Electromagnetically Induced Transparency with Rydberg Atoms

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(Received 8 June 2011; revised manuscript received 1 October 2011; published 14 November 2011)

We present a theory of electromagnetically induced transparency in a cold ensemble of strongly interacting Rydberg atoms. 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 model is computationally very efficient and is in quantitative agreement with the results of the recent experiment of Pritchard *et al.* [Phys. Rev. Lett. **105**, 193603 (2010)]

DOI: 10.1103/PhysRevLett.107.213601 PACS numbers: 42.50.Gy, 32.80.Ee, 42.65.−k

Strong dipole-dipole or van der Waals (vdW) interactions between atoms in highly excited Rydberg states [1] constitute the basis for promising quantum information schemes [2] and interesting many-body effects [3–8]. Many of these studies utilize the dipole blockade mechanism [9–13] which suppresses multiple Rydberg excitations within a certain interaction (blockade) volume. Electromagnetically induced transparency (EIT) [14] can translate the interactions between Rydberg atoms into sizable interactions between single photons [15–17].

Recently, several experiments on EIT [18–21], and the closely related CPT (coherent population trapping) [22], with Rydberg atoms were performed. Strong vdW interactions between the atomic Rydberg states were prominently manifest in Ref. [21]: 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. Here we develop a theoretical model for EIT with Rydberg atoms, whose predictions fully reproduce the experimental observations [21]. The crux of our approach is the coarse-grained treatment of the atomic medium composed of effective superatoms (SAs), with each SA represented by collective states of atoms in the blockade volume that can accommodate only one Rydberg excitation. A weak probe field propagates through the EIT medium with little attenuation, but for a stronger field with more than one photon per SA, the excess photons are subject to enhanced—essentially two-level atom—absorption. This leads to the field attenuation with the simultaneous buildup of an avoided volume between the probe photons [17]. The inclusion of two-photon correlations is the key feature of our work, not present in the numerical simulations of [21] and recent theoretical studies [23] which agreed with the experiment at weak probe fields but had significant discrepancies for stronger fields. Our theory is not limited to weak probe fields and/or low atomic densities, yet, despite intrinsic nonlinearity, it is intuitive and numerically efficient, amounting to the solution of a pair of coupled differential equations for the probe field intensity and its second-order correlation, in the spirit of the Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy.

Consider an ensemble of $N = \int_V d^3 r \rho(\mathbf{r})$ cold atoms of density $\rho(\mathbf{r})$ in the (quantization) volume V interacting with two optical fields. The quantized probe field $\hat{\mathcal{E}}_p$ of frequency ω_p acts on the atomic transition between the ground $|g\rangle$ and excited $|e\rangle$ states, and the control field of frequency ω_c drives the transition $|e\rangle \to |r\rangle$ with Rabi frequency Ω_c [see Fig. 1(a)]. A pair of atoms i and j at positions $\mathbf{r}_i$ and $\mathbf{r}_j$ excited to the Rydberg states $|r\rangle$ interact with each other via a vdW potential $\hbar\Delta(\mathbf{r}_i - \mathbf{r}_j) = \hbar C_6/|\mathbf{r}_i - \mathbf{r}_j|^6$ [24]. In the frame rotating with frequencies

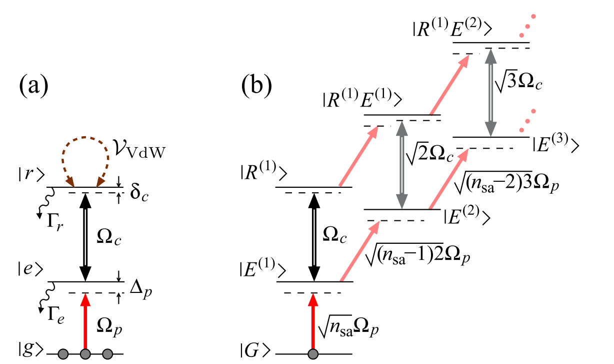

FIG. 1 (color online). (a) Level scheme of atoms interacting with probe Ω_p and control Ω_c fields with detunings Δ_p and δ_c. Γ_e and Γ_r are (population) decay rates of states $|e\rangle$ and $|r\rangle$, and $\mathcal{V}_{\mathrm{vdW}}$ denotes vdW interaction between atoms in Rydberg state $|r\rangle$. (b) Truncated level scheme of the superatom, composed of n_{SA} atoms, with corresponding transition amplitudes due to the probe and control fields.

$\omega_{p,c}$, the system Hamiltonian $\mathcal{H} = \mathcal{H}_{\rm a} + \mathcal{V}_{\rm af} + \mathcal{V}_{\rm vdW}$ contains the unperturbed atomic part, $\mathcal{H}_{\rm a} = -\hbar\sum_j^N[\Delta_p\hat{\sigma}_{ee}^j + (\Delta_p + \delta_c)\hat{\sigma}_{rr}^j]$, and the atom-field and vdW interactions, $\mathcal{V}_{\rm af} = -\hbar\sum_j^N[\hat{\Omega}_p(\mathbf{r}_j)\hat{\sigma}_{eg}^j + \Omega_c\hat{\sigma}_{re}^j + {\rm H.c.}]$ and $\mathcal{V}_{\rm vdW} = \hbar\sum_{i<j}^N\hat{\sigma}_{rr}^i\Delta(\mathbf{r}_i - \mathbf{r}_j)\hat{\sigma}_{rr}^j$, where $\hat{\sigma}_{\mu\nu}^j \equiv |\mu\rangle_{jj}\langle\nu|$ is the transition operator for atom j at position $\mathbf{r}_j$, $\Delta_p = \omega_p - \omega_{eg}$ and $\delta_c = \omega_c - \omega_{re}$ are the detunings of the probe and control fields, and $\hat{\Omega}_p = \eta\hat{\mathcal{E}}_p$ is the operator of the probe Rabi frequency, with $\eta = \wp_{ge}\sqrt{\omega_p/(2\hbar\epsilon_0 V)}$ the atom-field coupling strength on the $|g\rangle \to |e\rangle$ transition with dipole moment $\wp_{ge}$.

We consider the evolution of the probe field only along its propagation z axis, and assume uniform, undepleted control field Ω_c. Using Hamiltonian $\mathcal{H}$, we obtain Heisenberg-Langevin equations for the field $\hat{\mathcal{E}}_p(\mathbf{r})$ and continuous, appropriately averaged, atomic $\hat{\sigma}_{\mu\nu}(\mathbf{r})$ operators:

$$(\partial_t + c\partial_z)\hat{\mathcal{E}}_p(\mathbf{r}) = i\eta N\hat{\sigma}_{ge}(\mathbf{r}), \tag{1a}$$

$$\partial_t\hat{\sigma}_{ge}(\mathbf{r}) = (i\Delta_p - \gamma_e)\hat{\sigma}_{ge}(\mathbf{r}) + i\Omega_c^*\hat{\sigma}_{gr}(\mathbf{r}) + i\hat{\Omega}_p(\mathbf{r})[\hat{\sigma}_{gg}(\mathbf{r}) - \hat{\sigma}_{ee}(\mathbf{r})], \tag{1b}$$

$$\partial_t\hat{\sigma}_{gr}(\mathbf{r}) = \{i[\Delta_2 - \hat{S}(\mathbf{r})] - \gamma_r\}\hat{\sigma}_{gr}(\mathbf{r}) - i\hat{\Omega}_p(\mathbf{r})\hat{\sigma}_{er}(\mathbf{r}) + i\Omega_c\hat{\sigma}_{ge}(\mathbf{r}), \tag{1c}$$

where $\gamma_e \geq \frac{1}{2}\Gamma_e$ and γ_r ($\ll \gamma_e$) are the transversal relaxation rates, with the associated noise operators dropped, $\Delta_2 = \Delta_p + \delta_c$ is the two-photon detuning, and $\hat{S}(\mathbf{r}) \equiv \int d^3r'\rho(\mathbf{r}')\Delta(\mathbf{r} - \mathbf{r}')\hat{\sigma}_{rr}(\mathbf{r}')$ 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. (1) are highly nonlocal. We therefore need to contrive an efficient method to evaluate the vdW shift $\hat{S}(\mathbf{r})$.

We shall be concerned with stationary interaction and drop in Eqs. (1) all the time derivatives. Consider for the moment Eqs. (1b) and (1c) without the vdW shift $\hat{S}$ and small relaxation γ_r terms. When $\Omega_{p,c}, |\Delta_{p,2}| < \gamma_e$, we can approximate the population of Rydberg state $|r\rangle$ by a Lorentzian function of Δ_2: $\langle\hat{\sigma}_{rr}\rangle \approx \langle\hat{\Omega}_p^\dagger\hat{\Omega}_p\rangle/(|\Omega_c|^2 + \Delta_2^2\gamma_e^2/|\Omega_c|^2)$, with the half-width $w \equiv |\Omega_c|^2/\gamma_e$. Observe now that an atom in Rydberg state $|r\rangle$ would induce vdW shift $\Delta(R)$ of level $|r\rangle$ for another atom separated by distance R, which effectively translates into the two-photon detuning Δ_2. The vdW interaction then blocks the excitation of all the atoms for which $\Delta(R) \gtrsim w$. This is the essence of the Rydberg blockade [9]. We therefore define the blockade radius $R_{\rm SA} \simeq \sqrt[6]{C_6/w}$ and call the ensemble of $n_{\rm SA} = \rho V_{\rm SA}$ atoms within volume $V_{\rm SA} = \frac{4\pi}{3}R_{\rm SA}^3$ "superatom" (SA). Since in general the atomic density $\rho(\mathbf{r})$ varies with position $\mathbf{r}$, so does $n_{\rm SA}(\mathbf{r})$, but the density of SAs $\rho_{\rm SA} = V_{\rm SA}^{-1} = \frac{3}{4\pi}\sqrt{|\Omega_c|^2/(\gamma_e C_6)}$ is constant.

Each SA can contain only one Rydberg excitation delocalized over $V_{\rm SA}$. We may therefore 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 $2R_{\rm SA}$ [25,26]. The total vdW shift $\hat{S}(\mathbf{r})$ at position $\mathbf{r}$ can then be expressed as

$$\hat{S}(\mathbf{r}) \approx \sum_j^{N_{\rm SA}}\Delta(\mathbf{r} - \mathbf{r}_j)\hat{\Sigma}_{RR}(\mathbf{r}_j) = \bar{\Delta}\hat{\Sigma}_{RR}(\mathbf{r}) + \hat{s}(\mathbf{r}), \tag{2}$$

where $\hat{\Sigma}_{RR}(\mathbf{r}_j)$ is the projector onto the Rydberg excitation of the SA at $\mathbf{r}_j$. The physical meaning of the first term on the right-hand side of Eq. (2) 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: $\bar{\Delta} \simeq \frac{1}{V_{\rm SA}}\int_{V_{\rm SA}}\Delta(\mathbf{r}')d^3r' \to \infty$. Actually, for a small cutoff in the interatomic separation, $|\bar{\Delta}| \gg \gamma_e$ is finite but very large, which is the only relevant property. 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 $\langle\hat{s}(\mathbf{r})\rangle = \frac{w}{8}\langle\hat{\Sigma}_{RR}(\mathbf{r})\rangle$.

Assuming that the probe field $\hat{\Omega}_p$ varies little over distance $\sim R_{\rm SA}$, we can describe the dynamics of individual SAs in terms of collective states and operators defined within the blockade volume $V_{\rm SA}$. The level scheme of the SA is shown in Fig. 1(b): $|G\rangle = |g_1, g_2, \ldots, g_{n_{\rm SA}}\rangle$ is the ground state, and $|R^{(1)}\rangle = \frac{1}{\sqrt{n_{\rm SA}}} \times \sum_j^{n_{\rm SA}}|g_1, g_2, \ldots, r_j, \ldots, g_{n_{\rm SA}}\rangle$ 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 $\langle E^{(1)}|\mathcal{V}_{\rm af}|G\rangle = \sqrt{n_{\rm SA}}\hat{\Omega}_p$, $\langle R^{(1)}|\mathcal{V}_{\rm af}|E^{(1)}\rangle = \Omega_c$, etc., depend on the number of atoms $n_{\rm SA}$ in $V_{\rm SA}$. In order to calculate $\hat{\Sigma}_{RR}$, we now proceed along the lines similar to the single atom treatment. Starting with the SA in $|G\rangle$, we adiabatically eliminate all the excited states $|E^{(k)}\rangle$ having large widths $\sim k\gamma_e$. Note that state $|R^{(1)}\rangle$ is reached from $|G\rangle$ by 2-photon transition, while all the other states $|R^{(1)}E^{(k)}\rangle$ require $2 + k$ photon transitions; therefore their adiabatic elimination affects little the populations of $|G\rangle$ and $|R^{(1)}\rangle$. 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 $|G\rangle \to |R^{(1)}\rangle$ when the number density of probe photons $\rho_{\rm phot}$ is comparable to, or larger than, the density of SAs $\rho_{\rm SA}$, we take $\hat{\Sigma}_{GG} + \hat{\Sigma}_{RR} = \mathbb{1}$, which finally yields [27]

$$\hat{\Sigma}_{RR} = \frac{|\Omega_c|^2 n_{\rm SA}\hat{\Omega}_p^\dagger\hat{\Omega}_p}{|\Omega_c|^2 n_{\rm SA}\hat{\Omega}_p^\dagger\hat{\Omega}_p + [|\Omega_c|^2 - \Delta_p\Delta_2]^2 + \Delta_2^2\gamma_e^2}. \tag{3}$$

We next examine the probe field propagation in the atomic medium. For moderate Rabi frequency $\Omega_p < \gamma_e$ and number density of photons $\rho_{\rm phot} \ll \rho$, we can assume linear response of individual atoms to the applied field, setting $\hat{\sigma}_{ee}, \hat{\sigma}_{er} \to 0$ and $\hat{\sigma}_{gg} = \mathbb{1}$. We then arrive at the propagation equation for the probe field amplitude, $\partial_z \hat{\mathcal{E}}_p = i \frac{\kappa}{2} \hat{\alpha} \hat{\mathcal{E}}_p$, where $\kappa = \varsigma_0 \rho$ is the resonant (intensity) absorption coefficient proportional to the atomic absorption cross section $\varsigma_0 = \omega_p |\wp_{ge}|^2/(\hbar \epsilon_0 c \gamma_e)$, while

$$\hat{\alpha}(\mathbf{r}) = \hat{\Sigma}_{RR}(\mathbf{r}) \frac{i\gamma_e}{\gamma_e - i\Delta_p} + [1 - \hat{\Sigma}_{RR}(\mathbf{r})] \times \frac{i\gamma_e}{\gamma_e - i\Delta_p + |\Omega_c|^2[\gamma_r - i(\Delta_2 - \langle \hat{s}(\mathbf{r}) \rangle)]^{-1}} \quad (4)$$

is the operator-valued polarizability. Here the first fraction is the polarizability $\alpha_{\rm TLA}$ of a two-level atom, while the second fraction, barring the small shift $\langle \hat{s}(\mathbf{r}) \rangle$, is the usual EIT polarizability $\alpha_{\rm EIT}$ [14]. Physically, if the SA at position $\mathbf{r}$ contains a Rydberg excitation $[\hat{\Sigma}_{RR}(\mathbf{r}) \to 1]$, the two-photon detuning is shifted out of the EIT window $[\bar{\Delta} \gg \gamma_e]$ and the probe field $\hat{\mathcal{E}}_p$ sees an absorbing two-level system; if no Rydberg excitation is present, the medium response is that of usual EIT with a small mean-field shift due to the vdW interaction with the external SAs. Then the expectation value of the probe field intensity obeys the equation

$$\partial_z \langle \hat{\mathcal{E}}_p^\dagger(\mathbf{r}) \hat{\mathcal{E}}_p(\mathbf{r}) \rangle = -\kappa(\mathbf{r}) \langle \hat{\mathcal{E}}_p^\dagger(\mathbf{r}) {\rm Im}[\hat{\alpha}(\mathbf{r})] \hat{\mathcal{E}}_p(\mathbf{r}) \rangle. \quad (5)$$

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 [17] originating from nonlinear response of the atoms to the Rydberg excitations. We therefore proceed more carefully and replace $\hat{\alpha}(\mathbf{r})$ in Eq. (5) by its expectation value *conditioned* upon the presence of a photon at $\mathbf{r}$, denoted by $\langle \cdots \rangle_{\mathbf{r}}$,

$$\langle \hat{\alpha}(\mathbf{r}) \rangle_{\mathbf{r}} = \langle \hat{\Sigma}_{RR}(\mathbf{r}) \rangle_{\mathbf{r}} \alpha_{\rm TLA} + [1 - \langle \hat{\Sigma}_{RR}(\mathbf{r}) \rangle_{\mathbf{r}}] \alpha_{\rm EIT}. \quad (6)$$

The conditional Rydberg population $\langle \hat{\Sigma}_{RR} \rangle_{\mathbf{r}}$ of the SA at $\mathbf{r}$ is obtained from Eq. (3) 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 $V_{\rm SA}^{(\mathbf{r})}$. The field intensity is now coupled to its two-photon correlation $g_p^{(2)}(\mathbf{r})$ which in turn evolves upon propagation. 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 $\propto {\rm Im}[\langle \alpha(\mathbf{r}) \rangle - \alpha_{\rm EIT}]$ modifies $g_p^{(2)}$, which therefore obeys the equation of motion [28]

$$\partial_z g_p^{(2)}(\mathbf{r}) = -\kappa(\mathbf{r}) \langle \hat{\Sigma}_{RR}(\mathbf{r}) \rangle {\rm Im}[\alpha_{\rm TLA} - \alpha_{\rm EIT}] g_p^{(2)}(\mathbf{r}). \quad (7)$$

Hence, within the EIT window, where ${\rm Im}[\alpha_{\rm EIT}] \simeq 0$, the correlations between the photon pairs with relative distance smaller than the blockade (SA) radius decay with the rate proportional to the probability of SA excitation $\langle \hat{\Sigma}_{RR} \rangle$ and the absorption rate of a two-level system ${\rm Im}[\alpha_{\rm TLA}]$. We note that our treatment involves only a single transverse mode of the probe field, which is effectively defined by the SA cross section. If, however, during propagation there is strong mixing of the transverse modes, it would preclude the buildup of (anti)correlations between the photons.

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 spatially integrate the coupled coarse-grained Eqs. (5)–(7) 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. (6).

We employ our theory to simulate the experiment of Ref. [21] 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} μm^6 [24] 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 is $\rho(z) = \rho_0 \exp[-(z - z_0)^2/2\sigma_\rho^2]$ with peak $\rho_0 = 1.32 \times 10^7$ mm^{-3} and half-width $\sigma_\rho = 0.7$ mm; indistinguishable results are obtained for homogeneous ensemble of density $\bar{\rho} = 1.2 \times 10^7$ mm^{-3} and length $L = 1.3$ mm, leading to the resonant optical depth of $\bar{\kappa} L = 4.524$. The control field $\Omega_c/2\pi = 2.25 \times 10^6$ s^{-1} [29] is slightly detuned by $\delta_c/2\pi = -10^5$ s^{-1}. The corresponding blockade radius is $R_{\rm SA} \simeq 6.6$ μm and each SA contains on average $\bar{n}_{\rm SA} \simeq 14.7$ atoms. We emphasize that our simulations are insensitive to moderate variations ($\pm 20\%$) of the SA volume $V_{\rm SA}$ and the number of atoms $n_{\rm SA}$ ($\simeq 14 \pm 3$) it contains.

In Fig. 2 we compare the transmission spectra for different input probe intensities with the corresponding plots of Ref. [21]. Already for $\Omega_p(0)/2\pi \gtrsim 0.1$ MHz the vdW interaction induced nonlinearities play an important role. 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 3 summarizes the results of our simulations involving the continuous polarizability of Eq. (6). The weak field of $\Omega_p/2\pi \lesssim 0.01$ MHz encounters linear EIT response. Increasing the input probe intensity leads to lesser

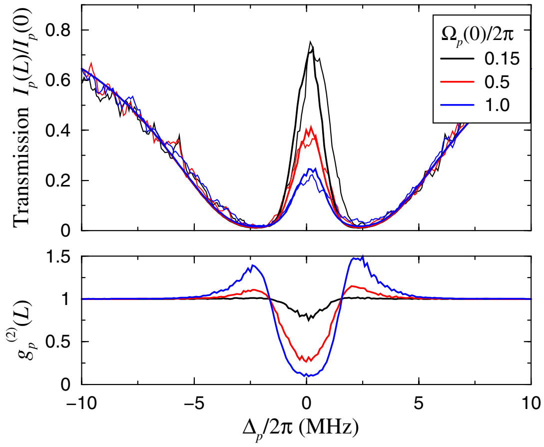

FIG. 2 (color online). Top: Probe field transmission $I_p(L)/I_p(0)$ versus detuning Δ_p, for different input intensities corresponding to $\Omega_p(0)/2\pi = 0.15, 0.5, 1.0$ MHz. Thin lines are experimental curves from [21], thicker lines are obtained via stochastic simulations of Eqs. (5)–(7) averaged over 10 independent realizations. Bottom: The corresponding intensity correlation functions $g_p^{(2)}(L)$.

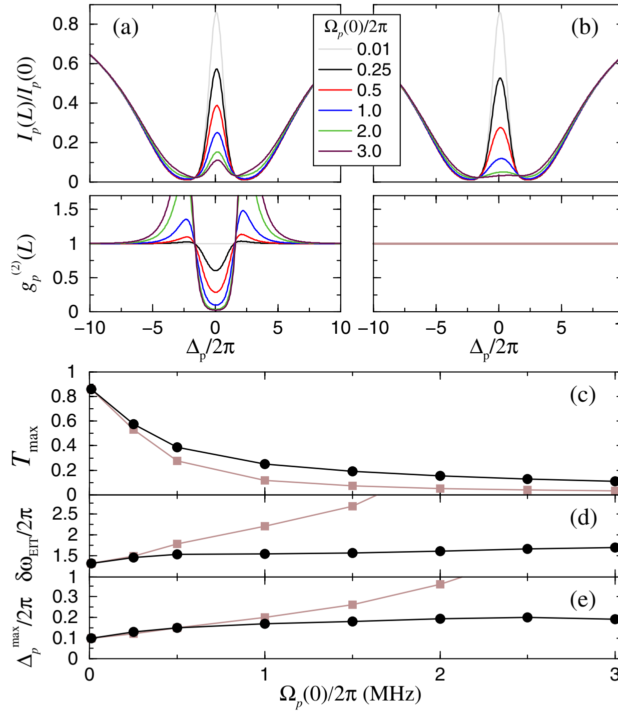

FIG. 3 (color online). (a) Transmission (top) and intensity correlation (bottom) spectra of the probe field, for various input intensities. (b) The same, but setting $g_p^{(2)}(z) = 1\ \forall\ z \in [0, L]$. (c) Peak probe transmission $T_{\max}$ around the EIT line center, (d) EIT linewidth $\delta\omega_{\rm EIT}$ (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). Δ_p, $\delta\omega_{\rm EIT}$, Ω_p are in MHz.

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 $\bar{\Delta}$ 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 $I_p = \langle \hat{\Omega}_p^\dagger \hat{\Omega}_p \rangle$ 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 $v = 2|\Omega_c|^2/(\bar{\kappa}\gamma_e)$ ($\simeq 6000$ m/s) is the probe group velocity within the EIT window $|\Delta_2| \lesssim \delta\omega_{\rm EIT}$, we have that $\rho_{\rm phot} = (\rho/4) \times \langle \hat{\Omega}_p^\dagger \hat{\Omega}_p \rangle / |\Omega_c|^2$ and the maximal saturation intensity is $I_p^{\max} \simeq (4\rho_{\rm SA}/\rho)|\Omega_c|^2$. 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. 3(b), we would have had 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 [21].

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. (7) $\mathrm{Im}[\alpha_{\rm TLA} - \alpha_{\rm EIT}] < 0$. In other words, linear absorption is larger than the conditional absorption, which results in photon bunching but very low flux.

We finally note that for relatively strong input fields $\Omega_p(0) \lesssim \gamma_e$ of Fig. 3, the validity of linear response of the atoms inherent in polarizability of Eq. (4) may not be *a priori* justified. This is indeed the case for an optically thin atomic medium. But in the optically thick medium, within a few absorption lengths, even a strong probe field and its two-photon correlation function quickly decay to the level at which the above approximation is justified.

To conclude, EIT via atomic Rydberg states is suppressed by collective Rydberg excitations of SAs which depend on the local probe field intensity and its two-particle correlation within the SA (blockade) volume. For strong input fields, 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. Conversely, suitably antibunched input fields should exhibit large transmission affected only by small linear absorption.

In a one-dimensional configuration, the spatial correlations between the photons in the medium translate at the output into temporal correlations in free space, which can be measured by coincidence detection. The limit of maximal saturation intensity $I_p^{\max}$ of the transmitted through the EIT window field then corresponds to a train of nonoverlapping single-photon pulses with the temporal separation δt of a few ns.

We are grateful to J. D. Pritchard and C. S. Adams for sharing with us the experimental details. We thank A. Gorshkov, Th. Pohl, and M. D. Lukin for stimulating discussions. This work was supported by the Humboldt Foundation (D. P.), the Harvard Quantum Optics Center (J. O.), and SFB TR49 (M. F.).

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[28] A formal derivation of Eq. (7) involves expanding the full derivative of $g_p^{(2)}(\mathbf{r})$ in terms of $k = 0, 1, 2$ photon states of the probe field within the blockade volume.
[29] Our definition of the Rabi frequencies $\Omega_{p,c}$ differ from that in [21] by a factor of $\frac{1}{2}$.