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Electromagnetically induced transparency and photon-photon interactions with Rydberg atoms

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Abstract. We discuss electromagnetically induced transparency in a cold ensemble of atoms optically excited to the Rydberg states. Strong dipole-dipole or van der Waals interactions between the atomic Rydberg states translate into large nonlocal nonlinearities for a propagating probe field. In the case of weak quantum fields, this can be used to attain a conditional phase shift of π between pairs of single photons realizing a deterministic photonic phase gate. For stronger fields, long-range interactions between the atoms constrain the medium to behave as a collection of superatoms, each comprising a blockade volume that can accommodate at most one Rydberg excitation. The propagation of a probe field is affected by its two-photon correlations within the blockade distance, which are strongly damped due to low saturation threshold of the superatoms. Our calculations reproduce and interpret the results of recent experiments.

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1. Introduction

Strong dipole–dipole (DD) or van der Waals (VdW) interactions between highly excited Rydberg states of atoms [1] constitute the basis for a number of promising schemes for quantum information applications [2] and interesting many-body effects [3, 4, 5, 6, 7]. Some of these schemes employ the dipole blockade mechanism [8] which suppresses multiple Rydberg excitations within a certain interaction (blockade) volume. Employing electromagnetically induced transparency (EIT) [9, 10] in a ladder configuration, one can translate the DD or VdW interactions between optically excited Rydberg states of atoms into sizable interactions between the photons [11, 12, 13]. Several experiments on EIT with Rydberg atoms were recently performed [14, 15, 16, 17].

Due to the strong, long-range interactions of the Rydberg atoms, the theoretical description of EIT in Rydberg media is a highly non-trivial many-body problem [11, 13, 17, 18, 19]. Here we outline our recent work on this subject, which elucidated two regimes of field intensities: interactions of weak, single-photon pulses [11], which can be used to implement universal photonic logic gates [20, 21]; and propagation and attenuation of strong input fields studied in a recent prominent experiment [17], whose theoretical treatment reveals the importance of two-photon correlations [19].

2. Interaction of weak quantum fields via Rydberg atoms

EIT can translate strong DD interactions between the Rydberg states of atoms into sizable interactions between single photons [11], as described below.

2.1. Equations of motion

Consider an ensemble of cold alkali atoms with level configuration as in Fig. 1. All the atoms are initially prepared in the ground state $|g\rangle$. Two distinguishable, weak (quantum) fields $E_{1,2}$ propagate in the opposite directions along the z axis and resonantly interact with the atoms on the transitions $|g\rangle \to |e_{1,2}\rangle$, respectively. The intermediate states $|e_{1,2}\rangle$ are resonantly coupled by two strong (classical) control fields

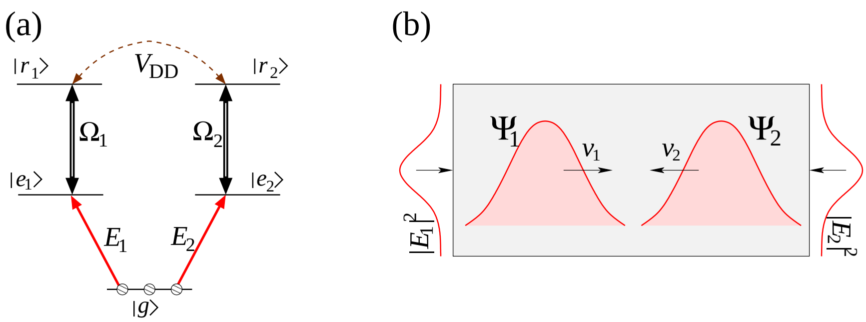

Figure 1. (a) Level scheme of atoms interacting with weak (quantum) fields $E_{1,2}$ on the transitions $|g\rangle \to |e_{1,2}\rangle$ and strong control fields of Rabi frequencies $\Omega_{1,2}$ on the transitions $|e_{1,2}\rangle \to |r_{1,2}\rangle$, respectively. V_{DD} denotes the DD interaction between pairs of atoms in Rydberg states $|r\rangle$. (b) Upon entering the medium, each field is converted into the corresponding polariton $\Psi_{1,2}$ representing a coupled excitation of the field and atomic coherence. These polaritons propagate in the opposite directions with slow group velocities $v_{1,2}$ and interact via the DD interaction.

with Rabi frequencies $\Omega_{1,2}$ to the highly excited Rydberg states $|r_{1,2}\rangle$. In a static electric field $E_{\rm st}\mathbf{e}_z$, the Rydberg states $|r\rangle$ possess large permanent dipole moments $\mathbf{p} = \frac{3}{2}nqea_0\mathbf{e}_z$, where n and $q \equiv n_1 - n_2$ are, respectively, the (effective) principal and parabolic quantum numbers, e is the electron charge, and a_0 is the Bohr radius [1]. A pair of atoms i and j at positions $\mathbf{r}_i$ and $\mathbf{r}_j$ excited to states $|r\rangle$ interact with each other via the DD potential $\hbar\Delta(\mathbf{r}_i - \mathbf{r}_j) = \hbar C_3(1 - 3\cos^2\vartheta)|\mathbf{r}_i - \mathbf{r}_j|^{-3}$, where ϑ is the angle between vectors $\mathbf{e}_z$ and $\mathbf{r}_i - \mathbf{r}_j$, and $C_3 \propto \wp_r^2$ is a constant proportional to the product of atomic dipole moments $\wp_r = \langle r_l|\,\mathbf{p}\,|r_l\rangle \propto n^2$ assumed the same for both states $|r_{1,2}\rangle$. In the frame rotating with the frequencies of the optical fields, the interaction Hamiltonian $H = V_{\rm af} + V_{\rm DD}$ contains the atom-field interaction $V_{\rm af} = -\hbar\sum_{l=1,2}\sum_j^N [g_l^j\hat{\mathcal{E}}_l\hat{\sigma}^j_{e_l g} + \Omega_l\hat{\sigma}^j_{r_l e_l} + \text{H.c.}]$, and the DD interaction $V_{\rm DD} = \hbar\sum_{i<j}^N \hat{\sigma}^i_{rr}\Delta(\mathbf{r}_i - \mathbf{r}_j)\hat{\sigma}^j_{rr}$, where $N = \rho V$ is the total number of atoms of density ρ in the (quantization) volume V, $\hat{\sigma}^j_{\mu\nu} \equiv |\mu\rangle_{jj}\langle\nu|$ is the transition operator of the jth atom, $\hat{\mathcal{E}}_l$ is the slowly-varying operator corresponding to the electric field E_l ($l = 1, 2$), and g_l^j is the corresponding atom-field coupling constant on the transition $|g\rangle_j \to |e_l\rangle_j$.

We assume that the transverse profile of both weak fields E_l is described by a Gaussian $e^{-r_\perp^2/w^2}$ of width w, where $r_\perp = |\mathbf{r}_\perp|$ is the distance from the field propagation z-axis. Then the (transverse-averaged) coupling constants are given by $g_l = (\wp_{ge_l}/\hbar)\sqrt{\hbar\omega/2\epsilon_0 V}$, $\wp_{ge_l}$ being the dipole matrix element on the transition $|g\rangle \to |e_l\rangle$, $V = \pi w^2 L$, and L the medium length. The Rabi frequencies of strong control fields Ω_l are assumed uniform over the entire volume V. Using Hamiltonian H, we derive the Heisenberg-Langevin equations for the atomic operators $\hat{\sigma}_{ge_l}(\mathbf{r})$, $\hat{\sigma}_{gr_l}(\mathbf{r})$ and propagation equations for the quantum fields $\hat{\mathcal{E}}_l(z)$. Solving for the atomic operators perturbatively in the small parameters $g_l\hat{\mathcal{E}}_l/\Omega_l$ and in the adiabatic approximation [9, 10, 11], and after substituting into the equations for the fields, we obtain the following propagation equations for the dark-state polaritons $\hat{\Psi}_l = \sqrt{c/v_l}\,\hat{\mathcal{E}}_l$ [9],

$$(\partial_t \pm v_l\partial_z)\,\hat{\Psi}_l(z,t) = -i\sin^2\theta_l\hat{S}(z,t)\hat{\Psi}_l(z,t), \tag{1}$$

the sign "+" or "−" corresponding to $l = 1$ or 2, respectively, $v_l = c\cos^2\theta_l$ is the group velocity of the corresponding field in the EIT medium, and the mixing angles θ_l are defined through $\tan^2\theta_l = g_l^2N/|\Omega_l|^2$. Operator $\hat{S}(z,t)$ is responsible for the self- and cross-phase modulation between the fields,

$$\hat{S}(z,t) = \frac{1}{L}\int_0^L dz'\Delta(z-z')\big[\sin^2\theta_1\hat{\mathcal{I}}_1(z',t) + \sin^2\theta_2\hat{\mathcal{I}}_2(z',t)\big], \tag{2}$$

where $\hat{\mathcal{I}}_l \equiv \hat{\Psi}_l^\dagger\hat{\Psi}_l = (c/v_l)\hat{\mathcal{E}}_l^\dagger\hat{\mathcal{E}}_l$ are the polariton intensity (excitation number) operators in the EIT medium, which correspond to the photon number operators outside the medium ($v_l = c$) [9], while the effective one-dimensional DD potential $\Delta(z-z')$ result from $\Delta(\mathbf{r}-\mathbf{r}')$ upon double integration over the transverse coordinate,

$$\begin{aligned}\Delta(z-z') &= \frac{1}{(\pi w^2)^2}\int d^2r_\perp\int d^2r'_\perp e^{-(r_\perp^2+r_\perp'^2)/w^2}\Delta(\mathbf{r}-\mathbf{r}') \\ &= \frac{C_3}{\sqrt{2}w^3}\big[2|\zeta| - \sqrt{\pi}(1+2\zeta^2)e^{\zeta^2}\text{erfc}(|\zeta|)\big], \qquad \zeta \equiv (z-z')/\sqrt{2}w. \end{aligned} \tag{3}$$

As seen in Fig. 2(a), $\Delta(\zeta)$ is sharply peaked around $\zeta = 0$.

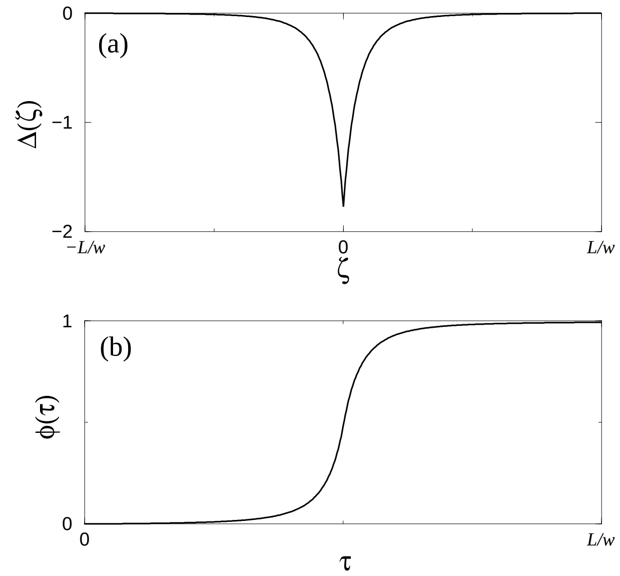

Figure 2. (a) One-dimensional DD potential $\Delta(\zeta)$ of Eq. (3) as a function of dimensionless distance ζ, in units of $C_3/\sqrt{2}w^3$ Hz. (b) The resulting phase-shift $\phi(\tau) \equiv \phi(vt, L - vt, t)$ of Eq. (6) as a function of dimensionless time $\tau = vt/w$, in units of $C_3/(vw^2)$ rad.

It follows from Eq. (1) that the intensity operators $\hat{\mathcal{I}}_l$ are constants of motion: $\hat{\mathcal{I}}_l(z,t) = \hat{\mathcal{I}}_l(z \mp v_l t, 0)$, the upper (lower) sign corresponding to $l = 1$ ($l = 2$). The solution for the field operators then reads

$$\hat{\Psi}_l(z,t) = \exp\left[- i \sin^2\theta_l \int_0^t dt' \hat{S}(z \mp v_l(t-t'), t') \right] \hat{\Psi}_l(z \mp v_l t, 0). \quad (4)$$

The validity of this dissipation-free solution hinges on the following assumptions: (i) The duration T of the pulses exceeds the inverse of the corresponding EIT bandwidth $\delta\omega = |\Omega_l|^2/(\gamma_{e_l}\sqrt{\kappa_l L})$, where γ_{e_l} is the transversal relaxation rate and $\kappa_l \simeq 3\lambda_l^2/(2\pi)\rho$ is the resonant absorption coefficient on the transition $|g\rangle \to |e_l\rangle$. With $v_l = 2|\Omega_l|^2/(\kappa_l \gamma_{e_l})$, this yields the condition $(\kappa_l L)^{-1/2} \ll T v_l / L < 1$ which requires a medium with large optical depth $\kappa_l L \gg 1$ [10]. (ii) The DD induced frequency shifts lie within the EIT bandwidths, $\langle \hat{S}(z) \rangle < \delta\omega$, $\forall\, z \in [0, L]$. (iii) The propagation/interaction time of each pulse $t_{\text{out}} = L/v_l$ is limited by the relaxation rate γ_{r_l} of the Rydberg state coherence via $t_{\text{out}}\gamma_{r_l} \ll 1$.

For simplicity of notation, we set $\theta_{1,2} = \theta$, i.e., $g_1^2 N/|\Omega_1|^2 = g_2^2 N/|\Omega_2|^2$.

2.2. Photonic phase gate

We now employ Eq. (4) to demonstrate the quantum phase gate between two single-photon pulses. We are concerned with the evolution of input state $|\Phi_{\text{in}}\rangle = |1_1\rangle\,|1_2\rangle$ composed of two single-excitation wavepackets $|1_l\rangle = \left[\frac{1}{L}\int dz f_l(z) \hat{\Psi}_l^\dagger(z)\right]|0\rangle$ whose spatial envelopes inside the medium $f_l(z) = \langle 0|\,\hat{\Psi}_l(z,0)\,|1_l\rangle$ are normalized as $\frac{1}{L}\int dz |f_l(z)|^2 = 1$. With the operator solution (4), for the (equal-time) correlation amplitude or the "two-photon wavefunction" $F_{12}(z_1, z_2, t) = \langle 0|\,\hat{\Psi}_1(z_1,t)\hat{\Psi}_2(z_2,t)\,|\Phi_{\text{in}}\rangle$ [21] we obtain

$$F_{12}(z_1, z_2, t) = f_1(z_1 - vt) f_2(z_2 + vt) \exp[i\phi(z_1, z_2, t)], \quad (5)$$

$$\phi(z_1, z_2, t) = -\sin^4\theta \int_0^t dt' \Delta\big(z_1 - z_2 - 2v(t - t')\big). \tag{6}$$

Hence, the two polaritons counterpropagate in a shape-preserving manner with group velocities $\pm v$. Since $\hat{\mathcal{I}}_l \hat{\Psi}_l |1_l\rangle = 0$, the self-interaction within each pulse is absent, while the cross-interaction between the pulses results in the phase-shift (6) shown in Fig. 2(b). Assume that at $t = 0$ the first pulse is centered at $z_1 = 0$ and the second pulse at $z_2 = L$, while after the interaction, $t_{\rm out} = L/v$, the coordinates of the two pulses are $z_1 = L$ and $z_2 = 0$, respectively. The accumulated phase-shift is then $\phi(L, 0, L/v) = -\sin^4\theta/v \int_0^L dz' \Delta_{12}(2z' - L)$. To evaluate the integral, we replace the variable $(2z' - L)/\sqrt{2}w \to \zeta'$ and extend the integration limits to $L/\sqrt{2}w \to \infty$, obtaining a *spatially uniform* $\phi = C_3/(vw^2)$ $(\sin\theta \simeq 1)$. The state of the system at $t_{\rm out}$ is then $|\Phi_{\rm out}\rangle = e^{\phi}|\Phi_{\rm in}\rangle$. Since for input states $|m_1\rangle|n_2\rangle$ $(m, n = 0, 1)$ there is no phase shift when $m + n < 2$, the conditional two-photon phase shift $\phi = \pi$ is equivalent to the CPHASE gate $|\Phi_{\rm out}\rangle = (-1)^{mn}|m_1\rangle|n_2\rangle$ [20].

2.3. Experimental considerations

To relate the foregoing discussion to a realistic experiment, we assume that the quantum fields are confined in a hollow-core waveguide of length $L \sim 1$ cm with the lowest transverse mode of width $w \simeq 2\mu$m [22]. The waveguide is filled with $N \simeq 5 \times 10^4$ cold Rb atoms at density $\rho \simeq 2 \times 10^{11}$ cm^{-3}. For the two quantum fields tuned to the D1 and D2 transitions $|g\rangle \to |e_{1,2}\rangle$ $(\lambda_1 = 795$ nm, $\lambda_2 = 780$ nm), the corresponding optical depths are $\kappa_1 L \simeq 600$ and $\kappa_2 L \simeq 580$. With $\gamma_{e_1} \simeq 1.8 \times 10^7\,{\rm s}^{-1}$, $\gamma_{e_2} \simeq 1.9 \times 10^7\,{\rm s}^{-1}$, and taking $\Omega_1 \simeq 7.35 \times 10^6$ rad/s, $\Omega_2 \simeq 7.43 \times 10^6$ rad/s, the group velocities are $v_{1,2} = 100$ m/s. The bandwidth of the pulses $T^{-1} \gtrsim v/L = 10^4\,{\rm s}^{-1}$ is smaller than the EIT bandwidth $\delta\omega \simeq 1.2 \times 10^5$ rad/s. To realize the CPHASE gate, we choose the Rydberg states $|r_{1,2}\rangle$ with $\wp_{r_1} = \wp_{r_2} = 315 e a_0$ (quantum numbers $n = 15$ and $q = n - 1$), leading to the conditional phase shift $\phi = \pi$. We have verified that the DD frequency shift is within the EIT window $\delta\omega$.

3. Strong field EIT with Rydberg atoms

We now consider propagation of a single (multiphoton) probe field $\hat{\mathcal{E}}_p$ of variable intensity through a ladder EIT medium with the VdW interactions between the atomic Rydberg states, as shown in Fig. 3(a). This system was studied experimentally in Ref. [17], where increasing the probe field amplitude led to reduction of its transmission within the EIT window, which, quite surprisingly, was accompanied by negligible broadening and indiscernible shift of the EIT line. We have developed an efficient theoretical model for EIT with Rydberg atoms [19], whose predictions fully reproduce the experimental observations [17], as detailed below.

3.1. Equations of motion

In the frame rotating with the frequencies of the probe ω_p and control ω_c fields, the system Hamiltonian $H = H_{\rm a} + V_{\rm af} + V_{\rm VdW}$ now contains the unperturbed atomic part, $H_{\rm a} = -\hbar \sum_j^N [\Delta_p \hat{\sigma}_{ee}^j + (\Delta_p + \delta_c)\hat{\sigma}_{rr}^j]$, where $\Delta_p = \omega_p - \omega_{eg}$ and $\delta_c = \omega_c - \omega_{re}$ are the corresponding detunings; and the atom-field and VdW interactions, $V_{\rm af} = -\hbar \sum_j^N [\hat{\Omega}_p(\mathbf{r}_j)\hat{\sigma}_{eg}^j + \Omega_c \hat{\sigma}_{re}^j + {\rm H.c.}]$ and $V_{\rm VdW} = \hbar \sum_{i<j}^N \hat{\sigma}_{rr}^i \Delta(\mathbf{r}_i - \mathbf{r}_j)\hat{\sigma}_{rr}^j$, where $\hat{\Omega}_p = g\hat{\mathcal{E}}_p$ is the probe Rabi frequency, while $\hbar\Delta(\mathbf{r}_i - \mathbf{r}_j) = \hbar C_6 |\mathbf{r}_i - \mathbf{r}_j|^{-6}$ is the VdW

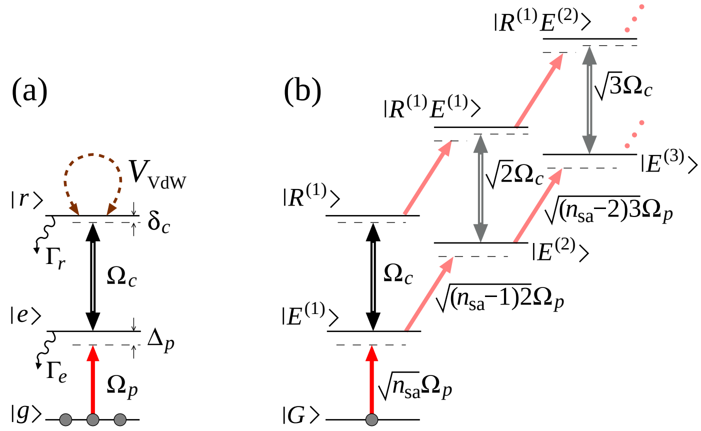

Figure 3. (a) Level scheme of atoms interacting with the probe Ω_p and control Ω_c fields on the corresponding transitions with detunings Δ_p and δ_c, respectively. Γ_e and Γ_r are the (population) decay rates of states $|e\rangle$ and $|r\rangle$, and $V_{\rm VdW}$ denotes the VdW interaction between the atoms in the Rydberg state $|r\rangle$. (b) Truncated level scheme of a superatom, composed of $n_{\rm sa}$ atoms, with the corresponding transition amplitudes due the probe and control fields.

potential between a pair of atoms i and j at positions $\mathbf{r}_i$ and $\mathbf{r}_j$. We consider stationary propagation of the probe field along the z axis, and assume uniform, undepleted control field Ω_c. Using Hamiltonian H, we derive Heisenberg-Langevin equations for the field $\hat{\mathcal{E}}_p(\mathbf{r})$ and atomic $\hat{\sigma}_{\mu\nu}(\mathbf{r})$ operators. For moderate Rabi frequency $\Omega_p < \gamma_e$ and number density of probe photons $\rho_{\rm phot} \ll \rho$, we can assume linear response of individual atoms to the applied field and arrive at the propagation equation for the probe field amplitude, $\partial_z \hat{\mathcal{E}}_p = i\frac{\kappa}{2}\hat{\alpha}\hat{\mathcal{E}}_p$ with the polarizability

$$\hat{\alpha}(\mathbf{r}) = \frac{i\gamma_e}{\gamma_e - i\Delta_p + |\Omega_c|^2[\gamma_r - i(\Delta_2 - \hat{S}(\mathbf{r}))]^{-1}}, \tag{7}$$

where $\hat{S}(\mathbf{r}) \equiv \int d^3r' \rho(\mathbf{r}')\,\Delta(\mathbf{r}-\mathbf{r}')\hat{\sigma}_{rr}(\mathbf{r}')$ is the total VdW induced shift of level $|r\rangle$ for an atom at position $\mathbf{r}$. Since $\hat{S}(\mathbf{r})$ involves integration over all spatial coordinates $\mathbf{r}' \in V$, Eqs. (7) is highly nonlocal. We therefore need to contrive an efficient method to evaluate the VdW shift $\hat{S}(\mathbf{r})$.

The strong, long-range interactions between the Rydberg atoms suppresses multiple Rydberg excitations within a blockade volume $V_{\rm sa} = \frac{4\pi}{3}R_{\rm sa}^3$ where $R_{\rm sa} \simeq \sqrt[6]{C_6\gamma_e/|\Omega_c|^2}$ is the blockade radius [8]. We therefore call the ensemble of $n_{\rm sa} = \rho V_{\rm sa}$ atoms "superatom" (SA) [19]. Each SA can contain only one Rydberg excitation delocalized over $V_{\rm sa}$. The level scheme of SA is shown in Fig. 1(b): $|G\rangle = |g_1, g_2, \ldots, g_{n_{\rm sa}}\rangle$ is the ground state, and $|R^{(1)}\rangle = \frac{1}{\sqrt{n_{\rm sa}}}\sum_j^{n_{\rm sa}} |g_1, g_2, \ldots, r_j, \ldots, g_{n_{\rm sa}}\rangle$ is the single collective Rydberg excitation state, while $|E^{(k)}\rangle$ are the properly symmetrized (Dicke) states with k atoms in $|e\rangle$. The corresponding transition amplitudes follow from $\langle E^{(1)}|\, V_{\rm af}\, |G\rangle = \sqrt{n_{\rm sa}}\hat{\Omega}_p$, $\langle R^{(1)}|\, V_{\rm af}\, |E^{(1)}\rangle = \Omega_c$, etc. We then obtain for the SA operators $\hat{\Sigma}_{GR} \equiv |G\rangle\langle R^{(1)}| = \Omega_c\sqrt{n_{\rm sa}}\hat{\Omega}_p\hat{\Sigma}_{GG}/[(\Delta_p + i\gamma_e)\Delta_2 - |\Omega_c|^2]$ and $\hat{\Sigma}_{RR} = \hat{\Sigma}_{RG}\hat{\Sigma}_{GR}$. To account for possible saturation of transition $|G\rangle \to |R^{(1)}\rangle$, we take $\hat{\Sigma}_{GG} + \hat{\Sigma}_{RR} = 1$, which finally yields

$$\hat{\Sigma}_{RR} = \frac{|\Omega_c|^2 n_{\rm sa}\hat{\Omega}_p^\dagger\hat{\Omega}_p}{|\Omega_c|^2 n_{\rm sa}\hat{\Omega}_p^\dagger\hat{\Omega}_p + [|\Omega_c|^2 - \Delta_p\Delta_2]^2 + \Delta_2^2\gamma_e^2}. \tag{8}$$

We can now treat the medium as a collection of $N_{\rm sa} = \rho_{\rm sa} V$ SAs at positions $\mathbf{r}_j$, which implies a spatial coarse-graining with the grain size $2R_{\rm sa}$ [23, 24]. The total VdW shift $\hat{S}(\mathbf{r})$ at position $\mathbf{r}$ can then be expressed as

$$\hat{S}(\mathbf{r}) \approx \sum_{j}^{N_{\rm sa}} \Delta(\mathbf{r}-\mathbf{r}_j)\hat{\Sigma}_{RR}(\mathbf{r}_j) = \bar{\Delta}\hat{\Sigma}_{RR}(\mathbf{r}) + \hat{s}(\mathbf{r}). \tag{9}$$

The physical meaning of the first term on the rhs of Eq. (9) is that an excited SA at $\mathbf{r}_j \simeq \mathbf{r}$ $[\hat{\Sigma}_{RR}(\mathbf{r}) \to 1]$ induces divergent VdW shift averaged over the SA volume: $\bar{\Delta} \simeq \frac{1}{V_{\rm sa}} \int_{V_{\rm sa}} \Delta(\mathbf{r}')d^3r' \to \infty$. The last term $\hat{s}(\mathbf{r}) \equiv \sum_{j\neq j_{\mathbf{r}}}^{N_{\rm sa}} \Delta(\mathbf{r}-\mathbf{r}_j)\hat{\Sigma}_{RR}(\mathbf{r}_j)$ describes the VdW shift induced by the external SAs outside the volume $V_{\rm sa}^{(\mathbf{r})}$ centered at $\mathbf{r}$. It can be evaluated by replacing the summation by an integration over the entire volume V, excluding the SA at $\mathbf{r}$, which, upon using the mean-field approximation, yields a small shift $\langle \hat{s}(\mathbf{r})\rangle = \frac{w}{8}\langle \hat{\Sigma}_{RR}(\mathbf{r})\rangle$.

Using $\hat{\Sigma}_{RR}(\mathbf{r})$ as a projector onto the Rydberg excitation of SA at $\mathbf{r}$ [while $\bar{\Delta} \gg \gamma_e$], we now recast the polarizability of Eq. (7) as

$$\hat{\alpha}(\mathbf{r}) = \hat{\Sigma}_{RR}(\mathbf{r})\frac{i\gamma_e}{\gamma_e - i\Delta_p} + [1-\hat{\Sigma}_{RR}(\mathbf{r})]\frac{i\gamma_e}{\gamma_e - i\Delta_p + |\Omega_c|^2[\gamma_r - i(\Delta_2 - \langle \hat{s}(\mathbf{r})\rangle)]^{-1}}. \tag{10}$$

Here the first fraction is the polarizability $\alpha_{\rm TLA}$ of a two-level atom, which applies when the SA at position $\mathbf{r}$ contains a Rydberg excitation $[\hat{\Sigma}_{RR}(\mathbf{r}) \to 1]$, while the second fraction, barring the small mean-field shift $\langle \hat{s}(\mathbf{r})\rangle$, is the usual EIT polarizability $\alpha_{\rm EIT}$ [10] acting when no Rydberg excitation is present $[\hat{\Sigma}_{RR}(\mathbf{r}) \to 0]$.

The expectation value of the probe field intensity obeys the equation

$$\partial_z \langle \hat{\mathcal{E}}_p^\dagger(\mathbf{r})\hat{\mathcal{E}}_p(\mathbf{r})\rangle = -\kappa \langle \hat{\mathcal{E}}_p^\dagger(\mathbf{r}){\rm Im}[\hat{\alpha}(\mathbf{r})]\hat{\mathcal{E}}_p(\mathbf{r})\rangle. \tag{11}$$

Note that factorizing out ${\rm Im}[\langle\hat{\alpha}(\mathbf{r})\rangle]$ in a mean-field sense would amount to neglecting the essential two-particle quantum correlations [13] originating from nonlinear response of the atoms to the Rydberg excitations. We therefore replace $\hat{\alpha}(\mathbf{r})$ in Eq. (11) by its expectation value *conditioned* upon the presence of photon at $\mathbf{r}$, denoted by $\langle\cdot\rangle_{\mathbf{r}}$,

$$\langle\hat{\alpha}(\mathbf{r})\rangle_{\mathbf{r}} = \langle\hat{\Sigma}_{RR}(\mathbf{r})\rangle_{\mathbf{r}}\alpha_{\rm TLA} + [1-\langle\hat{\Sigma}_{RR}(\mathbf{r})\rangle_{\mathbf{r}}]\alpha_{\rm EIT}. \tag{12}$$

The conditional Rydberg population $\langle\hat{\Sigma}_{RR}\rangle_{\mathbf{r}}$ of the SA at $\mathbf{r}$ is obtained from Eq. (8) by the replacement $\hat{\Omega}_p^\dagger(\mathbf{r})\hat{\Omega}_p(\mathbf{r}) \to \langle\hat{\Omega}_p^\dagger(\mathbf{r})\hat{\Omega}_p(\mathbf{r})\rangle\, g_p^{(2)}(\mathbf{r})$, where the probe field intensity correlation function $g_p^{(2)}(\mathbf{r}) = \frac{\langle\hat{\mathcal{E}}_p^\dagger(\mathbf{r})\hat{\mathcal{E}}_p^\dagger(\mathbf{r})\hat{\mathcal{E}}_p(\mathbf{r})\hat{\mathcal{E}}_p(\mathbf{r})\rangle}{\langle\hat{\mathcal{E}}_p^\dagger(\mathbf{r})\hat{\mathcal{E}}_p(\mathbf{r})\rangle\langle\hat{\mathcal{E}}_p^\dagger(\mathbf{r})\hat{\mathcal{E}}_p(\mathbf{r})\rangle}$ quantifies the probability of having simultaneously at least two photons in the blockade volume $V_{\rm sa}^{(\mathbf{r})}$. Note that linear, e.g. bare EIT, response of the medium does not change the correlation function of the propagating field, and only nonlinear, i.e. conditional, absorption $\propto {\rm Im}[\langle\hat{\alpha}(\mathbf{r})\rangle - \alpha_{\rm EIT}]$ modifies $g_p^{(2)}$, which therefore obeys the equation of motion

$$\partial_z g_p^{(2)}(\mathbf{r}) = -\kappa\langle\hat{\Sigma}_{RR}(\mathbf{r})\rangle{\rm Im}[\alpha_{\rm TLA} - \alpha_{\rm EIT}]g_p^{(2)}(\mathbf{r}). \tag{13}$$

We note that our treatment involves only single transverse mode of the probe field, which is effectively defined by the SA cross-section.

Given the input field "intensity" $I_p \equiv \langle\hat{\Omega}_p^\dagger\hat{\Omega}_p\rangle$ and its correlation function $g_p^{(2)}$ [for "classical" coherent field $g_p^{(2)}(0) = 1$], we then use the following stochastic procedure to integrate the coupled Eqs. (11)-(13) for $z \in [0, L]$: We divide the propagation distance L into $L/(2R_{\rm sa})$ intervals corresponding to SAs, and for z within each SA we determine via Monte-Carlo sampling of $\langle\hat{\Sigma}_{RR}(\mathbf{r})\rangle_{\mathbf{r}}$ whether the SA is excited, $\hat{\Sigma}_{RR}(\mathbf{r}) \to 1$, or not, $\hat{\Sigma}_{RR}(\mathbf{r}) \to 0$. We then average over several independent realizations. The limit of infinitely many such realizations corresponds to continuous polarizability of Eq. (12).

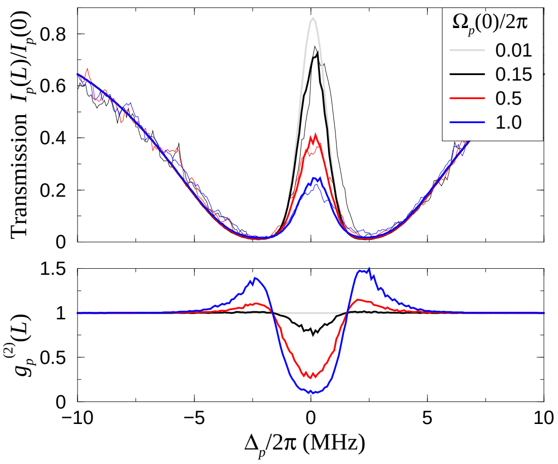

Figure 4. Top: Probe field transmission $I_p(L)/I_p(0)$ versus detuning Δ_p, for input intensities corresponding to $\Omega_p(0)/2\pi = 0.01, 0.15, 0.5, 1.0$ MHz. Thin lines are experimental curves from [17], thicker lines are obtained via stochastic simulations of Eqs. (11)-(13) averaged over 10 independent realizations. Bottom: The corresponding intensity correlation functions $g_p^{(2)}(L)$.

3.2. Numerical simulations and comparison with the experiment

We employ our theory to simulate the experiment of Ref. [17] with an ensemble of cold ^{87}Rb atoms: $|g\rangle \equiv 5S_{1/2}|F = 2, m_F = 2\rangle$, $|e\rangle \equiv 5P_{3/2}|F = 3, m_F = 3\rangle$ with $\Gamma_e = 3.8 \times 10^7$ s^{-1}, and $|r\rangle \equiv 60S_{1/2}$ with $\Gamma_r = 5 \times 10^3$ s^{-1} and $C_6/2\pi = 1.4 \times 10^{11}$ s$^{-1}\mu$m^6 corresponding to repulsive VdW interactions. $\gamma_{e,r}$ also include the one- and two-photon laser linewidths $\delta\omega_{1,2}/2\pi \simeq (5.7, 11) \times 10^4$ s^{-1}. The atomic density $\rho = 1.2 \times 10^7$ mm^{-3} and the medium length $L = 1.3$ mm lead to the resonant optical depth of $\kappa L = 4.524$. The control field $\Omega_c/2\pi = 2.25 \times 10^6$ s^{-1}‡ is slightly detuned by $\delta_c/2\pi = -10^5$ s^{-1}. The corresponding blockade radius is $R_{\rm sa} \simeq 6.6\,\mu$m and each SA contains on average $\bar{n}_{\rm sa} \simeq 14.7$ atoms.

In Fig. 4 we compare the transmission spectra for different input probe intensities with the corresponding plots of Ref. [17]. The weak field of $\Omega_p/2\pi \lesssim 0.01$ MHz leads to linear EIT response of the medium; the VdW interaction induced nonlinearities become important only at higher intensities. The agreement between our stochastic simulations and the experiment is remarkable. We also show the local intensity correlation $g_p^{(2)}(L)$ at the exit from the medium.

Figure 5 summarizes the results of our simulations involving the continuous polarizability of Eq. (12). Increasing the input probe intensity leads to lesser transmission through the EIT window ($\Delta_2 \sim 0$) and to small mean-field shift and broadening of the EIT line. This is due to the higher probability of two or more photons, exciting Rydberg states $|r\rangle$, to be at the same SA. The induced large VdW level shift $\bar{\Delta}(0)$ results in strong photon absorption, simultaneously reducing the photon coincidence probability within the SA volume $V_{\rm sa}$. Hence, both $I_p(z)$ and $g_p^{(2)}(z)$ decay, but once $g_p^{(2)}(z) \ll 1$, the attenuation of the probe field intensity $I_p(z)$ slows down. Eventually I_p saturates at a value corresponding to less than one photon per SA, $\rho_{\rm phot} \lesssim \rho_{\rm sa}$, with vanishing coincidence probability. With $\rho_{\rm phot} = \hbar\epsilon_0 c I_p/(2\wp_{ge}^2\omega_p v)$, where the probe group velocity $v = 2|\Omega_c|^2/(\kappa\gamma_e)$ ($\simeq 6000$ m/s),

‡ Our definition of the Rabi frequencies $\Omega_{p,c}$ differ from that in [17] by a factor of $\frac{1}{2}$

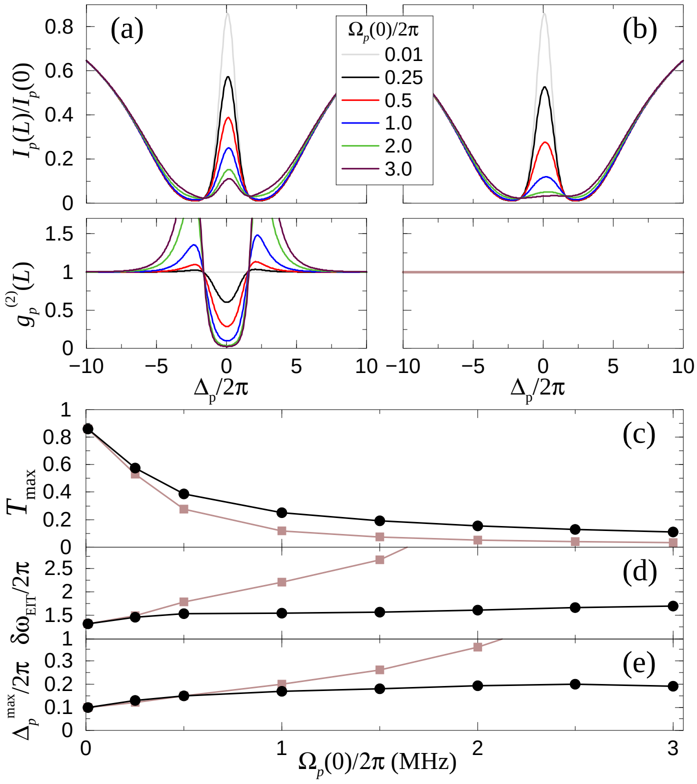

Figure 5. (a) Transmission (top) and intensity correlation (bottom) spectra of the probe field, for various input intensities. (b) The same, but setting $g_p^{(2)}(z) = 1 \, \forall \, z \in [0, L]$. (c) Peak probe transmission $T_{\max}$ around the EIT line center, (d) EIT linewidth $\delta\omega_{\rm EIT}$ (FWHM), and (e) detuning $\Delta_p^{\max}$ at the maximum $T_{\max}$, versus the input probe Rabi frequency $\Omega_p(0)$. The black lines (circles) correspond to case (a) and the brown lines (squares) to (b). $\Delta_p, \delta\omega_{\rm EIT}, \Omega_p$ are in MHz.

we have that $\rho_{\rm phot} = (\rho/4)\langle\hat{\Omega}_p^\dagger\hat{\Omega}_p\rangle/|\Omega_c|^2$ and the maximal saturation intensity is $\langle\hat{\Omega}_p^\dagger\hat{\Omega}_p\rangle \simeq (4\rho_{\rm sa}/\rho)|\Omega_c|^2$. In the medium the photons are anticorrelated (antibunched) within the temporal window of $\delta t \simeq 2R_{\rm sa}/v$ ($\simeq 1.6$ ns), which does not change when they leave the medium for free space.

Had we not taken into account the probe field intensity correlation, equivalent to setting $g_p^{(2)}(z) = 1 \, \forall \, z \in [0, L]$, Fig. 5(b), we would have faster, exponential decay of $I_p(z)$, unrestrained by the buildup of avoided volume between the photons, as well as sizable shift and broadening of the EIT line, which contradict the observations of [17].

Outside the EIT window, around the Autler-Townes doublet $\Delta_2 \sim \pm\Omega_c$, the probe is strongly absorbed, $\mathrm{Im}[\langle\alpha\rangle] \simeq 1$, but the correlation function is amplified, since in Eq. (13) $\mathrm{Im}[\alpha_{\rm TLA} - \alpha_{\rm EIT}] < 0$. In other words, linear absorption is larger than the conditional absorption, which results in photon bunching but very low flux.

4. Conclusions

Electromagnetically induced transparency in a medium of strongly interacting Rydberg atoms offers novel, highly non-linear regimes of field propagation. At the low-light level, the induced giant cross-phase modulation enables the realization of universal phase gate between single photon pulses. For stronger input fields, EIT via atomic Rydberg states is suppressed by collective excitations of SAs, which depend on the local probe field intensity and its two-particle correlation within the SA (blockade) volume. The buildup of anticorrelations between the photons upon propagation through the medium leads to the saturation of transmitted field intensity to a value corresponding to one photon per blockade volume. The transmitted field then corresponds to a train of non-overlapping single-photon pulses with the temporal separation δt of a few ns.

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