Photon-Photon Interactions via Rydberg Blockade

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We develop the theory of light propagation under the conditions of electromagnetically induced transparency in systems involving strongly interacting Rydberg states. Taking into account the quantum nature and the spatial propagation of light, we analyze interactions involving few-photon pulses. We show that this system can be used for the generation of nonclassical states of light including trains of single photons with an avoided volume between them, for implementing photon-photon gates, as well as for studying many-body phenomena with strongly correlated photons.

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The phenomenon of electromagnetically induced transparency (EIT) [1] in systems involving Rydberg states [2] has recently attracted significant experimental [3–10] and theoretical [11–21] attention. While EIT allows for strong atom-light interactions without absorption, Rydberg states provide strong long-range atom-atom interactions. Therefore, the resulting combination of EIT with Rydberg atoms is ideal for implementing mesoscopic quantum gates [2,16] and for inducing strong photon-photon interactions, with applications to photonic quantum information processing [2,11–14,19–22] and to the realization of many-body phenomena with strongly interacting photons [23]. At the same time, the many-body theoretical description of EIT with arbitrarily strongly interacting Rydberg atoms, taking into account the full quantum dynamics and the spatial propagation of light, has not been reported previously.

In this Letter, we develop such a theory by analyzing the problem for at most two incident photons, which, in turn, provides intuition for understanding the full multiphoton problem. We show that Rydberg atom interactions induce photon-photon interactions, which, below a critical interphotonic distance, turn the EIT medium into an effective two-level medium. This can be used to implement photon-atom and photon-photon phase gates and to enable deterministic single-photon sources. Furthermore, our novel nonperturbative analysis reveals a possibility of photons behaving as "hard-sphere" objects with strong anticorrelations characterized by an avoided volume, which could lead to a number of interesting many-body phenomena.

The basic physics is illustrated by a simple case [Fig. 1(b)], in which a single-photon wave packet $\mathcal{E}$ propagates in an EIT medium [level scheme in Fig. 1(a)] with a central control atom at $z = 0$ prepared in a Rydberg state $|r'\rangle$. Atoms in another Rydberg state $|r\rangle$, coupled by the EIT control laser [Fig. 1(a)], experience a van der Waals potential $V(z) = C_6/z^6$ due to the interaction with the control atom, which is decoupled from the applied fields. Alternatively, one could apply an electric field to induce dipole moments in states $|r\rangle$ and $|r'\rangle$, resulting in $V \propto 1/z^3$ [2].

Far away from $z = 0$, the incident photon propagates in a standard EIT medium. This medium features a control field with single-photon detuning Δ and Rabi frequency Ω, which creates a frequency window, in which the incident photon propagates with negligible absorption, near-unity refractive index, and reduced group velocity v_g [1,24]. In the vicinity of $z = 0$, however, the state $|r\rangle$ is shifted so strongly out of resonance that the photon sees only a two-level ($|g\rangle$, $|e\rangle$) medium with transition linewidth 2γ. As we will derive below, the critical z, at which the interaction is equal to the EIT linewidth $\Omega^2/|\gamma + i\Delta|$ [1], separates these two regimes and corresponds to the Rydberg blockade radius [11,25]. When $\Delta = 0$, the resonant blockade radius z_b is thus defined by $V(z_b) = \Omega^2/\gamma$ ($\hbar = 1$), while for $|\Delta| \gg \gamma$, we define the off-resonant blockade radius z_B

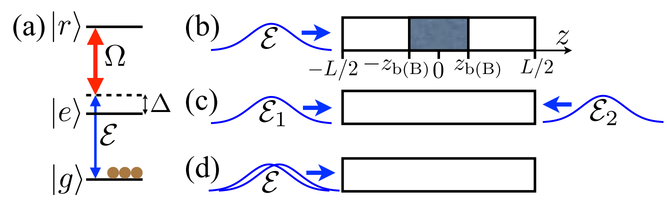

FIG. 1 (color online). (a) EIT level scheme, in which a ground state $|g\rangle$ (the initial state of each atom), an excited state $|e\rangle$, and a Rydberg state $|r\rangle$ are coupled by a quantum probe field $\mathcal{E}$ and a classical control field with Rabi frequency Ω and single-photon detuning Δ. (b) Interaction of one photon with a Rydberg excitation stored at $z = 0$, which modifies the propagation within the blockade region $|z| < z_{b(B)}$. (c),(d) Interaction of two counterpropagating (c) or copropagating (d) photons.

via $V(z_B) = \Omega^2/\Delta$ (we assumed $\Delta/C_6 > 0$). The propagation becomes a one-dimensional problem (see Ref. [26] for 3D effects) if the transverse extent of the photon is smaller than $z_{b(B)}$, which can be satisfied via tight focusing or by using waveguides [27–30]. Since the blockade region extends over $2z_{b(B)}$ [Fig. 1(b)], the presence of the control atom locally creates an absorbing or refractive medium with optical depth $d_{b(B)} = 2dz_{b(B)}/L$, where d is the resonant optical depth of the $|g\rangle - |e\rangle$ medium ($\Omega = 0$) of length L. Interesting effects occur at large blockaded optical depths $d_{b(B)}$. On resonance, assuming $d_b \gg 1$, the $|r'\rangle$ atom causes complete scattering of the incoming photon. Off resonance, for $d_B \gg 1$ and $d_B(\gamma/\Delta)^2 \ll 1$, the $|r'\rangle$ atom imprints a phase $\sim d_B\gamma/\Delta$ on the probe photon and reduces its group delay by $\sim d_B\gamma/\Omega^2$, as its group velocity is increased to the speed of light c within the blockade region.

In the off-resonant case, this simple system has direct practical applications. First, by encoding a qubit in the ground and $|r'\rangle$ states of the central control atom, one can implement a phase gate between the probe photon and the atom. Second, the protocol of Ref. [31] allows us to implement a phase gate between two photons by successively sending them past the control atom that is appropriately prepared and manipulated between the passes. Selective manipulation of the control atom can be achieved particularly simply if it is of a different species or isotope. Third, a phase gate between two photons can also be achieved by storing one of them in the $|r'\rangle$ state of the control atom and sending the other one through the medium. While storing a photon in a single atom is difficult, the same effect can be achieved by storing [24,32] the photon in a collective $|r'\rangle$ excitation (see below).

The results of this simple problem can be extended to the case of multiphoton EIT propagation in Rydberg media. First, off-resonance, two counterpropagating photons [Fig. 1(c)] can pick up a phase $\sim d_B\gamma/\Delta$, enabling the implementation of a two-photon phase gate [12,14]. Second, a pulse of copropagating photons [Fig. 1(d)] will evolve into a nonclassical state corresponding to a train of single photons [19] and exhibiting correlations similar to those of hard-sphere particles with radius $z_{b(B)}/2$. These correlations arise from scattering of photon pairs within the blockade region. Third, in the regime where z_b is larger than the EIT-compressed pulse length σ, both co- and counterpropagating resonant setups are usable as single-photon sources since all but one excitation will be extinguished. In the following, we present a detailed theoretical analysis of these phenomena.

Interaction of a photon with a stationary excitation.— We begin by detailing the solution of the problem of a single-photon wave packet with carrier wave vector k propagating in a medium where state $|r\rangle$ experiences a potential $V(z)$ [Fig. 1(b)]. Treating the medium in a one-dimensional continuum approximation, working in the dipole and rotating-wave approximations, and adiabatically eliminating the polarization on the $|g\rangle - |e\rangle$ transition, the slowly varying electric field amplitude $\mathcal{E}$ of the single-photon wave packet and the polarization S on the $|g\rangle - |r\rangle$ transition obey [24,32]

$$(\partial_t + c\partial_z)\mathcal{E}(z,t) = -\frac{g^2 n}{\Gamma}\mathcal{E}(z,t) - \frac{g\sqrt{n}\Omega}{\Gamma}S(z,t), \tag{1}$$

$$\partial_t S(z,t) = -iU(z,t) - \frac{\Omega^2}{\Gamma}S(z,t) - \frac{g\sqrt{n}\Omega}{\Gamma}\mathcal{E}(z,t). \tag{2}$$

Here $\Gamma = \gamma - i\Delta$, $U(z,t) = V(z)S(z,t)$, g is the atom-field coupling constant, and n is the atomic density. We have neglected the depletion of state $|g\rangle$ and the finite lifetime of the Rydberg state $|r\rangle$, which is typically much longer than the propagation times considered here [2]. Assuming that all atoms are in state $|g\rangle$ before the arrival of the photon, Eqs. (1) and (2) can be solved to give

$$\mathcal{E}\left(\frac{L}{2},t\right) = \int_{-\infty}^{\infty} d\omega \tilde{\mathcal{E}}\left(-\frac{L}{2},\omega\right) \times \exp\left[-i\omega\left(t - \frac{L}{c}\right) + i\frac{k}{2}\int_{-L/2}^{L/2} dz\chi(z,\omega)\right], \tag{3}$$

where the susceptibility χ is

$$\chi(z,\omega) = \frac{1}{kL}\frac{d\gamma[\omega - V(z)]}{\Omega^2 - (\Delta + i\gamma)[\omega - V(z)]} \tag{4}$$

and $\tilde{\mathcal{E}}(-L/2,\omega)$ is the Fourier transform of the wave packet incident at $z = -L/2$. For small ω, the medium becomes effectively two-level (i.e., Ω plays no role) if $|V| \gg \Omega^2/|\Delta + i\gamma|$, hence our definition of $z_{b(B)}$.

For narrowband pulses, we expand χ in ω and, assuming $|\Delta| \gg \gamma$ and $L \gg 2z_B$, reduce Eq. (3) to

$$\mathcal{E}(L/2,t) \approx \mathcal{E}(-L/2, t - L'/v_g)e^{i\varphi - \eta}, \tag{5}$$

where $v_g \approx c\Omega^2/(g^2 n) = 2\Omega^2 L/(d\gamma)$ is the EIT group velocity. In order to avoid the Raman resonance at $V + \Omega^2/\Delta = 0$, we assumed $\Delta/C_6 > 0$. Since the photon travels at c within the blockade region, the group delay comes from a reduced medium length $L' = L - \frac{7}{9}\pi z_B \approx L - 2z_B$. Additionally, the emergence of a two-level medium within $|z| < z_B$ gives an intensity attenuation of $e^{-2\eta}$ with $2\eta = \frac{5\pi}{18}d_B(\gamma/\Delta)^2 \approx d_B(\gamma/\Delta)^2$ and a picked-up phase of $\varphi = -\frac{\pi}{6}d_B(\gamma/\Delta) \approx -\frac{1}{2}d_B(\gamma/\Delta)$. Thus, with $d_B \gg 1$ and a properly chosen $|\Delta| \gg \gamma$, one can get a considerable phase and/or change in group delay without significant absorption. For the same derivation on resonance ($\Delta = 0$), the main effect is an intensity attenuation of $\approx \exp(-d_b)$, as expected for a two-level medium of length $2z_b$.

It is straightforward to extend our analysis to a delocalized $|r'\rangle$ excitation, i.e., a spin wave, that is spread over many atoms. Far off resonance, the effect of the control atom is independent of its position, such that a single control atom and a corresponding spin wave affect the incident photon identically. On resonance, with $d_b \gg 1$, the $|r'\rangle$ spin wave causes complete scattering of the incoming photon.

Interaction of propagating photons.—We now consider the problem of propagating photons interacting with each other. Regarding $\mathcal{E}$ and S in Eqs. (1) and (2) as operators with same-time commutation relations $[\mathcal{E}(z), \mathcal{E}^\dagger(z')] = [S(z), S^\dagger(z')] = \delta(z - z')$ [32] and taking $U(z) = \int dz' V(z - z') S^\dagger(z') S(z') S(z)$, Eqs. (1) and (2) become Heisenberg operator equations [33] for the case of photons copropagating in a Rydberg EIT medium [Fig. 1(d)]. Alternatively, for the case of two counterpropagating photons [Fig. 1(c)], we define operators $\mathcal{E}_{1(2)}$ and $S_{1(2)}$ for the right- (left-)moving photon. For S_1, $U(z) = \int dz' V(z - z') S_2^\dagger(z') S_2(z') S_1(z)$, and vice versa for S_2.

Since the physics of two counterpropagating photons is similar to the spin-wave problem above, we begin our analysis with this case [Fig. 1(c)]. Letting $|\psi(t)\rangle$ be the two-excitation wave function [34], we define $ee(z_1, z_2, t) = \langle 0|\mathcal{E}_1(z_1)\mathcal{E}_2(z_2)|\psi(t)\rangle$, $es(z_1, z_2, t) = \langle 0|\mathcal{E}_1(z_1) S_2(z_2)|\psi(t)\rangle$, $se(z_1, z_2, t) = \langle 0|S_1(z_1)\mathcal{E}_2(z_2)|\psi(t)\rangle$, and $ss(z_1, z_2, t) = \langle 0|S_1(z_1) S_2(z_2)|\psi(t)\rangle$. Equations (1) and (2) then yield a system of equations for these four variables. Defining $es_\pm = (es \pm se)/2$, one finds that es_- is small and does not significantly affect the dynamics. Dropping es_-, defining center-of-mass and relative coordinates $R = (z_1 + z_2)/2$ and $r = z_1 - z_2$, and taking a Fourier transform in time, one obtains $c\partial_r \mathbf{v} = \mathbf{M}(r, \omega)\mathbf{v}$, where $\mathbf{v} = \{ee(R, r, \omega), es_+(R, r, \omega)\}$ and

$$\mathbf{M}(r, \omega) = \begin{bmatrix} i\frac{w}{2} - \frac{g^2 n}{\Gamma} & -\frac{g\sqrt{n}\Omega}{\Gamma} \\ -\frac{g\sqrt{n}\Omega}{\Gamma} & i\omega - \frac{\Omega^2}{\Gamma} + \frac{ig^2 n[\omega - V(r)]}{2\Omega^2 + iV(r)\Gamma - i\omega\Gamma} \end{bmatrix}. \quad (6)$$

R enters only through boundary conditions and is, thus, not important in the present case. For narrowband pulses, we can expand $\mathbf{M}(r, \omega) \approx \mathbf{M}_0(r) + \omega \mathbf{M}_1(r)$, with

$$\mathbf{M}_0 = -\frac{1}{\Gamma}\begin{bmatrix} g^2 n & g\sqrt{n}\Omega \\ g\sqrt{n}\Omega & \Omega^2 + g^2 n \mathcal{V} \end{bmatrix}, \quad (7a)$$

$$\mathbf{M}_1 = i\begin{bmatrix} \frac{1}{2} & 0 \\ 0 & 1 - 2g^2 n \frac{\Omega^2 \mathcal{V}^2}{\Gamma^2 V^2} \end{bmatrix}. \quad (7b)$$

Here we defined the effective potential $\mathcal{V} = \Gamma V/(\Gamma V - i2\Omega^2)$. Outside (inside) the blockade region, $\mathcal{V} \approx i\Gamma V/(2\Omega^2)$ ($\mathcal{V} \approx 1$). For $|r| \gg z_{b(\mathrm{B})}$, the two photons propagate as dark-state polaritons [24]; i.e., we have $es_+/ee = -g\sqrt{n}/\Omega$, which is an eigenstate of $\mathbf{M}_0$ with eigenvalue 0. Since $g\sqrt{n} \gg \Omega$, the group velocity can be read out from the last entry of $\mathbf{M}_1$, which gives twice the EIT group velocity v_g since the two polaritons propagate towards each other. Within the blockade radius, where $\mathcal{V} \approx 1$ and $\mathcal{V}/V \approx 0$, the polariton solution ceases to be an eigenstate of $\mathbf{M}_0$, and Eq. (7b) predicts a speed up to $\sim c$. Since the time $\sim z_{b(\mathrm{B})}/c$ it takes to cross the blockade region is much less than the inverse width of the EIT window, the dynamics is highly nonadiabatic (see below) such that the main result of the interactions is a picked-up factor of $\exp[-\int dr g^2 n \mathcal{V}(r)/(c\Gamma)] = \exp(i\varphi - \eta)$. This is a generalization of the result of Refs. [12,14] (where $\mathcal{V} \propto V$) beyond the perturbative regime.

On resonance, $2\eta \approx d_b$. Thus, analogously to the spin-wave problem above, the entire EIT-compressed two-particle wave function decays provided it fits inside the medium and $d_b \gg 1$. The resulting single-excitation state is a statistical mixture of right- and left-moving excitations.

Off resonance, es_+ picks up $\varphi \approx -\frac{\pi}{2^{1/6}6}\frac{\gamma}{\Delta} d_\mathrm{B} \approx -\frac{\gamma}{2\Delta} d_\mathrm{B}$ and $\eta \approx \frac{5\pi}{2^{1/6}36}\frac{\gamma^2}{\Delta^2} d_\mathrm{B} \approx \frac{\gamma^2}{2\Delta^2} d_\mathrm{B}$. Additionally, the off-diagonal terms in $\mathbf{M}_0$ result in a small admixture of the bright-state polariton [24], which decays after the wave function exits the blockade region.

To verify these conclusions, we show in Fig. 2 and in the movie in the Supplemental Material [35] the results of numerical solutions of the full equations for ee, es, se, and ee in the off-resonant case. Despite the bright-polariton-induced oscillations of ee inside and near the blockade region [35], the final phase of the outgoing two-photon pulse perfectly agrees with our analytical prediction [Fig. 2(e)]. While also showing good agreement with the analytical result, the obtained loss is slightly larger due to the bright-state polariton admixture, which was neglected within the above approximate treatment.

Provided the EIT-compressed two-particle wave function fits inside the medium, this process thus allows for the implementation of a nearly lossless phase gate between two photons. Taking a specific example of cold Rb atoms with $|e\rangle = 5^2P_{1/2}$ and $|r\rangle = 70^2S_{1/2}$ and using $\Omega/2\pi = 2$ MHz [10] and $\Delta = 20\gamma$, we find $z_\mathrm{B} = 15\ \mu$m, which, for a dense cloud with $n = 10^{12}\ \mathrm{cm}^{-3}$, gives $d_\mathrm{B} = \frac{3}{2\pi}\lambda^2(2z_\mathrm{B})n \approx 9$ [36,37]. This yields a significant phase of $\varphi \approx -0.2$ and a very small attenuation $2\eta \approx 0.02$. One can increase d_B further by using photonic waveguides [27–30] and working with a Bose-Einstein condensate [30].

In the copropagating case, we define $ee(z_1, z_2, t) = \langle 0|\mathcal{E}(z_1)\mathcal{E}(z_2)|\psi(t)\rangle$, $es(z_1, z_2, t) = \langle 0|\mathcal{E}(z_1)S(z_2)|\psi(t)\rangle$, and $ss(z_1, z_2, t) = \langle 0|S(z_1)S(z_2)|\psi(t)\rangle$ [Fig. 1(d)]. Defining $es_\pm(z_1, z_2) = [es(z_1, z_2) \pm es(z_2, z_1)]/2$, dropping es_-, and taking the Fourier transform in time, we obtain $c\partial_R \mathbf{v} = 2\mathbf{M}(r, \omega)\mathbf{v}$. That is, the only difference from the counterpropagating case is the replacement of ∂_r with $(1/2)\partial_R$. The resulting equations can be solved separately at each r. As before, outside the blockade radius, the two-photon dark-state polariton propagates with group velocity v_g. Inside the blockade radius, $\mathbf{M}_0$ results in exponential attenuation of the two-excitation wave

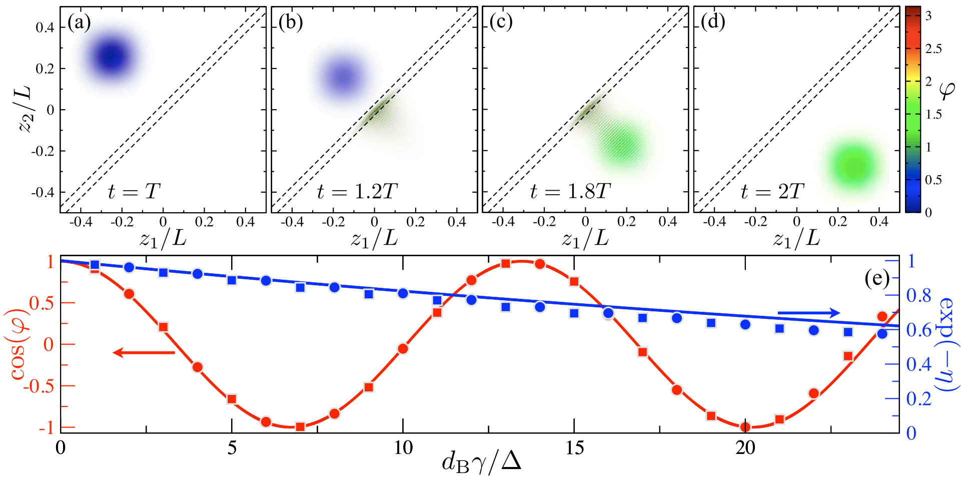

FIG. 2 (color online). (a)–(d) Two-photon counterpropagation for $\Delta = 20\gamma$, $\Omega = 2\Delta$, $g\sqrt{n} = 20\Delta$, and $z_B = 0.055\sigma$, where σ is the compressed pulse length inside the medium. The color coding shows the local phase of ee, while the opacity reflects the two-photon density $|ee|^2$. The dashed lines are $|z_1 - z_2| = z_B$. The full movie is provided in the Supplemental Material [35]. (e) Numerically obtained phase shift φ [we plot $\cos\varphi$] and attenuation $e^{-\eta}$ as a function of d_B compared to the analytical predictions (lines). The numerical data correspond to two different parameter scans $g\sqrt{n} = 400\Delta$, $z_B = 0.0025\sigma, \ldots, 0.03\sigma$ (dots) and $z_B = 0.03\sigma$, $g\sqrt{n} = 80, \ldots, 390$ (squares).

function on a length scale $\sim \frac{L}{d}(\gamma^2 + \Delta^2)/\gamma^2$, giving rise to an avoided volume between the remaining photons. This is confirmed by our numerical calculations, shown in Fig. 3 and in the movie in the Supplemental Material [35]. Therefore, the two-excitation wave function evolves into a statistical mixture of a single excitation and a correlated train of two photons separated by $z_{b(B)}$. We emphasize that the photon-density-independent avoided volume is a unique feature of our system. On resonance, if z_b is larger than the EIT-compressed pulse length σ, a single excitation will be generated deterministically. For a coherent input pulse, one similarly expects the wave packet to evolve with some probability into a correlated train of blockade-radius-separated photons. Furthermore, if $z_b > \sigma$, such a system can function as a deterministic single-photon source, in analogy with Refs. [38,39].

In summary, we have shown that Rydberg blockade in EIT media can be harnessed for inducing strong photon-photon interactions, with applications to generating non-classical states of light, implementing nonlinear photonic gates, and studying many-body phenomena with strongly correlated light. Besides providing a framework for describing experiments [10], this work opens several promising avenues of research. With an eye towards single-photon generation, one can extend the presented wave function treatment to a density matrix approach and explicitly analyze the propagation of the remaining excitation after the interaction-induced decay of multiphoton states. In addition, a gas of bosons (Rydberg polaritons) with a hard-sphere core (of radius $z_{b(B)}/2$) can be investigated both theoretically and experimentally in the copropagating case. In particular, the previously neglected

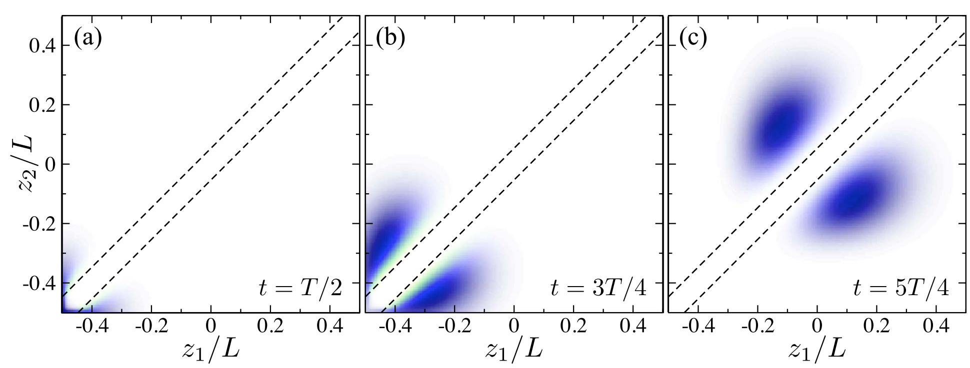

FIG. 3 (color online). Time evolution of $|ee|^2$ for two copropagating photons for $\Omega = \gamma$, $g\sqrt{n} = 100\gamma$, and $z_b = 0.08\sigma$. As in Fig. 2, the opacity shows the two-photon density $|ee|^2$, while the color indicates the local phase of ee. The dashed lines are $|z_1 - z_2| = z_b$, in agreement with the numerical results, which show the decay of ee within the dashed lines. The full movie is provided in the Supplemental Material [35].

effects of es_{-} endow these bosons with an effective mass $\propto -id\gamma/(Lv_g\Gamma)$, which plays a significant role for propagation distances larger than those considered in the present Letter. By including the effects of the coordinates transverse to the propagation axis, one can extend this problem to higher dimensions. Furthermore, for $\Delta/C_6 < 0$, the effective potential shows a resonant feature, which can give rise to two-polariton bound states.

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[35] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.107.133602 for the movies for counterpropagating (Fig. 2) and copropagating (Fig. 3) photons.
[36] EIT requires stray electric fields to be $\lesssim 30$ mV/cm to keep two-photon detuning below the width of the EIT transparency window $\Omega^2/(\gamma\sqrt{d_{\mathrm{B}}}) \approx (2\pi)0.5$ MHz.
[37] Motional effects are negligible: For a typical atomic coherence length $w = 40$ nm, a typical force $F \sim |V'(z_B)| \sim 10^{-24}$ N between two Rb atoms of mass m in state $|r\rangle$ a distance z_B apart gives a negligible dephasing rate $\sim\sqrt{F/(2wm)} \sim (2\pi)1$ kHz.
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