of Zeeman degeneracy for the van der Waals blockade between Rydberg atoms, *Phys. Rev. A* **77**, 032723 (2008).
- [36] K. Afrousheh, P. Bohloulou-Zanjani, D. Vagale, A. Mugford, M. Fedorov, and J. D. D. Martin, Spectroscopic Observation of Resonant Electric Dipole-Dipole Interactions between Cold Rydberg Atoms, *Phys. Rev. Lett.* **93**, 233001 (2004).
- [37] E. A. Goldschmidt, T. Boulier, R. C. Brown, S. B. Koller, J. T. Young, A. V. Gorshkov, S. L. Rolston, and J. V. Porto, Anomalous Broadening in Driven Dissipative Rydberg System, *Phys. Rev. Lett.* **116**, 113001 (2016).
- [38] T. Boulier, E. Magnan, C. Bracamontes, J. Maslek, E. A. Goldschmidt, J. T. Young, A. V. Gorshkov, S. L. Rolston, and J. V. Porto, Spontaneous avalanche dephasing in large Rydberg ensembles, *Phys. Rev. A* **96**, 053409 (2017).
- [39] J. de Hond, N. Cisternas, R. J. C. Spreeuw, H. B. van Linden van den Heuvell, and N. J. van Druten, Interplay between van der Waals and dipole-dipole interactions among Rydberg atoms, *J. Phys. B: At. Mol. Opt. Phys.* **53**, 084007 (2020).
- [40] P. Bienias, J. Douglas, A. Paris-Mandoki, P. Titum, I. Mirgorodskiy, C. Tresp, E. Zeuthen, M. J. Gullans, M. Manzoni, S. Hofferberth, D. Chang, and A. V. Gorshkov, Photon propagation through dissipative Rydberg media at large input rates, *Phys. Rev. Res.* **2**, 033049 (2020).
- [41] T. Wang, S. F. Yelin, R. Côté, E. E. Eyler, S. M. Farooqi, P. L. Gould, M. Koštrun, D. Tong, and D. Vranceanu, Super-radiance in ultracold Rydberg gases, *Phys. Rev. A* **75**, 033802 (2007).
- [42] L. Hao, Z. Bai, J. Bai, S. Bai, Y. Jiao, G. Huang, J. Zhao, W. Li, and S. Jia, Observation of blackbody radiation enhanced superradiance in ultracold Rydberg gases, *New J. Phys.* **23**, 083017 (2021).
- [43] D. P. Sadler, E. M. Bridge, D. Boddy, A. D. Bounds, N. C. Keegan, G. Lochead, M. P. A. Jones, and B. Olmos, Radiation trapping in a dense cold Rydberg gas, *Phys. Rev. A* **95**, 013839 (2017).
- [44] A. Derevianko, P. Kómár, T. Topcu, R. M. Kroeze, and M. D. Lukin, Effects of molecular resonances on Rydberg blockade, *Phys. Rev. A* **92**, 063419 (2015).
- [45] T. J. Carroll, K. Claringbould, A. Goodsell, M. J. Lim, and M. W. Noel, Angular Dependence of the Dipole-Dipole Interaction in a Nearly One-Dimensional Sample of Rydberg Atoms, *Phys. Rev. Lett.* **93**, 153001 (2004).
- [46] A. Reinhard, T. Cubel Liebisch, K. C. Younge, P. R. Berman, and G. Raithel, Rydberg-Rydberg Collisions: Resonant Enhancement of State Mixing and Penning Ionization, *Phys. Rev. Lett.* **100**, 123007 (2008).

- [47] M. Eder, A. Lesak, A. Plone, T. Yoda, M. Highman, and A. Reinhard, Quantifying the impact of state mixing on the Rydberg excitation blockade, *Phys. Rev. Res.* **2**, 023234 (2020).
- [48] C. Ates, T. Pohl, T. Pattard, and J. M. Rost, Antiblockade in Rydberg Excitation of an Ultracold Lattice Gas, *Phys. Rev. Lett.* **98**, 023002 (2007).
- [49] T. Amthor, C. Giese, C. S. Hofmann, and M. Weidemüller, Evidence of Antiblockade in an Ultracold Rydberg Gas, *Phys. Rev. Lett.* **104**, 013001 (2010).
- [50] N. Šibalić, J. D. Pritchard, C. S. Adams, and K. J. Weatherill, ARC: An open-source library for calculating properties of alkali Rydberg atoms, *Comput. Phys. Commun.* **220**, 319 (2017).
- [51] A. André, M. Bajcsy, A. S. Zibrov, and M. D. Lukin, Nonlinear Optics with Stationary Pulses of Light, *Phys. Rev. Lett.* **94**, 063902 (2005).
- [52] Y.-W. Lin, W.-T. Liao, T. Peters, H.-C. Chou, J.-S. Wang, H.-W. Cho, P.-C. Kuan, and I. A. Yu, Stationary Light Pulses in Cold Atomic Media and without Bragg Gratings, *Phys. Rev. Lett.* **102**, 213601 (2009).
- [53] J. L. Everett, D. B. Higginbottom, G. T. Campbell, P. K. Lam, and B. C. Buchler, Stationary light in atomic media, *Adv. Quantum Technol.* **2**, 1800100 (2019).
- [54] M. Fleischhauer, J. Otterbach, and R. G. Unanyan, Bose-Einstein Condensation of Stationary-Light Polaritons, *Phys. Rev. Lett.* **101**, 163601 (2008).