



CHORUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

Dissipative Many-Body Quantum Optics in Rydberg Media

Alexey V. Gorshkov, Rejish Nath, and Thomas Pohl

Phys. Rev. Lett. **110**, 153601 — Published 9 April 2013

DOI: [10.1103/PhysRevLett.110.153601](https://doi.org/10.1103/PhysRevLett.110.153601)

Dissipative Many-body Quantum Optics in Rydberg Media

Alexey V. Gorshkov,¹ Rejish Nath,² and Thomas Pohl³

¹*Institute for Quantum Information and Matter,
California Institute of Technology, Pasadena, CA 91125, USA*

²*Institut for Quantum Optics and Quantum Information and Institute for Theoretical Physics,
University of Innsbruck, A-6020 Innsbruck, Austria*

³*Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany*

We develop a theoretical framework for the dissipative propagation of quantized light under conditions of electromagnetically induced transparency in atomic media involving strongly interacting Rydberg states. The theory allows us to determine the peculiar spatiotemporal structure of the output of the recently demonstrated single-photon filter and the recently proposed single-photon subtractor, which, respectively, let through and absorb a single photon. In addition to being crucial for applications of these and other optical quantum devices, the theory opens the door to the study of exotic dissipative many-body dynamics of strongly interacting photons in nonlinear nonlocal media.

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

Dissipation has recently been recognized as a powerful tool for quantum information and many-body physics [1–11]. A particular example, realized in recent experiments [12–14], is the propagation of quantized light fields in Rydberg media [15–18] under the conditions of electromagnetically induced transparency (EIT) [19]. While Rydberg states provide strong long-range atom-atom interactions, EIT provides strong atom-light interactions with controlled dissipation. The resulting combination gives rise to strong and often dissipative photon-photon interactions [20–23], which can be used to generate a variety of non-classical states of light [12–14, 24–38] and to implement photon-photon and atom-photon quantum gates [21, 23, 39, 40]. First wavefunction-based descriptions of two-photon propagation in Rydberg EIT media have revealed the emergence of correlated two-photon losses that could enable the deterministic generation of single photons [13, 21]. Yet, the fate of the remaining photon as well as the underlying dissipative many-body dynamics have remained unclear despite their essential role in the performance of future Rydberg-EIT-based nonlinear optical quantum devices.

In this Letter, we address these outstanding questions and develop a theory for the dissipative many-body dynamics of quantized light fields in a strongly interacting medium. In contrast to earlier studies [13, 21], our theory provides information about the many-body density matrix of the light field, i.e. it faithfully describes the process of populating the m -photon states from the $(m+1)$ -photon manifold as a photon scatters. In addition to opening the door to the study of photonic dissipative many-body physics, the theory allows one to compute the complex spatiotemporal structure of the generated non-classical light fields, whose understanding is crucial for applications. As two important examples that illustrate this point and evince the power of our method, we consider the recently demonstrated single-photon filter [13, 21] and the recently proposed single-photon subtractor

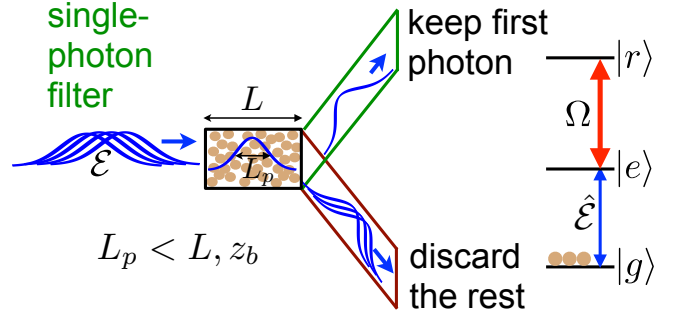


FIG. 1. Single-photon filter. A classical field with Rabi frequency Ω resonantly couples the excited state $|e\rangle$ to the Rydberg state $|r\rangle$ controls the propagation of the quantum field $\hat{\mathcal{E}}$. The EIT-compressed pulse length L_p is assumed to be smaller than the length L of the medium and the blockade radius z_b .

tor [26]. In the limit of strong interactions, our approach yields exact solutions to the dissipative many-body evolution, provides an intuitive picture of the underlying physics, and highlights the importance of the entrance dynamics of the incoming photons. This dynamics may be crucial for photon storage in a nonlinear quantum memory [12], while the developed theoretical framework should enable the understanding of recent experiments [13, 14] beyond the limit of extremely weak input.

The basic physics can be illustrated by considering the example of a single-photon filter shown in Fig. 1. In the absence of interactions, the probe field $\hat{\mathcal{E}}$ couples with an effective spin wave of Rydberg atoms $|r\rangle$ to form a slow-light polariton [19]. Whenever two polaritons are within the so-called blockade radius z_b of each other, the strong interactions between $|r\rangle$ atoms destroy EIT and lead to strong dissipation [21]. If the EIT-compressed pulse length L_p is smaller than the length L of the medium and the blockade radius z_b , the first photon propagates without losses under EIT conditions but causes scatter-

ing of all subsequent photons [21]. The density matrix of the first photon is obtained by tracing over all the subsequent photons. Since the first photon must already be inside the medium to cause scattering, the timing of the first scattering event [t_2 in Eqs. (7,10)] carries information about the first photon. Therefore, the transmitted single photon is impure. In the following, we develop a master-equation-type framework that allows one to determine the state of the output photons in this and other Rydberg-EIT problems, including those that do not satisfy the condition $L_p < L, z_b$.

Setup.—Let $\hat{\mathcal{E}}^\dagger(z)$, $\hat{P}^\dagger(z)$ ($\sim |e\rangle\langle g|$), and $\hat{S}^\dagger(z)$ ($\sim |r\rangle\langle g|$) be the slowly-varying operators for the creation of a photon, an excitation in state $|e\rangle$, and a Rydberg excitation $|r\rangle$, respectively, at position z [41–43]. Assuming that almost all atoms are in $|g\rangle$ at all times, the operators satisfy the same-time commutation relations $[\hat{\mathcal{E}}(z), \hat{\mathcal{E}}^\dagger(z')] = [\hat{P}(z), \hat{P}^\dagger(z')] = [\hat{S}(z), \hat{S}^\dagger(z')] = \delta(z - z')$ [41–43]. The Heisenberg equations of motion inside the medium $z \in [0, L]$ are [13, 21, 41–43]

$$\begin{aligned} \partial_t \hat{\mathcal{E}}(z, t) &= -\partial_z \hat{\mathcal{E}}(z, t) + ig\hat{P}(z, t), & (1) \\ \partial_t \hat{P}(z, t) &= -\hat{P}(z, t) + ig\hat{\mathcal{E}}(z, t) + i\Omega\hat{S}(z, t) + \hat{F}_P(z, t), & (2) \\ \partial_t \hat{S}(z, t) &= i\Omega\hat{P}(z, t) - i\int dz' V(z - z')\hat{S}^\dagger(z')\hat{S}(z')\hat{S}(z). & (3) \end{aligned}$$

Here $V(z) = C_6/z^6$ is the Rydberg-Rydberg interaction whose spatial non-locality contrasts with typical nonlinear quantum optical systems [44–46]. \hat{F}_P is a δ -correlated vacuum Langevin noise operator [47]. g is the collective atom-photon coupling constant, time and frequencies were rescaled by γ (the halfwidth of the $|g\rangle$ - $|e\rangle$ transition), while z was rescaled by c/γ . In these units, the blockade radius is given by $z_b = (C_6/\Omega^2)^{1/6}$ [21] and $c = 1$. Outside the medium ($z \notin [0, L]$), $\hat{S}(z, t)$ and $\hat{P}(z, t)$ are not defined and $(\partial_t + \partial_z)\hat{\mathcal{E}}(z, t) = 0$.

Incoming n -photon Fock state.—For simplicity, throughout the paper, we assume that the incoming pulse is confined to a single – for simplicity, real – arbitrary spatiotemporal mode $h(t)$ satisfying $\int dt h^2(t) = 1$. Then an incoming n -photon Fock state – before entering the medium – can be written as

$$|\psi(t)\rangle = \frac{1}{\sqrt{n!}} \left[\int_{-\infty}^{\infty} dx h(t-x)\hat{\mathcal{E}}^\dagger(x) \right]^n |0\rangle, \quad (4)$$

while its full density matrix at all times has the form

$$\rho(t) = \sum_{m=0}^n \rho_m(t), \quad (5)$$

where ρ_m contains m (photonic or atomic) excitations. The detection of the scattered photons by the environment erases the correlations between the $m+1$ terms in Eq. (5). The Heisenberg equations of motion (1-3) yield master-equation-type evolution equations for the matrix elements of ρ_m . If all but at most one photon are scattered, the only nonvacuum matrix element that survives

in the output field is $ee(x, y, t) = \text{tr}[\rho_1(t)\hat{\mathcal{E}}^\dagger(x)\hat{\mathcal{E}}(y)]$. As shown in the supplementary material [48], the dissipative propagation can be solved analytically for arbitrary photon number n under conditions of perfect EIT with $L_p < L, z_b$ and numerically for $n = 2$ without any restriction on the experimental parameters. In the former case, the resulting dynamics can be derived within a more general and simpler framework outlined below.

Perfect EIT with $L_p < L$ requires a large optical depth of the medium [19], implying that the absorption length is much smaller than the blockade radius z_b and the compressed pulse length L_p . Since $L_p < z_b$, at most one photon can propagate through the medium without losses. Then a fundamental question directly relevant to the experiments in Refs. [13, 14] is whether all n incoming photons are lost as they blockade each other's propagation or whether one photon indeed survives.

To answer this question, we work in the Schrödinger picture [49] and rewrite the input pulse [Eq. (4)] outside the medium by time-ordering the photons:

$$|\psi(t)\rangle = \sqrt{n!} \int_{t_n > \dots > t_1} \left[\prod_{i=1}^n dt_i h(t_i) \mathcal{E}^\dagger(t - t_i) \right] |0\rangle, \quad (6)$$

so that each set of t_i in Eq. (6) appears in Eq. (4) $n!$ times. While the n incoming photons are in the same spatial mode and hence indistinguishable, the possibility of time ordering is the crucial conceptual step in the derivation. As the first photon ($i = 1$) enters the medium, it turns into a Rydberg spin-wave excitation \hat{S} moving at the EIT group velocity $v_g = (\Omega/g)^2 c \ll c$. Since $L_p < z_b$, this single Rydberg excitation turns the entire medium seen by the remaining $n - 1$ photons into a resonant two-level medium. As the absorption length is much smaller than L_p , all the remaining photons get scattered [see term $-P(z, t)$ in Eq. (2)] into some other mode \hat{Q} as soon as they enter the medium. We will later trace over those loss channels, so we can assume without loss of generality that \hat{Q} is also a one-dimensional mode with commutation relation $[\hat{Q}(z), \hat{Q}^\dagger(z')] = \delta(z - z')$ [50]. Once the entire pulse is inside the medium, we, therefore, have

$$|\psi(t)\rangle = \sqrt{n!} \int_{t_n > \dots > t_2} \left[\prod_{i=2}^n dt_i h(t_i) \hat{Q}^\dagger(t - t_i) \right] |\psi_{t_2}(t)\rangle, \quad (7)$$

where

$$|\psi_{t_2}(t)\rangle = -\sqrt{v_g} \int_{-\infty}^{t_2} dt_1 h(t_1) \hat{S}^\dagger(v_g(t - t_1)) |0\rangle. \quad (8)$$

Taking $|\psi(t)\rangle\langle\psi(t)|$ and tracing over photons in mode \hat{Q} , we obtain

$$\rho(t) = \int dx dy S(x, y, t) S^\dagger(y) |0\rangle\langle 0| S(x) = \quad (9)$$

$$\int dt_2 n(n-1) h^2(t_2) \left[\int_{t_2}^{\infty} h^2(\tau) d\tau \right]^{n-2} |\psi_{t_2}(t)\rangle\langle\psi_{t_2}(t)|, \quad (10)$$

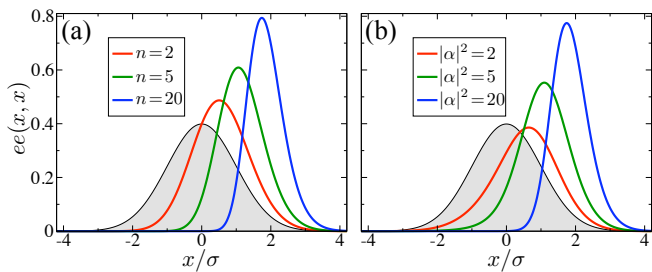


FIG. 2. The narrowing and advancing of the produced single-photon intensity profile $ee(x, x)$ with increasing input intensity. The input mode $h^2(x) \propto \exp[-x^2/(2\sigma^2)]$ is shaded. (a) Input is a Fock state with the indicated value of n . (b) Input is a coherent state with the indicated mean photon number $|\alpha|^2$.

where $ss(x, y, t) = \phi(x/v_g - t, y/v_g - t)/v_g$ is the density matrix of the remaining spin wave with

$$\phi(x, y) = nh(-x)h(-y) \left[\int_{-\infty}^{\min(x, y)} dz h^2(-z) \right]^{n-1}, \quad (11)$$

which together with Eq. (9) yields the dynamics inside the medium. For the purposes of this derivation, we have ignored the small photonic component $ee = v_g ss$. This solution has a simple physical interpretation in the spirit of master-equation unraveling [51]: The trace of the integrand in Eq. (10) is the probability that the second photon enters the medium (and immediately scatters) in the time interval $[t_2, t_2 + dt_2]$, while $|\psi_{t_2}(t)\rangle$ is the unnormalized spin wave that would be propagating in the medium had we detected that scattering event.

Transforming to a moving frame of reference, the density matrix of the output photon becomes $ee(x, y) = \phi(x, y)$ [see Eq. (11)]. This result shows that exactly one photon indeed survives the dissipative entrance dynamics: $\text{tr}[\rho] = \int dx \phi(x, x) = 1$. It also yields a remarkably simple result for the purity of the created photon:

$$\text{tr}[\rho^2] = \frac{n}{2n-1}. \quad (12)$$

As expected, the purity is smaller than unity because the timing of the scattering event carries some information about the remaining spin wave $|\psi_{t_2}(t)\rangle$. Crucially for applications, the purity does not vanish but approaches $1/2$ as $n \rightarrow \infty$. Surprisingly, it is independent of the mode shape $h(t)$. Furthermore, the eigenvalues p_i and eigenvectors $\phi_i(x)$ of $\phi(x, y)$ can be easily found [48] by using the change of variables $x \rightarrow \int_{-\infty}^x dz h^2(-z)$, which makes the density matrix and hence p_i independent of $h(t)$. Physically, this surprising behavior emphasizes the fact that the key role is played simply by the arrival *order* of n identical photons and not by the shape of the mode.

This dynamics at the medium boundary leads to a slight narrowing and advancing of the single-photon pulse

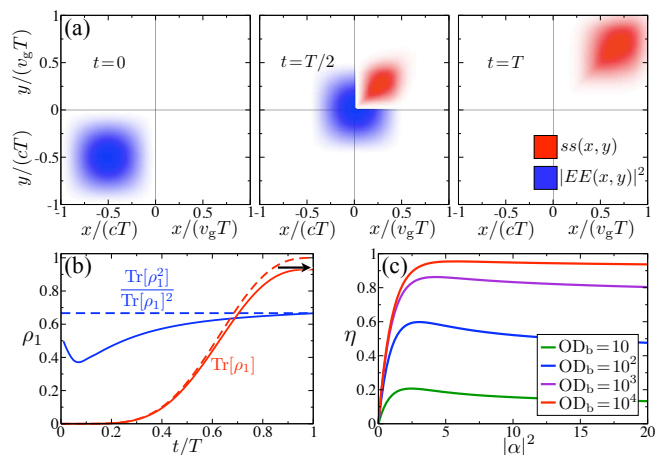


FIG. 3. (a) For an incoming $n = 2$ Fock state, the two-photon amplitude and the single-spin-wave density matrix at different times. See Ref. [48] for the full movie. (b) Approximate analytical (dashed) and exact numerical (solid) results for the efficiency $\text{tr}[\rho_1]$ (red) and purity $\text{tr}[\rho_1^2]/\text{tr}[\rho_1]^2$ (blue) of the single excitation. The purple arrow indicates the efficiency of retrieving Eq. (9) with $n = 2$. (c) As a function of the mean photon number $|\alpha|^2$ of the incoming coherent pulse, for the indicated values of the blocked optical depth OD_b , the estimated efficiency η of the single-photon source.

$\phi(x, x)$, as shown in Fig. 2(a) for a typical Gaussian input mode. This behavior can be traced back to the first scattering event, which projects the leading photon into the medium. This effect becomes more pronounced with increasing n , since the larger n is the sooner the first scattering event takes place. More succinctly, the probability distribution of the *first photon* is obviously advanced and narrower relative to the normalized probability distribution $h^2(t)$ of the *entire incident pulse*. Fortunately, for our Gaussian $h(t)$, $\phi(x, x)$ and $\phi_i(x)$ shorten extremely slowly with n as $\sim 1/\sqrt{\log n}$, keeping the associated losses at a minimum.

To verify this intuitive picture, we carried out numerical simulations for $n = 2$ incoming photons and $h(t) \propto 1 - 4(t/T - 0.5)^2$ ($t \in [0, T]$) using the full propagation equations derived from Eqs. (1-3) [48]. This form of $h(t)$ is motivated by optimal photon storage [43, 52]. The results are shown in Fig. 3(a,b). In Fig. 3(a), the left-bottom quadrant corresponds to both photons being still outside the medium, so $t = 0$ is described by Eqs. (4,6). The top-right quadrant corresponds to both photons being inside the medium, so $t = T$ is described by Eq. (9). Finally the remaining two quadrants correspond to the first photon being already inside the medium while the second photon is still outside. Fig. 3(b) shows a comparison to the analytical prediction from Eqs. (9,11) [53]. While imperfections keep the single-photon conversion efficiency slightly away from unity, the overall physical picture is very well confirmed by our numerical simula-

tions. To verify that losses induced by the finite width of the EIT transparency window – and not the correlated photon dynamics upon pulse entrance – constitute the dominant imperfection, the purple arrow in Fig. 3(b) indicates the efficiency of retrieving Eq. (9) with $n = 2$ back out of the medium.

Arbitrary incoming state.— Since any mixed state can be represented as a classical mixture of pure states, it is sufficient to consider an arbitrary pure input state

$$|\psi\rangle = \sum_n c_n |n\rangle, \quad (13)$$

where $|n\rangle$ is given in Eq. (4). Since $|0\rangle$ and $|1\rangle$ scatter no photons while the scattered photons for $n \geq 2$ destroy the associated coherences, tracing over all photons except for the first one yields

$$\rho = (c_0|0\rangle + c_1|1\rangle)(c_0^*\langle 0| + c_1^*\langle 1|) + \sum_{n \geq 2} |c_n|^2 \rho_1^{(n)}, \quad (14)$$

where $\rho_1^{(n)}$, given in Eq. (11), is the single photon obtained from the Fock state $|n\rangle$. The single-photon conversion efficiency is $1 - |c_0|^2$, i.e. limited by the vacuum component of the input state. The corresponding purity is $\text{tr}[\rho_1^2]/(1 - |c_0|^2)^2$, where

$$\text{tr}[\rho_1^2] = \sum_{m,n \geq 1} |c_m|^2 |c_n|^2 \frac{2mn}{(m+n-1)(m+n)}. \quad (15)$$

For a coherent input with mean photon number $|\alpha|^2$, $|c_i|^2 = e^{-|\alpha|^2} |\alpha|^{2i} / i!$, the efficiency is thus $1 - e^{-|\alpha|^2}$, while the single-photon purity is $(1 - e^{-|\alpha|^2})^{-2} (1 - e^{-2|\alpha|^2} (1 + 2|\alpha|^2)) / 2$, which falls off monotonically from 1 to 1/2 with increasing $|\alpha|^2$. Since $|c_0|^2$ drops exponentially with $|\alpha|^2$, a small average number of incoming photons $|\alpha|^2 \sim 10$ is sufficient to make the single-photon source deterministic. Repeating the above derivations, one obtains for the output density matrix

$$\phi(x, y) = |\alpha|^2 h(-x) h(-y) \exp \left[-|\alpha|^2 \int_{\min(x,y)}^{\infty} h^2(-z) dz \right], \quad (16)$$

which can easily be diagonalized [48]. As for a Fock-state input with Gaussian $h(x)$, the output pulse shortens extremely slowly with increasing $|\alpha|^2$ ($\sim 1/\sqrt{\log|\alpha|}$), as shown in Fig. 2(b).

The efficiency of this single-photon source – imperfect due to the finite width of the EIT window – can be estimated from the analytical form of the density matrix without involving interactions. We assume that the incoming pulse is stored without interactions into the spin-wave $s(z) \propto 1 - 4(z/L - 0.5)^2$ and that the single-spin-wave density matrix [Eq. (11) or Eq. (16)] is retrieved forward. The efficiency η of the single-photon source can then be estimated as the product of these two – storage and retrieval – efficiencies. The $|\alpha|^2$ -dependence of η for

a coherent input is shown in Fig. 3(c) for different blockaded optical depths OD_b , assuming the entire medium is blockaded. The relatively poor scaling of the efficiency with OD_b results from the cusp of the density matrix $\phi(x, y)$ along the diagonal ($x = y$), which carries high-frequency components. In a magneto-optical trap (density $\mathcal{N} \sim 10^{12} \text{ cm}^{-3}$), $\text{OD}_b \sim 10$ [13], and hence $\eta \approx 0.2$ can be achieved. In a BEC [37, 54], $\mathcal{N} \sim 10^{14} \text{ cm}^{-3}$ can give $\text{OD}_b \sim 1000$ and $\eta \approx 0.9$. The efficiency can be further increased by using photonic waveguides [55–58] and by further optimizing $h(t)$ and retrieving backwards [43, 52].

Despite their impurity, the single photons produced with this method are a valuable resource. In particular, the impurity, which can be measured [59] using Hong-Ou-Mandel interference [61], would not interfere with applications that do not rely on this effect, such as optical quantum computing with impurity-insensitive two-qubit gates (e.g. [62]) or the BB84 quantum key distribution protocol [63]. For applications that rely on Hong-Ou-Mandel interference, the photon can be purified in the following ways. First, the detection of the first scattered photon would yield a pure photon. Second, the impure single photon can be purified to the dominant eigenvector $|\phi_1\rangle$ with probability p_1 ($p_1 = 0.69$ for $|\alpha|^2 \gg 1$). This can be accomplished, e.g., using an atomic ensemble in a cavity [64] to store only the mode $|\phi_1\rangle$, heralded by the absence of a click at the cavity output, followed by retrieval into any desired mode [65].

Photon subtraction.— To demonstrate the versatility of the developed theory, we now apply it to the single-photon subtractor [26]. The subtractor requires adding a large single-photon detuning to the level diagram of Fig. 1 and relies on inhomogeneous dephasing of the $|g\rangle$ - $|r\rangle$ coherence to incoherently absorb the photon into state $|r\rangle$. We show that this scheme also yields impure output. The detailed physics of such a setting [48] is complementary to the single-photon source in so far as the density matrix of the remaining photons is obtained by tracing out the first one. Since the timing of the absorption carries information about the remaining photons, the density matrix of the latter is impure. In fact, the single-photon subtractor and the single-photon filter complement each other to make the original pure state. Hence, the impurity and the entire eigenspectrum of the reduced density matrix are identical in the two cases.

This can be shown by tracing over the first photon in Eq. (13) to obtain

$$\rho = |c_0|^2 |0\rangle\langle 0| + \int_{-\infty}^{\infty} dt_1 h^2(t_1) |\psi_{t_1}(t)\rangle\langle \psi_{t_1}(t)|, \quad (17)$$

where

$$|\psi_{t_1}(t)\rangle = \sum_{n \geq 1} c_n \frac{\sqrt{n!}}{(n-1)!} \left[\int_{t_1}^{\infty} dt' h(t') \mathcal{E}^\dagger(t-t') \right]^{n-1} |0\rangle,$$

which has the same eigenspectrum as Eqs. (14).

Outlook.—In conclusion, we extended the dynamics of open quantum systems of Rydberg atoms [66–75] to include the dissipative quantum dynamics of the propagating light field, which is crucial for the understanding of recent experiments [13, 14]. While we focused on the case $z_b > L_p$, the developed framework also applies to the opposite regime [13, 14] and may lead to a quantitative description of the saturation behavior in Ref. [13]. It can also be easily extended to a time-dependent blockade radius, as relevant for photon storage via time-dependent control fields. Extensions to non-dissipative unitary evolution [40], media with longitudinal density variations, incomplete transverse blockade, as well as finite Rydberg-state lifetime are straightforward [13]. Finally, we expect our calculations to be extendable to other light-processing modules, such as the quantum filter [76, 77]. Most importantly, our approach may lead to a simplified effective theory for the many-body dissipative dynamics of correlated photons in strongly interacting media.

We thank M. Lukin, V. Vuletić, O. Firstenberg, T. Peyronel, Q.-Y. Liang, J. Preskill, M. Fleischhauer, J. Otterbach, D. Petrosyan, J. Honer, T. Lee, S. Hofferberth, T. Pfau, P. Grangier, and C. Adams for discussions. This work was supported by NSF, IQIM, the Lee A. DuBridge and Gordon and Betty Moore foundations, CUA and the EU through the Marie Curie ITN COHERENCE and STREP QuILMI.

-
- [1] F. Verstraete, M. M. Wolf, and J. Ignacio Cirac, *Nature Phys.* **5**, 633 (2009).
- [2] F. Pastawski, L. Clemente, and J. I. Cirac, *Phys. Rev. A* **83**, 012304 (2011).
- [3] K. G. H. Vollbrecht, C. A. Muschik, and J. I. Cirac, *Phys. Rev. Lett.* **107**, 120502 (2011).
- [4] H. Krauter, C. A. Muschik, K. Jensen, W. Wasilewski, J. M. Petersen, J. I. Cirac, and E. S. Polzik, *Phys. Rev. Lett.* **107**, 080503 (2011).
- [5] B. Kraus, H. P. Büchler, S. Diehl, A. Kantian, A. Micheli, and P. Zoller, *Phys. Rev. A* **78**, 042307 (2008).
- [6] S. Diehl, A. Micheli, A. Kantian, B. Kraus, H. P. Büchler, and P. Zoller, *Nature Phys.* **4**, 878 (2008).
- [7] S. Diehl, W. Yi, A. J. Daley, and P. Zoller, *Phys. Rev. Lett.* **105**, 227001 (2010).
- [8] H. Weimer, M. Müller, I. Lesanovsky, P. Zoller, and H. P. Büchler, *Nature Phys.* **6**, 382 (2010).
- [9] J. T. Barreiro, M. Müller, P. Schindler, D. Nigg, T. Monz, M. Chwalla, M. Hennrich, C. F. Roos, P. Zoller, and R. Blatt, *Nature* **470**, 486 (2011).
- [10] E. G. D. Torre, J. Otterbach, E. Demler, V. Vuletic, and M. D. Lukin, arxiv:1209.1991.
- [11] S. Diehl, A. Tomadin, A. Micheli, R. Fazio, and P. Zoller, *Phys. Rev. Lett.* **105**, 015702 (2010).
- [12] Y. O. Dudin and A. Kuzmich, *Science* **336**, 887 (2012).
- [13] T. Peyronel, O. Firstenberg, Q.-Y. Liang, S. Hofferberth, A. V. Gorshkov, T. Pohl, M. D. Lukin, and V. Vuletic, *Nature (London)* **488**, 57 (2012).
- [14] D. T. Maxwell, D. J. Szwer, D. P. Barato, H. Busche, J. D. Pritchard, A. Gauguier, K. J. Weatherill, M. P. A. Jones, and C. S. Adams, arxiv:1207.6007 (2012).
- [15] M. Saffman, T. G. Walker, and K. Mølmer, *Rev. Mod. Phys.* **82**, 2313 (2010).
- [16] D. Comparat and P. Pillet, *J. Opt. Soc. Am. B* **27**, A208 (2010).
- [17] T. Pohl, C. S. Adams, and H. R. Sadepour, *J. Phys. B: At. Mol. Opt. Phys.* **44**, 180201 (2011).
- [18] R. Löw, H. Weimer, J. Nipper, J. B. Balewski, B. Butscher, H. P. Büchler, and T. Pfau, *J. Phys. B: At. Mol. Opt. Phys.* **45**, 113001 (2012).
- [19] M. Fleischhauer, A. Imamoglu, and J. P. Marangos, *Rev. Mod. Phys.* **77**, 633 (2005).
- [20] J. D. Pritchard, D. Maxwell, A. Gauguier, K. J. Weatherill, M. P. A. Jones, and C. S. Adams, *Phys. Rev. Lett.* **105**, 193603 (2010).
- [21] A. V. Gorshkov, J. Otterbach, M. Fleischhauer, T. Pohl, and M. D. Lukin, *Phys. Rev. Lett.* **107**, 133602 (2011).
- [22] S. Sevinçli, N. Henkel, C. Ates, and T. Pohl, *Phys. Rev. Lett.* **107**, 153001 (2011).
- [23] D. Petrosyan, J. Otterbach, and M. Fleischhauer, *Phys. Rev. Lett.* **107**, 213601 (2011).
- [24] M. D. Lukin, M. Fleischhauer, R. Cote, L. M. Duan, D. Jaksch, J. I. Cirac, and P. Zoller, *Phys. Rev. Lett.* **87**, 037901 (2001).
- [25] A. E. B. Nielsen and K. Mølmer, *Phys. Rev. A* **81**, 043822 (2010).
- [26] J. Honer, R. Löw, H. Weimer, T. Pfau, and H. P. Büchler, *Phys. Rev. Lett.* **107**, 093601 (2011).
- [27] B. Olmos and I. Lesanovsky, *Phys. Rev. A* **82**, 063404 (2010).
- [28] T. Pohl, E. Demler, and M. D. Lukin, *Phys. Rev. Lett.* **104**, 043002 (2010).
- [29] M. Saffman and T. G. Walker, *Phys. Rev. A* **66**, 065403 (2002).
- [30] L. H. Pedersen and K. Mølmer, *Phys. Rev. A* **79**, 012320 (2009).
- [31] J. D. Pritchard, C. S. Adams, and K. Mølmer, *Phys. Rev. Lett.* **108**, 043601 (2012).
- [32] C. Guerlin, E. Brion, T. Esslinger, and K. Mølmer, *Phys. Rev. A* **82**, 053832 (2010).
- [33] Y. O. Dudin, F. Bariani, and A. Kuzmich, *Phys. Rev. Lett.* **109**, 133602 (2012).
- [34] Y. O. Dudin, L. Li, F. Bariani, and A. Kuzmich, *Nature Phys.* **8**, 790 (2012).
- [35] J. Stanojevic, V. Parigi, E. Bimbard, A. Ourjoumtsev, P. Pillet, and P. Grangier, *Phys. Rev. A* **86**, 021403 (2012).
- [36] F. Bariani, Y. O. Dudin, T. A. B. Kennedy, and A. Kuzmich, *Phys. Rev. Lett.* **108**, 030501 (2012).
- [37] P. Schauß, M. Cheneau, M. Endres, T. Fukuhara, S. Hild, A. Omran, T. Pohl, C. Gross, S. Kuhr, and I. Bloch, *Nature (London)* **491**, 87 (2012).
- [38] F. Bariani, P. M. Goldbart, and T. A. B. Kennedy, *Phys. Rev. A* **86**, 041802 (2012).
- [39] E. Shahmoon, G. Kurizki, M. Fleischhauer, and D. Petrosyan, *Phys. Rev. A* **83**, 033806 (2011).
- [40] V. Parigi, E. Bimbard, J. Stanojevic, A. J. Hilliard, F. Nogrette, R. Tualle-Brouiri, A. Ourjoumtsev, and P. Grangier, *Phys. Rev. Lett.*, in press.
- [41] M. Fleischhauer and M. D. Lukin, *Phys. Rev. Lett.* **84**, 5094 (2000).
- [42] M. Fleischhauer and M. D. Lukin, *Phys. Rev. A* **65**, 022314 (2002).

- [43] A. V. Gorshkov, A. André, M. D. Lukin, and A. S. Sørensen, Phys. Rev. A **76**, 033805 (2007).
- [44] P. D. Drummond, Phys. Rev. A **42**, 6845 (1990).
- [45] P. D. Drummond, *Quantum Theory of Nonlinear Optics*, Lecture Notes for the Nordic Summer School in Nonlinear Optics (1997).
- [46] M. Hillery, Acta Phys. Slovaca **1**, 59 (2009).
- [47] As shown in the supplementary information [48], vacuum Langevin noise \hat{F}_P does not affect our calculations.
- [48] See supplementary information.
- [49] M. Hafezi, D. E. Chang, V. Gritsev, E. Demler, and M. D. Lukin, Phys. Rev. A **85**, 013822 (2012).
- [50] Assuming that each photon goes into a separate mode Q_i gives the same answer.
- [51] H. J. Carmichael, *An Open Systems Approach to Quantum Optics* (Springer, Berlin, 1993).
- [52] A. V. Gorshkov, A. André, M. Fleischhauer, A. S. Sørensen, and M. D. Lukin, Phys. Rev. Lett. **98**, 123601 (2007).
- [53] To repeat the derivation of Eqs. (9,11) for times when $h(t)$ is only partially inside the medium, one takes into account only those photons that have already entered the medium and finds the same equations except the lower limit of integration in Eq. (11) changes from $-\infty$ to $-t$.
- [54] M. Viteau, M. G. Bason, J. Radogostowicz, N. Malossi, D. Ciampini, O. Morsch, and E. Arimondo, Phys. Rev. Lett. **107**, 060402 (2011).
- [55] C. A. Christensen, S. Will, M. Saba, G.-B. Jo, Y.-I. Shin, W. Ketterle, and D. Pritchard, Phys. Rev. A **78**, 033429 (2008).
- [56] M. Bajcsy, S. Hofferberth, V. Balic, T. Peyronel, M. Hafezi, A. S. Zibrov, V. Vuletic, and M. D. Lukin, Phys. Rev. Lett. **102**, 203902 (2009).
- [57] E. Vetsch, D. Reitz, G. Sague, R. Schmidt, S. T. Dawkins, and A. Rauschenbeutel, Phys. Rev. Lett. **104**, 203603 (2010).
- [58] A. Goban, K. S. Choi, D. J. Alton, D. Ding, C. Lacroûte, M. Pototschnig, T. Thiele, N. P. Stern, and H. J. Kimble, Phys. Rev. Lett. **109**, 033603 (2012).
- [59] The Hong-Ou-Mandel coincidence probability for two identical single photons with density matrix ρ_1 is $(1 + \text{tr}[\rho_1^2])/2$ [60].
- [60] R. Loudon, *Quantum Theory of Light* (Oxford University Press, New York, 2000).
- [61] C. K. Hong, Z. Y. Ou, and L. Mandel, Phys. Rev. Lett. **59**, 2044 (1987).
- [62] A. V. Gorshkov, J. Otterbach, E. Demler, M. Fleischhauer, and M. D. Lukin, Phys. Rev. Lett. **105**, 060502 (2010).
- [63] C. H. Bennett and G. Brassard, Proceedings of the IEEE International Conference on Computers, Systems, and Signal Processing, Bangalore, 175 (1984).
- [64] J. Simon, H. Tanji, J. K. Thompson, and V. Vuletić, Phys. Rev. Lett. **98**, 183601 (2007).
- [65] A. V. Gorshkov, A. André, M. D. Lukin, and A. S. Sørensen, Phys. Rev. A **76**, 033804 (2007).
- [66] T. E. Lee, H. Häffner, and M. C. Cross, Phys. Rev. Lett. **108**, 023602 (2012).
- [67] T. E. Lee and M. C. Cross, Phys. Rev. A **85**, 063822 (2012).
- [68] C. Ates, S. Sevinçli, and T. Pohl, Phys. Rev. A **83**, 041802 (2011).
- [69] B. Olmos, W. Li, S. Hofferberth, and I. Lesanovsky, Phys. Rev. A **84**, 041607 (2011).
- [70] M. Hoening, D. Muth, D. Petrosyan, and M. Fleischhauer, arXiv:1208.2911 (2012).
- [71] A. W. Glaetzle, R. Nath, B. Zhao, G. Pupillo, and P. Zoller, Phys. Rev. A **86**, 043403 (2012).
- [72] G. Günter, M. Robert-de-Saint-Vincent, H. Schempp, C. S. Hofmann, S. Whitlock, and M. Weidemüller, Phys. Rev. Lett. **108**, 013002 (2012).
- [73] M. R. de Saint-Vincent, C. S. Hofmann, H. Schempp, G. Günter, S. Whitlock, and M. Weidemüller, arXiv:1209.4728 (2012).
- [74] J. Nipper, J. B. Balewski, A. T. Krupp, S. Hofferberth, R. Löw, and T. Pfau, Phys. Rev. X **2**, 031011 (2012).
- [75] G. Pupillo, A. Micheli, M. Boninsegni, I. Lesanovsky, and P. Zoller, Phys. Rev. Lett. **104**, 223002 (2010).
- [76] A. V. Taichenachev, A. M. Tumaikin, and V. I. Yudin, Europhys. Lett. **72**, 562 (2005).
- [77] A. Taichenachev, V. Yudin, A. Samokotin, A. Akimov, A. Sokolov, V. Sorokin, and N. Kolachevskii, JETP Lett. **88**, 355 (2008).