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<td>doi:10.1103/PhysRevLett.107.133602</td>
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Photon-Photon Interactions via Rydberg Blockade

Alexey V. Gorshkov,1 Johannes Otterbach,2 Michael Fleischhauer,2 Thomas Pohl,3 and Mikhail D. Lukin4

1Institute for Quantum Information, California Institute of Technology, Pasadena, California 91125, USA
2Department of Physics and Research Center OPTIMAS,
Technische Universität Kaiserslautern, 67663, Kaiserslautern, Germany
3Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany
4Physics Department, Harvard University, Cambridge, Massachusetts 02138, USA

(Dated: March 22, 2011)

We develop the theory of light propagation under the conditions of electromagnetically induced transparency (EIT) 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 demonstrate 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 quantum gates, as well as for studying many-body phenomena with strongly correlated photons.

PACS numbers: 42.50.Nn, 32.80.Ee, 42.50.Gy, 34.20.Cf

The phenomenon of electromagnetically induced transparency (EIT) [1] in systems involving Rydberg states [2] has recently attracted significant experimental [3,4] and theoretical [5,6] 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 [7] and for inducing strong photon-photon interactions, with applications to photonic quantum information processing [2,5] and to the realization of many-body phenomena with strongly interacting photons [8]. 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 give rise to photon-photon interactions, which, below a critical inter-photonic 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.

The basic physics is illustrated by considering a simple case [Fig. 1(b)], in which a single-photon wavepacket $E$ propagates in an EIT medium [level scheme in Fig. 1(a)] with a central control atom at $z = 0$ prepared in a Rydberg state $|r⟩$. Atoms in another Rydberg state $|r′⟩$, 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 $|r⟩$ and $|r′⟩$, resulting in $V \propto 1/z^3$ [2].

Sufficiently far away from $z = 0$, the incident photon propagates in a standard EIT medium featuring a two-photon-resonant control field with Rabi frequency $Ω$. In the vicinity of the control atom, however, the state $|r⟩$ is shifted so strongly out of resonance that the photon sees only a two-level $(|g⟩, |e⟩)$ medium with transition linewidth $2γ$. The critical $z$, at which the interaction is equal to the EIT linewidth, separates these two regimes and corresponds to the Rydberg blockade radius $[11,24]$. When the single-photon detuning $Δ = 0$, the resonant blockade radius $z_B$ is thus defined by $V(z_B) = Ω^2/γ$ ($h = 1$), while for $Δ \gg γ$, we define the off-resonant blockade radius $z_{B'}$ via $V(z_{B'}) = Ω^2/Δ$ (we assumed $Δ/C_6 > 0$). Since the blockade region extends over $2z_{B'}$ [Fig. 1(b)], the presence of the control atom locally creates an absorbing or refractive medium with an effective optical depth $d_{b(B)} = 2z_{b(B)}/L$, where $d$ is the resonant optical depth of the $|g⟩−|e⟩$ medium ($Ω = 0$) of length $L$. Interesting effects occur at large bloackaded optical depths $d_{b(B)}$. In the resonant case, assuming $d_{b} \gg 1$, the presence of the $|r⟩$ excitation causes complete scattering, i.e.
loss, of the incoming photon. Off resonance, for $d_B \gg 1$ and $d_B(\gamma/\Delta)^2 \ll 1$, the interaction with the $|r'\rangle$-atom imprint 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. [25] allows 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 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 single photon in a single atom is difficult, the same effect can be achieved by storing [26, 27] the photon in a collective $|r'\rangle$ excitation, as we will discuss below.

The results of this simple problem can be extended to the case of multi-photon EIT propagation in Rydberg media. First, off-resonance, two counter-propagating photons [Fig. 1(c)] can pick up a phase $\sim d_B\gamma/\Delta$, enabling the implementation of a two-photon phase gate [23, 24]. Second, a pulse of co-propagating photons [Fig. 1(d)] will evolve into a non-classical state corresponding to a train of single photons [19] and exhibiting correlations similar to those of hard-sphere particles with radius $z_B(\Omega)/2$. 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, $\sigma$, both co- and counter-propagating resonant set-ups might be 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 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 wavepacket and the polarization $S$ on the $|g\rangle - |r\rangle$ transition obey [26, 27]

\[
\left(\partial_t + c\partial_z\right)\mathcal{E}(z,t) = -\frac{e^2}{2m} \mathcal{E}(z,t) - \frac{2\sqrt{\Gamma \Omega}}{\Gamma} S(z,t),
\]

\[
\partial_t S(z,t) = -iU(z,t) - \frac{\Omega^2}{\Gamma} S(z,t) - \frac{2\sqrt{\Gamma \Omega}}{\Gamma} \mathcal{E}(z,t).
\]

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. (1,2) can be solved to give

\[
\mathcal{E}(\frac{L}{2},t) = \int_{-\infty}^{\infty} dw e^{i\omega(t-\frac{L}{c})+\frac{kL}{2} dz \chi(z,\omega)} \tilde{\mathcal{E}}(-\frac{L}{2},\omega)
\]

where

\[
k\chi(z,\omega) = \frac{1}{L} \frac{d\gamma}[\omega - V(z)] = \frac{1}{L} \frac{d\gamma}{\Omega^2 - (\Delta + i\gamma)|\omega - V(z)|}
\]

and $\tilde{\mathcal{E}}(-L/2,\omega)$ is the Fourier transform of the wavepacket incident at $z = -L/2$.

For narrowband pulses, we expand $\chi$ in $\omega$ and, assuming $\Delta \gg \gamma$ and $L \gg 2\Omega$, reduce Eq. (3) to

\[
\mathcal{E}(L/2,t) \approx \mathcal{E}(-L/2, t - L'/v_k)e^{i\varphi - \eta},
\]

where $v_k \approx c\Omega^2/(\gamma^2 n) = 2\Omega^2 L/(d^2\Gamma)$ is the EIT group velocity. In order to avoid the Raman resonance at $V + \Omega^2/\Delta = 0$, we assumed $\Delta/\Gamma < 0$. Since the photon travels at $c$ within the blockade region, the group delay comes from a reduced medium length $L' = L - \frac{2}{\pi} \pi 2B \approx L - 2\Omega$. Additionally, the emergence of a two-level medium within $|z| < z_B$ gives an intensity attenuation of $e^{-2n}$ with $2\eta = \frac{6\pi}{18} d_B (\gamma/\Delta)^2 \approx d_B (\gamma/\Delta)^2$ and a picked-up phase of $\varphi = -\Delta d_B (\gamma/\Delta) \approx -\frac{1}{2} d_B (\gamma/\Delta)$. 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. At the same time, after tracing out the scattered photon, which carries information about the location of the scattering, the spin wave itself disentangles into a classical mixture of pieces of length $\sim z_B$.

**Interaction of propagating photons.**—We now consider the problem of propagating photons interacting with each other. Regarding $\mathcal{E}$ and $S$ in Eqs. (12) as operators with same-time commutation relations $[\mathcal{E}(z),\mathcal{E}'(z')] = [S(z),S'(z')] = \delta(z - z')$ [27] and taking $U(z) = \int dz' V(z - z')S(z')S'(z)S(z)$, Eqs. (12) become Heisenberg operator equations [28] for the case of photons co-propagating in a Rydberg EIT medium [Fig. 1(d)]. Alternatively, for the case of two counter-propagating photons [Fig. 1(c)], we define operators $\hat{E}_{1(2)}$ and $S_{1(2)}$ for
the right- (left-) moving photon. For $S_1$, the interaction is $U(z) = \int dz' V(z-z')S_1(z)S_2(z')S_1(z)$, and vice versa for $S_2$.

Since the physics of two counter-propagating 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 wavefunction $|\psi(0)\rangle = |0\rangle_1|\mathcal{E}(z_1)\mathcal{E}(z_2)\rangle|\psi(t)\rangle$, $es(z_1, z_2, t) = \langle 0|\mathcal{E}(z_1)\mathcal{E}(z_2)|\psi(t)\rangle$, $se(z_1, z_2, t) = \langle 0|\mathcal{S}(z_1)\mathcal{S}(z_2)|\psi(t)\rangle$, and $ss(z_1, z_2, t) = \langle 0|\mathcal{S}(z_1)\mathcal{S}(z_2)|\psi(t)\rangle$. Eqs. (12) 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 $\partial_t \psi = M(r, \omega)\psi$, where $\psi = \{ee(R, r, \omega), es_+(R, r, \omega)\}$ and

$$M(r, \omega) = \begin{bmatrix} \frac{i\omega - \frac{g^2}{\sqrt{\Omega}}}{-g\sqrt{\Omega} + i\omega - \frac{g\sqrt{\Omega}}{\sqrt{\Omega} + g\sqrt{\Omega}}} \\
\frac{g^2}{\sqrt{\Omega} + i\omega - \frac{g\sqrt{\Omega}}{\sqrt{\Omega} - g\sqrt{\Omega}}} \end{bmatrix} \begin{bmatrix} g\sqrt{\Omega} \\
 g\sqrt{\Omega} \end{bmatrix}$$

$(6)$

$R$ enters only through boundary conditions and is, thus, not important in the present case. For narrowband pulses, we can expand $M(r, \omega) \approx M_0(r) + \omega M_1(r)$, with

$$M_0 = \frac{1}{\Gamma} \begin{bmatrix} g^2n & g\sqrt{\Omega} \\
 g\sqrt{\Omega} & \Omega + g^2nN \end{bmatrix}$$

$(7a)$

$$M_1 = i \begin{bmatrix} \frac{1}{\gamma} & 0 \\
 0 & \frac{1}{\gamma} \end{bmatrix} \begin{bmatrix} 0 \\
 0 \end{bmatrix} \begin{bmatrix} 0 \\
 0 \end{bmatrix}$$

$(7b)$

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 $|r| \gg z_B$, the two photons propagate as dark-state polaritons $|\mathcal{D}\rangle$, i.e. we have $es_+ = -g\sqrt{\Omega}/\Omega$, which is an eigenstate of $M_0$ with eigenvalue 0. Since $g\sqrt{\Omega} \gg \Omega$, the group velocity can be read out from the last entry of $M_1$, which gives twice the EIT group velocity $v_B$ since the two polaritons propagate towards each other. Within the blockade radius, where $\mathcal{V} \approx 1$ and $\mathcal{V}/\mathcal{V} \approx 0$, the polariton solution ceases to be an eigenstate of $M_0$, and Eq. (7) predicts a speed up to $\sim c$. Since the time $\sim z_B/c$ it takes to cross the blockade region is much less than the inverse width of the EIT window, the dynamics is highly non-adiabatic such that the main result of the interactions is a picked-up factor of $\exp[\int d\rho g^2n\mathcal{V}(\rho)/(\Gamma\Delta)] = \exp(i\varphi - \eta)$. This is a generalization of the result of Refs. [22, 24] 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 wavefunction decays provided it fits inside the medium and $d_B \gg 1$. The resulting state is a statistical mixture of right- and left-moving excitations.

Off resonance, $es_+$ picks up $\varphi \approx -\frac{\pi}{2\gamma/\Omega} \approx \frac{\pi}{2\gamma} d_B \approx \frac{\gamma}{2\Omega} d_B$ and $\eta \approx \frac{5\pi}{2\gamma/\Omega} \approx \frac{5\pi}{2\Omega} d_B$. Additionally, the off-diagonal terms in $M_0$ result in a small admixture of the bright state polariton $|\mathcal{D}\rangle$, which decays after the wavefunction exits the blockade region.

To verify these conclusions, we show in Fig. 2 and in the supplementary movie [30] 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 [30], 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 results, the obtained loss is slightly larger due to the brightness of the polariton admixture, which was neglected within the above approximate treatment.

Provided the EIT-compressed two-particle wavefunction 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 $\Omega/2\pi = 2\text{MHz}$ and $\Delta = 20\gamma$, we find $z_B = 15\mu$m, which, for a dense cloud with $n = 10^{12}$ cm$^{-3}$, gives $d_B = \frac{\Delta}{2\pi} \sqrt{2} |\mathcal{D}|/\Omega \approx 9$. 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 [31, 34] and working with a BEC [34].

In the co-propagating case, we define $ee(z_1, z_2, t) = \langle 0|\mathcal{E}(z_1)\mathcal{E}(z_2)|\psi(t)\rangle$, $es(z_1, z_2, t) = \langle 0|\mathcal{S}(z_1)\mathcal{S}(z_2)|\psi(t)\rangle$, and $ss(z_1, z_2, t) = \langle 0|\mathcal{S}(z_1)\mathcal{S}(z_2)|\psi(t)\rangle$. Defining $es_\pm(z_1, z_2) = \{es(z_1, z_2) + es(z_2, z_1)\}/2$, dropping $es_-$, and taking the Fourier transform in time, we obtain $\partial_t \mathbf{v} = 2M(r, \omega)\mathbf{v}$. That is, the only difference from the counter-propagating case is the replacement of $\partial_t$ 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, $M_0$ results in fast attenuation on a lengthscale $\sim \frac{\hbar}{2}(\gamma^2 + \Delta^2)/\gamma^2$. This is confirmed by our numerical calculations, shown in Fig. 3 and in the supplementary movie [30]. Therefore, the two-excitation wavefunction evolves into a statistical mixture of a single excitation and a correlated train of two photons separated by $z_{b(B)}$. On resonance, if the photon is scattered when the EIT-compressed pulse of length less than $z_b$ is fully inside the medium, we expect the remaining excitation to propagate in its original spatiotemporal mode. For a coherent input pulse, one similarly expects the wavepacket to evolve with some probability into a correlated train of blockade-radius-separated photons. Furthermore, if the blockade radius is larger than the EIT-compressed pulse length, there may be a regime in which such a system could function as a deterministic single-photon source.

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. This work opens several promising avenues of research. With an eye towards single-photon generation, one can extend the presented wavefunction treatment to a density matrix approach and explicitly analyze the propagation of the remaining excitation after the interaction-induced decay of multi-photon 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 co-propagating case. In particular, the previously neglected effects of $g_s$ endow these bosons with an effective mass $\propto -i\hbar \gamma/(L v_g \Gamma)$, 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.

We thank S. Hofferberth, M. Bajcsy, T. Peyronel, M. Hafezi, and N. Yao for discussions. This work was supported by NSF, the Lee A. DuBridge Fellowship, DFG through the GRK 792, CUA, DARPA, and the Packard Foundation.

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