Experimental Demonstration of Quantum Stationary Light Pulses in an Atomic Ensemble

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We report an experimental demonstration of the nonclassical stationary light pulse (SLP) in a cold atomic ensemble. A single collective atomic excitation is created and heralded by detecting a Stokes photon in the spontaneous Raman scattering process. The heralded single atomic excitation is converted into a single stationary optical excitation or the single-photon SLP, whose effective group velocity is zero, effectively forming a trapped single-photon pulse within the cold atomic ensemble. The single-photon SLP is then released from the atomic ensemble as an anti-Stokes photon after a specified trapping time. The second-order correlation measurement between the Stokes and anti-Stokes photons reveals the nonclassical nature of the single-photon SLP. Our work paves the way toward quantum nonlinear optics without a cavity.

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I. INTRODUCTION

Coherent atom-light interactions are of great interest in many fields of physics. In particular, a strong interaction at the quantized light level via enhanced atom-light interaction promises significant advances in photonic quantum technologies [1–12]. A cavity quantum electrodynamics system has often been used to achieve the strong atom-photon interaction, in that a photon localized within the cavity may interact with an atom or atoms multiple times [8–13]. In this regard, the stationary light pulse (SLP), in which an optical pulse is trapped within an atomic ensemble with zero group velocity, has begun to attract attention lately [14–22]. However, experimental demonstrations of the SLP to date have been limited to classical light pulses only [19–24]. Here, we report the first experimental demonstration of the quantum SLP in which a heralded single-photon state is first trapped within a cold atomic ensemble without a cavity and then released after a specified trapping time. Our work paves the way toward quantum nonlinear optics without a cavity and may open up possibilities to explore novel photonic quantum matters [25–28].

A coherently driven atomic medium often provides an effect that allows dynamically controlling light propagation through the medium, such as electromagnetically induced transparency (EIT) [29–31]. Via EIT, the group velocity of an optical pulse can be manipulated significantly and, moreover, an optical pulse can be coherently mapped into and out of atomic coherence, enabling atomic quantum memory for photons [32–37]. However, the EIT effect cannot enhance nonlinear interactions between optical pulses because, in the limit of zero group velocity, only atomic coherence persists with no remaining optical excitations. The SLP effect, which may occur in a doubly driven atomic medium, on the other hand, supports nonzero optical excitations having zero group velocity, effectively trapping the optical pulses within the atomic medium. The SLP effect thus allows enhanced nonlinear interactions between optical pulses by increasing the atom-photon interaction time without a cavity [14]. Moreover, it has been theoretically proposed that the SLP effect could lead to novel photonic quantum matters, such as crystals of photons [25] and polariton condensates [26], as well as enable us to simulate dynamics of quantum particles [27,28].

Experimentally, the SLP was first reported using warm atomic vapor [19] and later using cold atoms [20–22]. However, all experimental demonstrations so far have been limited to classical light pulses only, i.e., the trapped optical pulses were strong classical coherent states [19–22], even though all the aforementioned applications of the SLP effect are based on formation of the SLP of quantum optical states, such as the single-photon SLP. In this paper, we report the first experimental demonstration of the quantum SLP in which a heralded single-photon state is first trapped within a cold atomic ensemble without a cavity and then released after a specified trapping time. We demonstrate via the second-order correlation measurement that the nonclassical

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nature of the heralded single-photon state is well preserved during the SLP process.

II. EXPERIMENTAL SETUP

We now elaborate our experimental demonstration of the quantum SLP; see Fig. 1. First, consider the energy level diagram shown in Fig. 1(a). Two hyperfine ground states are denoted as $|g\rangle$ and $|s\rangle$, and two excited states are denoted as $|e_1\rangle$ and $|e_2\rangle$. Initially, all atoms are prepared in the ground state $|g\rangle$. When a weak write pulse (Ω_w) with detuning Δ_1 is applied, the spontaneously Raman scattered Stokes photon $\hat{E}_s$ is emitted. The detection event of the Stokes photon heralds the successful preparation of the single collective atomic excitation or the spin wave $\hat{S}(z,t)$ [38–40].

In order to form the single-photon SLP, two counterpropagating read lasers Ω_r^+ and Ω_r^- are applied; see Fig. 1(b). [Note that $\Omega_r^\pm$ denote the Rabi frequencies of the two read lasers, with $\Omega_r^\pm(t)$ indicating the time-dependent Rabi frequencies.] The two counterpropagating read lasers drive the spin waves in the opposite directions with nonzero photonic excitations $\hat{E}_{as}^\pm(z,t)$, where $\hat{E}_{as}^+$ ($\hat{E}_{as}^-$) is the quantized slow varying electric field operator for the forward (backward) propagating anti-Stokes photon. As the quantized electric field operators $\hat{E}_{as}^\pm$ are coupled with the spin wave $\hat{S}$, the propagation dynamics for the involved fields $\hat{E}_{as}^+$, $\hat{E}_{as}^-$, and $\hat{S}$ is described as a single quasiparticle or the dark-state polariton [17,26,32,33]:

$$\hat{\Psi}_s(z,t) = \cos\theta(t)[\cos\phi(t)\hat{E}_{as}^+(z,t) + \sin\phi(t)\hat{E}_{as}^-(z,t)] - \sin\theta(t)\hat{S}(z,t). \quad (1)$$

The two mixing angles $\theta(t)$ and $\phi(t)$ play important roles in the formation of the single-photon SLP. The mixing angle $\theta(t)$ is defined as $\tan^2\theta(t) = g^2 n/\Omega^2(t)$, where g is the atom-photon coupling constant, n is the atomic density, and $\Omega^2(t) = \Omega_r^{+2}(t) + \Omega_r^{-2}(t)$. The mixing angle $\phi(t)$ is given as $\tan^2\phi(t) = \Omega_r^{-2}(t)/\Omega_r^{+2}(t)$. The single quasiparticle description in Eq. (1) implies that the quantized fields $E_{as}^\pm(z,t)$ and $\hat{S}(z,t)$ are coherently coupled and propagate together with group velocity $v_g = c\cos^2\theta(t)\cos 2\phi(t)$, where c is the light speed in vacuum [17,26]. If the Rabi frequencies of read lasers are balanced, i.e., $\Omega_r^{+2}(t) = \Omega_r^{-2}(t)$, the photonic components $E_{as}^\pm(z,t)$ have zero group velocity but with nonzero amplitudes, hence forming the single-photon SLP or a trapped single photon within an atomic ensemble without a cavity. After a specified trapping time τ, the single-photon SLP may be released from the atomic cloud by turning off one of the read lasers, e.g., $\Omega_r^-(t) \to 0$, as shown in Fig. 1(c).

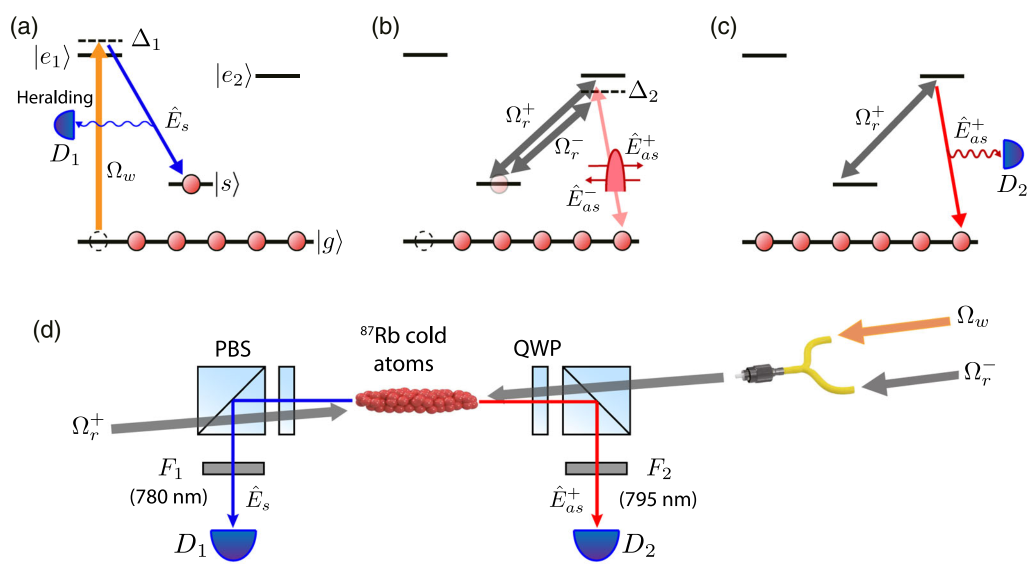

FIG. 1. Experimental sequence and schematic. (a)–(c) The energy level diagram and the experimental sequence. (a) Initially, all atoms are prepared in the ground state $|g\rangle$. When the write pulse Ω_w is applied with detuning Δ_1, creation of a single collective atomic excitation is heralded by the detection event of the Stokes photon $\hat{E}_s$ at detector D_1. (b) From the single collective atomic excitation, the single-photon SLP (denoted by $\hat{E}_{as}^\pm$) is formed by applying two counterpropagating read lasers $\Omega_r^\pm$. Detuning Δ_2 is necessary to suppress higher order atomic coherence in cold atoms. (c) After a specified trapping time τ, the single-photon SLP is released from the atomic ensemble as the anti-Stokes photon $\hat{E}_{as}^+$ by turning off the read lasers Ω_r^- and is detected at detector D_2. (d) Experimental schematic. The optical depth of the Rb MOT is approximately 120. All the optical fields have the same circular polarization in the atomic frame. Because of the phase-matching condition, the read/write lasers and the Stokes/anti-Stokes modes are tilted by approximately 0.35°. The bandpass filter F_1 (F_2) consists of a 780-nm (795-nm) interference filter and a solid etalon filter. The arrival time difference between the heralding photon at D_1 and the released single-photon SLP at D_2 is recorded with a time-correlated single-photon counting (TCSPC) module. PBS stands for polarizing beam splitter; QWP stands for quarter wave plate.

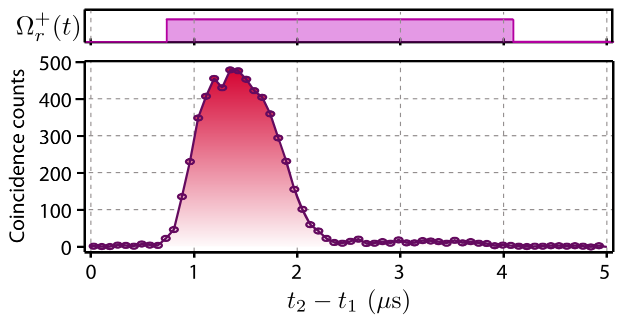

FIG. 2. Heralded generation of a single-photon state. The read laser Ω_r^+ is turned on 0.8 μs after the write pulse. The TCSPC histogram triggered by the Stokes photon at t_1 displays the waveform of the anti-Stokes single photon. The TCSPC histogram is accumulated for 3000 s.

III. HERALDED GENERATION OF SINGLE PHOTONS

We first demonstrate generation of a heralded single-photon state without trapping and then show our result on the single-photon trapping by using the SLP effect described above. The experimental schematic is shown in Fig. 1(d); see Appendix B for details of the experimental setup. For generation of a heralded single-photon state, the write pulse Ω_w with 0.3-mW peak power is applied to the atomic cloud, followed by the counterpropagating read laser Ω_r^+ with 1.2-mW peak power. The write pulse has a Gaussian temporal shape with the full width half maximum (FWHM) of 200 ns and the read laser Ω_r^+ is applied 0.8 μs after the write pulse. Both lasers are well collimated to illuminate the whole atomic ensemble (approximately 1.8 mm in diameter). All the optical fields have the same circular polarization in the atomic frame. The temporal correlation between the heralding Stokes photon $\hat{E}_s$ at D_1 and the anti-Stokes photon $\hat{E}_{as}^+$ at D_2 is measured with a time-correlated single-photon counting (TCSPC) module. Note that the process here is the same as the correlated photon pair generation used in the Duan-Lukin-Cirac-Zoller quantum repeater scheme [38–40]. The experimental data for heralded generation of a single-photon state are shown in Fig. 2. The TCSPC histogram shows the second-order temporal correlation between the Stokes and the anti-Stokes photon. The cross correlation between Stokes and anti-Stokes photons was measured to be $g_{s,as}^{(2)}(0) = 5.43 \pm 0.074$ with the pair detection probability $p_c = 9.6 \times 10^{-3}$. Note that $g_{s,as}^{(2)}(0) > 2$ indicates nonclassical correlation between the Stokes and anti-Stokes photons because the individual photons are thermal in nature.

IV. FORMATION OF THE SINGLE-PHOTON SLP

Let us now describe our experimental demonstration of the single-photon SLP. As described in Fig. 1(b), to form the single-photon SLP from the single collective atomic excitation, two counterpropagating read lasers $\Omega_r^\pm$ must be applied. While the forward read laser Ω_r^+ is on-resonant to $|s\rangle \to |e_1\rangle$, the backward read laser Ω_r^- is slightly detuned by $\Delta_2 = 2\pi \times 4.0$ MHz. This detuning ensures there is no effective higher order Raman coherence [16,20,41], which prevents the formation of stationary light. Also, the Rabi frequencies of the two read lasers need to be sufficiently well balanced, i.e., $|\Omega_r^+(t)| \simeq |\Omega_r^-(t)|$, to ensure formation of the single-photon SLP. We have carefully balanced $\Omega_r^+(t)$ and $\Omega_r^-(t)$ in a separate SLP with an external laser pulse; see the Supplemental Material [42]. Then, after a specified trapping time τ, we turn off one of the read lasers, $\Omega_r^-(t) \to 0$, and the single-photon SLP is released from the atomic medium as the anti-Stokes photon; see Fig. 1(c). The timing sequence for the experiment is schematically shown in Fig. 3(a) and the experimental results are shown in Fig. 3(b) for the

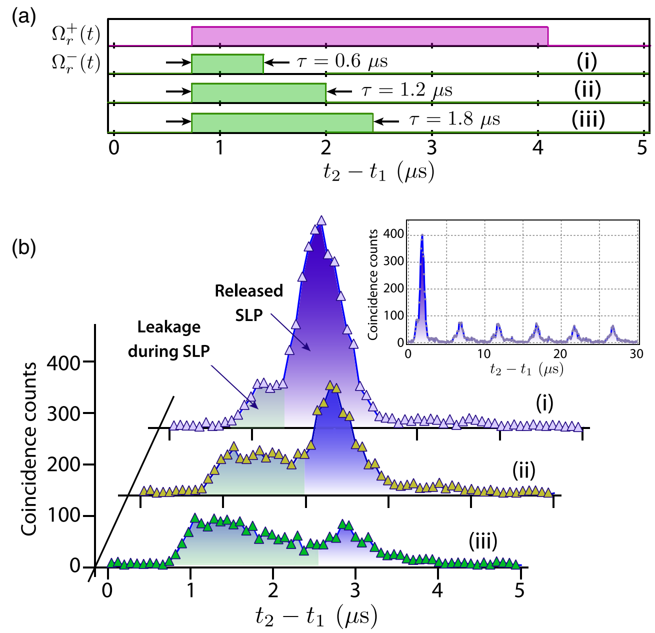

FIG. 3. Formation of the single-photon SLP. (a) The two counter-propagating read lasers Ω_r^+ and Ω_r^- are simultaneously turned on 0.8 μs after the write pulse. After a specified trapping time τ, Ω_r^- is turned off and released is the heralded single-photon from the atomic cloud as the anti-Stokes photon. The trapping times are, (i) $\tau = 0.6\ \mu$s, (ii) $\tau = 1.2\ \mu$s, (iii) $\tau = 1.8\ \mu$s. (b) The TCSPC histogram shows the waveform of the released single-photon. During the formation of the single-photon SLP, emission of the anti-Stokes photon is greatly suppressed, albeit showing some leakage. When the read laser Ω_r^- is turned off, the remaining single-photon SLP is released. Each TCSPC histogram is accumulated for 3000 s. Inset shows the extended region for case (i), in which uncorrelated counts are also presented. Since the write/read pulses are applied at every 5 μs, the coincidence count peaks appear at every 5 μs in the TCSPC histogram. The pronounced peak at 2 μs is due to the quantum-correlated photon pair generated by the corresponding write/read pulse sequence. The other low-lying periodic peaks represent uncorrelated random coincidence counts due to a Stokes photon by a write pulse and an anti-Stokes photon by subsequent read pulses.

trapping times $\tau = 0.6$, 1.2, and 1.8 μs. During the single-photon trapping time τ, we observe a small but non-negligible leakage. We find that the amount of leakage for the single-photon SLP experiment is larger when compared with the classical SLP experiment. Unlike the classical SLP experiment, the single atomic excitation heralded upon the detection of the Stokes photon is spread over the whole atomic medium. Therefore, the stored excitation at the edges of the medium is more likely to be leaked during the SLP formation. See the Supplemental Material [42] for the classical SLP experiment and for the numerical simulation results for the single-photon SLP.

From the TCSPC histograms, as in Fig. 3(b), we obtain the second-order correlation $g^{(2)}_{s,as}(0)$ between the Stokes and the anti-Stokes photons and the relative intensity of the anti-Stokes photon for different single-photon trapping times. The measured second-order correlation results are shown in Fig. 4(a). The second-order correlation $g^{(2)}_{s,as}(0)$ calculated with the raw data (without subtracting any noise counts) tends to decrease with increasing trapping times, but, once the noise counts are subtracted, $g^{(2)}_{s,as}(0)$ values are rather stable. The relative contribution from the noise to the calculated second-order correlation is evidenced in Fig. 4(b), which shows that

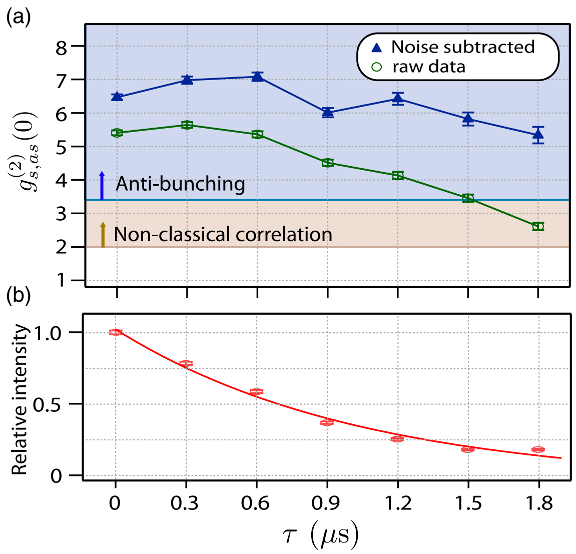

FIG. 4. Nonclassical nature of the single-photon SLP. (a) The second-order correlation $g^{(2)}_{s,as}(0)$ between Stokes and anti-Stokes photons as a function of the single-photon trapping time τ. The raw $g^{(2)}_{s,as}(0)$ was calculated without subtracting any noise counts. Unwanted noise counts due to unfiltered strong read lasers were independently measured and subtracted for the calculation of the noise-subtracted $g^{(2)}_{s,as}(0)$. The bound for nonclassical correlation, calculated from the Cauchy-Schwarz inequality, is $g^{(2)}_{s,as}(0) = 2$, and the bound for antibunching of the heralded anti-Stokes photon is $g^{(2)}_{s,as}(0) = 2 + \sqrt{2}$. (b) The relative intensity of the anti-Stokes photon as a function of τ. The SLP decay rate of $\gamma_\tau = 2\pi \times 154$ kHz was obtained from the exponential decay fit (solid line).

the relative intensity of the anti-Stokes photon is decreasing as a function of the trapping times. Thus, we attribute the main mechanism for the decay of the second-order correlation mostly to the loss associated with the ground-state dephasing and the phase mismatch.

As mentioned earlier, the bound for nonclassical correlation between the Stokes and the anti-Stokes photons is $g^{(2)}_{s,as}(0) = 2$. In addition, the lower bound for the anti-Stokes photon to show antibunching is given by $g^{(2)}_{s,as}(0) = 2 + \sqrt{2}$ with certain assumptions [42]. The noise-subtracted experimental data in Fig. 4(a) all violate the equality for antibunching, signaling that the nonclassical nature of a single-photon state is maintained during the single-photon SLP process within the atomic medium. Note that it is possible to achieve higher $g^{(2)}_{s,as}$ values by simply reducing the power of the write pulse, but at the cost of lowering the pair detection probability p_c [43].

V. CONCLUSION

We have reported the first experimental demonstration of nonclassical SLP in a cold atomic ensemble. A single collective atomic excitation is created and heralded by detecting a Stokes photon in the spontaneous Raman scattering process. The heralded single atomic excitation is then converted into a single stationary optical excitation or the single-photon SLP, whose effective group velocity is zero, effectively forming a trapped single-photon pulse within the cold atomic ensemble. The single-photon SLP is then released from the atomic ensemble as an anti-Stokes photon after a specified trapping time. The second-order correlation measurement between the Stokes and anti-Stokes photons reveals the nonclassical nature of the single-photon SLP. Our work paves the way toward quantum nonlinear optics without a cavity [14], simulating the dynamics of quantum particles [27,28], as well as novel photonic quantum matters, such as crystals of photons [25] and polariton condensates [26].

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APPENDIX A: PREPARATION OF THE COLD ATOMIC ENSEMBLE

A cold atomic ensemble of 87 Rb is prepared by a 2D magneto-optical trap (MOT) in a glass cell ultrahigh vacuum chamber (approximately 10^{-9} Torr) [44,45]. After the initial loading of the atoms, the MOT coils are turned off to reduce the effect due to the stray magnetic fields. The temporal dark MOT and compression techniques are applied, resulting in a high optical depth (OD) cold atom cloud about 20 mm in length and 1.8 mm in diameter [45]. All atoms are initially prepared in the ground state, $|g\rangle \equiv |5S_{1/2}, F = 1\rangle$. The other relevant atomic levels are $|s\rangle \equiv |5S_{1/2}, F = 2\rangle$, $|e_1\rangle \equiv |5P_{3/2}, F' = 2\rangle$, and $|e_2\rangle \equiv |5P_{1/2}, F' = 2\rangle$. The OD is measured as high as approximately 120 from the two-level transmission spectrum between $|g\rangle$ and $|e_2\rangle$ [45].

APPENDIX B: EXPERIMENTAL DETAILS

The write pulse Ω_w has the Gaussian temporal shape with 200 ns at FWHM, the peak power of 0.3 mW, and has detuning $\Delta_1 = 2\pi \times 134.9$ MHz. The read lasers have the rectangular temporal shapes with 1.2-mW peak power. Both the write and the read lasers are well collimated to illuminate the whole ensemble (approximately 1.8 mm in diameter). The collection diameters of the Stokes and anti-Stokes photons are defined by single-mode optical fibers and set to be approximately 250 μm at the medium. The bandwidth of the solid etalon filters is 470 MHz. The total channel efficiency is estimated to be approximately 0.17, which includes the single-mode fiber coupling efficiency of 0.6, the spectral filter transmittance of 0.65, the multimode fiber (coupled to the single-photon detector) coupling efficiency of 0.8, and the detector quantum efficiency of 0.55.

The whole experimental sequence is repeated at the period of 200 ms. Each experimental sequence consists of the cold atom preparation window and the main experimental window. The duration of atom preparation is 199.8 ms and the experimental window is set to be as short as 0.2 ms to minimize the variations of the medium properties, such as OD and the ground-state dephasing rate γ_{gs}. During the experimental window, the single-photon detectors (D_1 and D_2) are gated to the sequence of the single-photon SLP experiment. The duration of the SLP experimental sequence is 5 μs, so there are 40 single-photon SLP trials within each experimental window. Typically, we operate the experiment at the regime of the pair detection probability of $p_c \sim 10^{-2}$.

APPENDIX C: FORMATION OF THE SLP

For stable formation of the SLP, three conditions must be satisfied: (i) the phase-matching condition, (ii) suppression of higher order atomic coherence, and (iii) matching of the read laser Rabi frequencies. First, the phase-matching condition requires that all the fields involved in the stationary light formation satisfy energy and momentum conservation conditions [20,22]. The phase-matching condition for the SLP is given as $\vec{\kappa} = \vec{k}_{as}^{+} - \vec{k}_{r}^{+} = \vec{k}_{as}^{-} - \vec{k}_{r}^{-}$, where the initial spin wave is set as $\vec{\kappa} = \vec{k}_w - \vec{k}_s$. Here, $\vec{k}_w$, $\vec{k}_s$, $\vec{k}_{as}^{\pm}$, and $\vec{k}_{r}^{\pm}$ are the wave vectors for Ω_w, $\hat{E}_s$, $\hat{E}_{as}^{\pm}$, and $\Omega_r^{\pm}$, respectively. To satisfy the above phase-matching condition, the read lasers and the anti-Stokes collection modes are tilted by 0.35°. Second, the SLP is only supported when the forward (backward) read laser couples only with the forward (backward) anti-Stokes photon. Since the coupling between the forward (backward) read laser and the backward (forward) anti-Stokes photon is mediated by higher order atomic coherence, the higher order coherence should be suppressed. While the higher order atomic coherence is not induced in hot atoms due to the Doppler effect, it is induced in cold atomic medium. The higher order coherence in cold atoms, however, can be suppressed by providing the detuning Δ_2 for the backward read laser Ω_r^{-} so that the higher order coherence is dephased fast [16,20,41]. In our experiment, we set $\Delta_2 = 2\pi \times 4.0$ MHz, which is much larger than the EIT width. Finally, the effective Rabi frequencies of the two read lasers should be matched because the mixing angles and the group velocity for the stationary dark state polariton are controlled by the Rabi frequencies ($\Omega_r^{\pm}$) of the corresponding read lasers. In order to match the Rabi frequencies of the two read lasers, we performed a separate SLP experiment with a classical laser input; see the Supplemental Material. We also obtained the other relevant experimental parameters for the single-photon SLP experiment, such as OD ~ 100 and the ground-state dephasing rate $\gamma_{gs} = 2\pi \times 3.0$ kHz, by fitting the classical SLP experiment results with the numerical simulations of the coupled Maxwell-Bloch equations [20,46].

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