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Phys. Rev. Lett. **107**, 133602 — Published 22 September 2011

DOI: [10.1103/PhysRevLett.107.133602](https://doi.org/10.1103/PhysRevLett.107.133602)

Photon-Photon Interactions via Rydberg Blockade

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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 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.

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 [3–10] 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 multi-photon problem. We show that Rydberg atom interactions induce photon-photon interactions, which, below a critical inter-photon 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 non-perturbative analysis reveals a possibility of photons behaving as “hard-sphere” objects with strong anti-correlations 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 wavepacket \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.

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 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 focussing 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

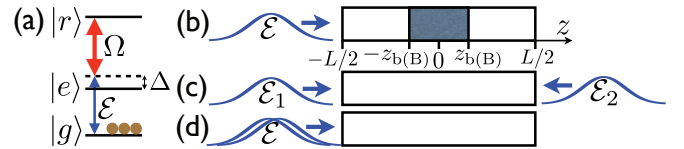


FIG. 1. (a) EIT level scheme, in which a ground state $|g\rangle$ (the initial state of each atom), an excited state $|e\rangle$, and a Rydberg state $|r\rangle$ are coupled by a quantum probe field \mathcal{E} and a classical control field with Rabi frequency Ω and single-photon detuning Δ . (b) Interaction 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 counter-propagating (c) or co-propagating (d) photons.

optical depth of the $|g\rangle\text{--}|e\rangle$ medium ($\Omega = 0$) of length L . Interesting effects occur at large blocked optical depths $d_{\text{b(B)}}$. On resonance, assuming $d_{\text{b}} \gg 1$, the $|r'\rangle$ atom causes complete scattering of the incoming photon. Off resonance, for $d_{\text{B}} \gg 1$ and $d_{\text{B}}(\gamma/\Delta)^2 \ll 1$, the $|r'\rangle$ atom imprints a phase $\sim d_{\text{B}}\gamma/\Delta$ on the probe photon and reduces its group delay by $\sim d_{\text{B}}\gamma/\Omega^2$, 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 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 multi-photon EIT propagation in Rydberg media. First, off-resonance, two counter-propagating photons [Fig. 1(c)] can pick up a phase $\sim d_{\text{B}}\gamma/\Delta$, enabling the implementation of a two-photon phase gate [12, 14]. 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 $z_{\text{b(B)}}/2$. 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 counter-propagating 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 with wavevector 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 $|g\rangle\text{--}|e\rangle$ transition, the slowly varying electric field amplitude \mathcal{E} of the single-photon wavepacket and the polarization S on the $|g\rangle\text{--}|r\rangle$ transition obey [24, 32]

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

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

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. (1,2) can be solved to give

$$\mathcal{E}\left(\frac{L}{2}, t\right) = \int_{-\infty}^{\infty} d\omega e^{-i\omega(t - \frac{L}{c}) + i\frac{k}{2} \int_{-\frac{L}{2}}^{\frac{L}{2}} dz \chi(z, \omega)} \tilde{\mathcal{E}}\left(-\frac{L}{2}, \omega\right) \quad (3)$$

where the susceptibility χ is

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

and $\tilde{\mathcal{E}}(-L/2, \omega)$ is the Fourier transform of the wavepacket 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 $z_{\text{b(B)}}$.

For narrowband pulses, we expand χ in ω and, assuming $\Delta \gg \gamma$ and $L \gg 2z_{\text{B}}$, reduce Eq. (3) to

$$\mathcal{E}(L/2, t) \approx \mathcal{E}(-L/2, t - L'/v_{\text{g}}) e^{i\varphi - \eta}, \quad (5)$$

where $v_{\text{g}} \approx c\Omega^2/(g^2 n) = 2\Omega^2 L/(d\gamma)$ 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 $L' = L - \frac{7}{9}\pi z_{\text{B}} \approx L - 2z_{\text{B}}$. Additionally, the emergence of a two-level medium within $|z| < z_{\text{B}}$ gives an intensity attenuation of $e^{-2\eta}$ with $2\eta = \frac{5\pi}{18}d_{\text{B}}(\gamma/\Delta)^2 \approx d_{\text{B}}(\gamma/\Delta)^2$ and a picked-up phase of $\varphi = -\frac{\pi}{6}d_{\text{B}}(\gamma/\Delta) \approx -\frac{1}{2}d_{\text{B}}(\gamma/\Delta)$. Thus, with $d_{\text{B}} \gg 1$ 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 $\approx \exp(-d_{\text{b}})$, as expected for a two-level medium of length $2z_{\text{b}}$.

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 $d_{\text{b}} \gg 1$, 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. (1,2) 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. (1,2) become Heisenberg operator equations [33] 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 $\mathcal{E}_{1(2)}$ 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 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 [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 $ss(z_1, z_2, t) = \langle 0 | S_1(z_1) S_2(z_2) | \psi(t) \rangle$. Eqs. (1,2) 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

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

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

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

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

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 $|r| \gg z_{b(B)}$, 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 $\sim z_{b(B)}/c$ it takes to cross the blockade region is much less than the inverse width of the EIT window, the dynamics is highly non-adiabatic (see below) such that the main result of the interactions is a pick-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 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 $\varphi \approx -\frac{\pi}{2^{1/6}6} \frac{\gamma}{\Delta} d_B \approx -\frac{\gamma}{2\Delta} d_B$ and $\eta \approx \frac{5\pi}{2^{1/6}36} \frac{\gamma^2}{\Delta^2} d_B \approx \frac{\gamma^2}{2\Delta^2} d_B$. Additionally, the off-diagonal terms in \mathbf{M}_0 result in a small admixture of the

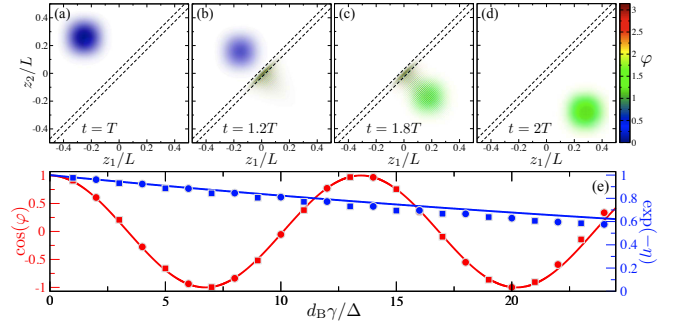


FIG. 2. (a)-(d) Two-photon counter-propagation for $\Delta = 20\gamma$, $\Omega = 2\Delta$, $g\sqrt{n} = 20\Delta$ and $z_B = 0.055\sigma$, where σ is the compressed pulse length inside the medium. The color coding shows the local phase of ee , while the opacity reflects the two-photon density $|ee|^2$. The dashed lines are $|z_1 - z_2| = z_B$. The full movie is provided in the supplementary 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 corresponds to two different parameter scans $g\sqrt{n} = 400\Delta$, $z_B = 0.0025\sigma, \dots, 0.03\sigma$ (dots) and $z_B = 0.03\sigma$, $g\sqrt{n} = 80, \dots, 390$ (squares).

bright-state polariton [24], which decays after the wavefunction exits the blockade region.

To verify these conclusions, we show in Fig. 2 and in the supplementary movie [35] the results of numerical solutions of the full equations for ee , es , se , and ss 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 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 $\Omega/2\pi = 2\text{MHz}$ [10] and $\Delta = 20\gamma$, we find $z_B = 15\mu\text{m}$, which, for a dense cloud with $n = 10^{12} \text{ cm}^{-3}$, gives $d_B = \frac{3}{2\pi} \lambda^2 (2z_B)n \approx 9$ [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 BEC [30].

In the co-propagating case, we define $ee(z_1, z_2, t) = \langle 0 | \mathcal{E}(z_1) \mathcal{E}(z_2) | \psi(t) \rangle$, $es(z_1, z_2, t) = \langle 0 | \mathcal{E}(z_1) S(z_2) | \psi(t) \rangle$, 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 counter-propagating 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

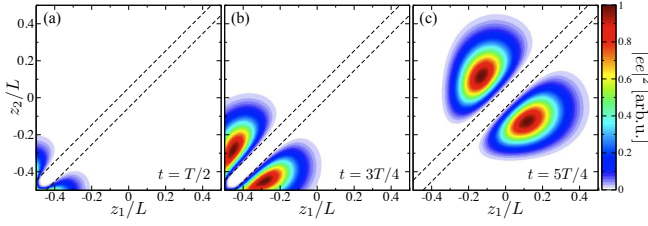


FIG. 3. Time evolution of $|ee|^2$ for two co-propagating photons for $\Omega = \gamma$, $g\sqrt{n} = 100\gamma$, and $z_b = 0.08\sigma$. The dashed lines are $|z_1 - z_2| = z_b$, in agreement with the numerical results, which show the decay of ee within the dashed lines. The full movie is provided in the supplementary material [35].

radius, the two-photon dark-state polariton propagates with group velocity v_g . Inside the blockade radius, M_0 results in exponential attenuation of the two-excitation wavefunction on a lengthscale $\sim \frac{L}{d}(\gamma^2 + \Delta^2)/\gamma^2$, giving rise to an avoided volume between the remaining photons. This is confirmed by our numerical calculations, shown in Fig. 3 and in the supplementary movie [35]. Therefore, the two-excitation wavefunction evolves into a statistical mixture of a single excitation and a correlated train of two photons separated by $z_{b(B)}$. 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 wavepacket 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 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 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 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.

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 and Packard Foundations, DFG through the GRK 792, CUA, and DARPA.

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 - [35] The movies for counter-propagating [Fig. 2] and co-propagating [Fig. 3] photons are provided in the supplementary material.
 - [36] EIT requires stray electric fields to be $\lesssim 30$ mV/cm to keep two-photon detuning below $\Omega^2/(\gamma\sqrt{d_B}) \approx 0.5$ MHz.

[37] For a 40 nm atomic coherence length, a force $|V'(z_B)| \sim 10^{-24}$ N gives a negligible dephasing of 1 kHz.