Supplementary online material to the manuscript: "Dissipative Many-Body Quantum Optics in Rydberg Media"

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I. EQUATIONS OF MOTION FOR TWO INCOMING PHOTONS

In this Section, we give the master-equation-type equations of motion used to do the numerics for two incoming photons and to obtain Fig. 3(a,b) in the main text. While Heisenberg equations of motion [Eqs. (1-3) in the main text] offer a concise description of the system dynamics, an equivalent Shroedinger-picture master equation offers a more straightforward method of solution.

In the case of two incoming photons, the full density matrix

$$\rho(t) = \epsilon(t)|0\rangle\langle 0| + \rho_1(t) + |\psi_2(t)\rangle\langle\psi_2(t)| \qquad \text{(S1)}$$

consists of the unnormalized two-excitation wavefunction

$$\begin{aligned}|\psi_2(t)\rangle = &\frac{1}{2}\int dx \int dy EE(x,y,t)\hat{\mathcal{E}}^\dagger(x)\hat{\mathcal{E}}^\dagger(y)|0\rangle \\ &+ \int dx \int' dy EP(x,y,t)\hat{\mathcal{E}}^\dagger(x)\hat{P}^\dagger(y)|0\rangle \\ &+ \int dx \int' dy ES(x,y,t)\hat{\mathcal{E}}^\dagger(x)\hat{S}^\dagger(y)|0\rangle \\ &+ \frac{1}{2}\int' dx \int' dy PP(x,y,t)\hat{P}^\dagger(x)\hat{P}^\dagger(y)|0\rangle \\ &+ \int' dx \int' dy PS(x,y,t)\hat{P}^\dagger(x)\hat{S}^\dagger(y)|0\rangle \\ &+ \frac{1}{2}\int' dx \int' dy SS(x,y,t)\hat{S}^\dagger(x)\hat{S}^\dagger(y)|0\rangle, \qquad \text{(S2)}\end{aligned}$$

the unnormalized single-excitation density matrix

$$\begin{aligned}\rho_1(t) = &\int dx \int dy\, ee(x,y,t)\hat{\mathcal{E}}^\dagger(y)|0\rangle\langle 0|\hat{\mathcal{E}}(x) \\ &+ \int dx \int' dy\, ep(x,y,t)\hat{P}^\dagger(y)|0\rangle\langle 0|\hat{\mathcal{E}}(x) \\ &+ \int' dx \int dy\, pe(x,y,t)\hat{\mathcal{E}}^\dagger(y)|0\rangle\langle 0|\hat{P}(x) \\ &+ \int dx \int' dy\, es(x,y,t)\hat{S}^\dagger(y)|0\rangle\langle 0|\hat{\mathcal{E}}(x) \\ &+ \int' dx \int dy\, se(x,y,t)\hat{\mathcal{E}}^\dagger(y)|0\rangle\langle 0|\hat{S}(x) \\ &+ \int' dx \int' dy\, pp(x,y,t)\hat{P}^\dagger(y)|0\rangle\langle 0|\hat{P}(x) \\ &+ \int' dx \int' dy\, ps(x,y,t)\hat{S}^\dagger(y)|0\rangle\langle 0|\hat{P}(x) \\ &+ \int' dx \int' dy\, sp(x,y,t)\hat{P}^\dagger(y)|0\rangle\langle 0|\hat{S}(x) \\ &+ \int' dx \int' dy\, ss(x,y,t)\hat{S}^\dagger(y)|0\rangle\langle 0|\hat{S}(x), \qquad \text{(S3)}\end{aligned}$$

and the vacuum component $\epsilon(t)|0\rangle\langle 0|$. Here $\int$ integrates over $(-\infty,\infty)$, while $\int'$ integrates over $[0,L]$. Without loss of generality, we take EE, PP, and SS to be symmetric [e.g. $EE(x,y) = EE(y,x)$]. If the input state had correlations between different Fock states, one would need to include coherences between manifolds of different photon number; the method we discuss can be naturally generalized to these situations.

All terms in $\rho(t=0)$ vanish except for $EE(x,y,0) = \sqrt{2}h(-x)h(-y)$, where we assume $h(t<0) = 0$. The equations of motion for EE, EP, ES, PP, PS, and SS can be obtained by expressing them in terms of $|\Psi_2\rangle$ [e.g. $ES(x,y) = \langle 0|\hat{\mathcal{E}}(x)\hat{S}(y)|\psi_2(t)\rangle$] and using Eqs. (1-3) in the main text. For $x \notin [0,L]$, $y \in [0,L]$, they are

$$(\partial_t + \partial_x + \partial_y)EE = igEP, \qquad \text{(S4)}$$
$$(\partial_t + \partial_x + 1)EP = igEE + i\Omega ES, \qquad \text{(S5)}$$
$$(\partial_t + \partial_x)ES = i\Omega EP, \qquad \text{(S6)}$$

and describe the EIT propagation of photon y, while photon x propagates outside the medium with the speed of light ($c = 1$ in our units). Using $EE(x,y,t) = \sqrt{2}h(t-x)h(t-y)$ to set the boundary conditions at $y = 0$, these equations are solved for $x \le 0$, $y \in [0,L]$ to give the boundary conditions for the equations in the region $x, y \in [0,L]$:

$$(\partial_t + \partial_x + \partial_y)EE = igEP_+, \qquad \text{(S7)}$$
$$(\partial_t + \partial_x + 1)EP = ig(EE + PP) + i\Omega ES, \qquad \text{(S8)}$$
$$(\partial_t + \partial_x)ES = igPS + i\Omega EP, \qquad \text{(S9)}$$
$$(\partial_t + 2)PP = igEP_+ + i\Omega PS_+, \qquad \text{(S10)}$$
$$(\partial_t + 1)PS = igES + i\Omega(PP + SS), \qquad \text{(S11)}$$
$$(\partial_t + iV(r))SS = i\Omega PS_+, \qquad \text{(S12)}$$

where $EP_\pm(x,y) = EP(x,y) \pm EP(y,x)$, $ES_\pm(x,y) = ES(x,y) \pm ES(y,x)$, $PS_\pm(x,y) = PS(x,y) \pm PS(y,x)$, and $r = x - y$. The solution to these equations can then be used to set the boundary conditions at $x = L$ for Eqs.

(S4-S6) in the region $x \geq L$, $y \in [0, L]$, which can in turn be used to calculate the outgoing two-photon pulse.

Now we turn to the evolution equations for the single-excitation density matrix ρ_1. We first note that

$$es(x, y) = \langle \hat{\mathcal{E}}^\dagger(x)\hat{S}(y)\rangle - \int dz EE^*(x, z)ES(z, y) \quad \text{(S13)}$$
$$- \int' dz EP^*(x, z)PS(z, y) - \int' dz ES^*(x, z)SS(z, y).$$

The equation of motion for $\langle \hat{\mathcal{E}}^\dagger(x)\hat{S}(y)\rangle$ follows from Eqs. (1-3) in the main text. Together with the equations of motion for the two-photon amplitudes, this yields the equation of motion for $es(x, y)$, and, similarly, for all matrix elements of ρ_1. The following source terms will describe the transfer of population from $|\psi_2\rangle$ to ρ_1:

$$f_{ee}(x, y) = 2\int' dz EP^*(x, z)EP(y, z), \quad \text{(S14)}$$

$$f_{ep}(x, y) = 2\int' dz EP^*(x, z)PP(y, z), \quad \text{(S15)}$$

$$f_{es}(x, y) = 2\int' dz EP^*(x, z)PS(z, y), \quad \text{(S16)}$$

$$f_{pp}(x, y) = 2\int' dz PP^*(x, z)PP(y, z), \quad \text{(S17)}$$

$$f_{ps}(x, y) = 2\int' dz PP^*(x, z)PS(z, y), \quad \text{(S18)}$$

$$f_{ss}(x, y) = 2\int' dz PS^*(z, x)PS(z, y). \quad \text{(S19)}$$

As expected, in the interaction-free case ($V = 0$) and assuming perfect EIT, the source terms vanish because $|e\rangle$ is never populated, so all components of $|\psi_2\rangle$ involving P vanish. With these definitions, for $x, y \in [0, L]$,

$$(\partial_t + \partial_x + \partial_y)ee = ig(ep - pe) + f_{ee}, \quad \text{(S20)}$$
$$(\partial_t + \partial_x + 1)ep = ig(ee - pp) + i\Omega es + f_{ep}, \quad \text{(S21)}$$
$$(\partial_t + \partial_x)es = i\Omega ep - ig\, ps + f_{es}, \quad \text{(S22)}$$
$$(\partial_t + 2)pp = ig(pe - ep) + i\Omega(ps - sp) + f_{pp}, \text{(S23)}$$
$$(\partial_t + 1)ps = -ig\, es + i\Omega(pp - ss) + f_{ps}, \quad \text{(S24)}$$
$$\partial_t ss = i\Omega(sp - ps) + f_{ss}, \quad \text{(S25)}$$

while $pe(x, y) = ep^*(y, x)$, $se(x, y) = es^*(y, x)$, and $sp(x, y) = ps^*(y, x)$. Equations of motion outside of $x, y \in [0, L]$ can be obtained in the same way.

We do numerical calculations in the regime of good EIT ($\lesssim 1\%$ single-photon loss). Thus, to a good approximation, photon scattering occurs only when both photons are inside the medium. We therefore solve Eqs. (S20-S25) with vanishing initial and boundary conditions.

We note that the equations can easily be extended [S1] to include longitudinally varying density, finite decoherence rate of S, as well as cases where the blockade radius is smaller than the transverse extent of the probe beam.

We also note that, as in Refs. [S2, S3, S4, S5], the vacuum Langevin noise operator $\hat{F}_P(z, t)$ in Eq. (2) does not play a role in the above derivation. This operator can be thought of as describing the effect of the mode $\hat{Q}$ [see Eq. (7) in the main text], which photons scatter into. Using the generalized Einstein relations, $F_P(z, t)$ can be shown to have only the following nonzero correlation: $\langle \hat{F}_P(z, t)\hat{F}_P^\dagger(z', t')\rangle = 2\delta(z - z')\delta(t - t')$ [S6, S2, S3, S4, S5]. Using this, one immediately sees that $F_P(z, t)$ does not contribute to Eqs. (S4-S12) since terms of the form $\langle 0|\hat{\mathcal{E}}(x, t)\hat{F}_P(y, t)|\psi_2(0)\rangle$ vanish. Similarly, since Eq. (S13) and the corresponding equations for the other elements of ρ_1 involve only normally ordered expectation values, Eqs. (S20-S25) are also unaffected by $\hat{F}_P(z, t)$. It is not surprising that we are able to derive the master equation from the Heisenberg equations without having to rely on vacuum Langevin noise operator $\hat{F}_P(z, t)$: indeed, the effects of the latter are completely determined by the decay term in Eq. (2) of the main text and by the generalized Einstein relations [S6].

II. IDEAL SINGLE-PHOTON GENERATION FROM 2 PHOTONS

In this Section, in the case of an input Fock state with $n = 2$, we show how Eqs. (9,11) in the main text arise from the full equations of motion presented above. This Section and Section III also justify the use of the simplified framework (that of tracing over all photons but the first one) employed in the main text.

Let's assume that EIT is perfect and that $L_p < L < z_b$. Then, for $x \leq 0$ and $y \in [0, L]$, Eqs. (S4-S6) give

$$ES(x, y, t) = -\sqrt{2/v_g}\, h(t - x)h(t - y/v_g), \quad \text{(S26)}$$

where $v_g = (\Omega/g)^2$ in our units. From Eqs. (S9,S11), we obtain $\partial_t ES \approx -\partial_x ES - g^2 ES$, so that, for $x, y \in [0, L]$,

$$PS(x, y, t) \approx igES(x, y, t) \approx igES(x = 0, y, t - x)e^{-g^2 x}$$
$$\approx -ig\sqrt{2/v_g}\, h(t)h(t - y/v_g)e^{-g^2 x}, \quad \text{(S27)}$$

which describes the absorption of the two-excitation amplitude over the absorption length $1/g^2 \ll L_p$. Inserting this expression into Eq. (S19), we obtain

$$f_{ss}(x, y, t) = 2h^2(t)h(t - x/v_g)h(t - y/v_g)/v_g. \text{(S28)}$$

Then from Eqs. (S22,S24,S25), to a good approximation,

$$es = -(\Omega/g)ss + (\Omega/g^3)\partial_x ss, \quad \text{(S29)}$$
$$\partial_t ss = -2\Omega^2 ss - \Omega g(se + es) + f_{ss}. \quad \text{(S30)}$$

Inserting Eq. (S29) into Eq. (S30), we obtain

$$\partial_t ss = -v_g \partial_R ss + f_{ss}(x, y, t), \quad \text{(S31)}$$

which describes how the source f_{ss} puts excitations into ss; and, as soon as the excitations are put in, they start moving at v_g along $R = (x+y)/2$. This equation assumes the wavepacket's frequency components all fit inside the EIT transparency window. Solving this equation gives

$$ss(x,y,t) = \frac{2}{v_g} h(t-\frac{x}{v_g}) h(t-\frac{y}{v_g}) \int_{t-\frac{\min(x,y)}{v_g}}^{t} dt' h^2(t'), \quad \text{(S32)}$$

which, for $n = 2$, generalizes Eqs. (9,11) in the main text to cases when the pulse has only partially entered the medium. Eq. (S32) yields $\mathrm{Tr}[\rho_1^2]/\mathrm{Tr}[\rho_1]^2 = 2/3$ for all t and $\mathrm{Tr}[\rho_1] = \left[\int_{-\infty}^{t} d\tau h^2(\tau)\right]^2$, which give the dashed lines in Fig. 3(b) in the main text. To a good approximation, ρ_1 satisfies the dark-state-polariton condition $ee = -\sqrt{v_g} es = v_g ss$. This derivation can easily be extended to include the effect of a finite EIT transparency window width, which partially explains the slight discrepancy between analytics and numerics in Fig. 3(b) in the main text.

III. IDEAL SINGLE-PHOTON GENERATION FROM n PHOTONS

In this Section, we generalize Sec. II to arbitrary n.

Let $\mathbf{x}_m \equiv x_1, \dots, x_m$, $E_m \equiv E \dots E$ (where E is repeated m times to denote the m-photon wavefunction), and $h(t - \mathbf{x}_m) \equiv \prod_{i=1}^{m} h(t - x_i)$. Then, for $\mathbf{x}_n < 0$, the incoming n-photon state is given by

$$E_n(\mathbf{x}_n) = \sqrt{n!} h(t - \mathbf{x}_n). \quad \text{(S33)}$$

Once the first two photons enter the medium ($\mathbf{x}_{n-2} < 0$ and $x_{n-1}, x_n > 0$), we have, in analogy with Eq. (S27),

$$\begin{aligned} &E_{n-2}PS(\mathbf{x}_n) \quad \text{(S34)} \\ &= -ig\sqrt{n!/v_g} h(t - \mathbf{x}_{n-2}) h(t) h(t - x_n/v_g) e^{-g^2 x_{n-1}}. \end{aligned}$$

So, by analogy with Eqs. (S19,S28),

$$\begin{aligned} &f_{e_{n-2}se_{n-2}s}(\mathbf{x}_{n-1}, \mathbf{x}'_{n-1}) \quad \text{(S35)} \\ &= 2\int_{}^{t} dz E_{n-2}PS^*(\mathbf{x}_{n-2}, z, x_{n-1}) E_{n-2}PS(\mathbf{x}'_{n-2}, z, x'_{n-1}) \\ &= \frac{n!}{v_g} h(t-\mathbf{x}_{n-2}) h(t-\mathbf{x}'_{n-2}) h^2(t) h(t-\frac{x_{n-1}}{v_g}) h(t-\frac{x'_{n-1}}{v_g}). \end{aligned}$$

Applying group velocity propagation along $(x_{n-1} + x'_{n-1})/2$ [as in Eq. (S31)], we have [as in Eq. (S32)]

$$\begin{aligned} e_{n-2}se_{n-2}s(\mathbf{x}_{n-1}, \mathbf{x}'_{n-1}) &= \frac{n!}{v_g} h(t - \mathbf{x}_{n-2}) h(t - \mathbf{x}'_{n-2}) \\ \times h(t-\frac{x_{n-1}}{v_g}) h(t-\frac{x'_{n-1}}{v_g}) &\int_{t-\frac{\min(x_{n-1},x'_{n-1})}{v_g}}^{t} dt' h^2(t'). \quad \text{(S36)} \end{aligned}$$

Allowing now the third photon to enter the medium ($x_{n-2}, x'_{n-2} > 0$), we have

$$\begin{aligned} &f_{e_{n-3}se_{n-3}s}(\mathbf{x}_{n-2}, \mathbf{x}'_{n-2}) \quad \text{(S37)} \\ &= e_{n-2}se_{n-2}s(\mathbf{x}_{n-3}, 0, x_{n-2}, \mathbf{x}'_{n-3}, 0, x'_{n-2}) \\ &= \frac{n!}{v_g} h(t - \mathbf{x}_{n-3}) h(t - \mathbf{x}'_{n-3}) h^2(t) \\ &\times h(t - \frac{x_{n-2}}{v_g}) h(t - \frac{x'_{n-2}}{v_g}) \int_{t-\frac{\min(x_{n-2},x'_{n-2})}{v_g}}^{t} dt' h^2(t'). \end{aligned}$$

Applying group velocity propagation along $(x_{n-2} + x'_{n-2})/2$, we have

$$\begin{aligned} &e_{n-3}se_{n-3}s(\mathbf{x}_{n-2}, \mathbf{x}'_{n-2}) \\ &= \frac{n!}{v_g} h(t - \mathbf{x}_{n-3}) h(t - \mathbf{x}'_{n-3}) h(t - \frac{x_{n-2}}{v_g}) h(t - \frac{x'_{n-2}}{v_g}) \\ &\times \frac{1}{2} \left[\int_{t-\frac{\min(x_{n-2},x'_{n-2})}{v_g}}^{t} dt' h^2(t')\right]^2. \quad \text{(S38)} \end{aligned}$$

Allowing the fourth photon to enter the medium, we have

$$\begin{aligned} &f_{e_{n-4}se_{n-4}s}(\mathbf{x}_{n-3}, \mathbf{x}'_{n-3}) \quad \text{(S39)} \\ &= e_{n-3}se_{n-3}s(\mathbf{x}_{n-4}, 0, x_{n-3}, \mathbf{x}'_{n-4}, 0, x'_{n-3}) \\ &= \frac{n!}{v_g} h(t - \mathbf{x}_{n-4}) h(t - \mathbf{x}'_{n-4}) h^2(t) \\ &\times h(t-\frac{x_{n-3}}{v_g}) h(t-\frac{x'_{n-3}}{v_g}) \frac{1}{2}\left[\int_{t-\frac{\min(x_{n-3},x'_{n-3})}{v_g}}^{t} dt' h^2(t')\right]^2. \end{aligned}$$

Applying group velocity propagation along $(x_{n-3} + x'_{n-3})/2$, we have

$$\begin{aligned} &e_{n-4}se_{n-4}s(\mathbf{x}_{n-3}, \mathbf{x}'_{n-3}) \\ &= \frac{n!}{v_g} h(t - x_1) \dots h(t - x'_{n-4}) h(t - \frac{x_{n-3}}{v_g}) h(t - \frac{x'_{n-3}}{v_g}) \\ &\times \frac{1}{3!} \left[\int_{t-\frac{\min(x_{n-3},x'_{n-3})}{v_g}}^{t} dt' h^2(t')\right]^3. \quad \text{(S40)} \end{aligned}$$

We continue in this way until we reach

$$\begin{aligned} &ss(x_1, x'_1) = \\ &= \frac{n}{v_g} h(t - \frac{x_1}{v_g}) h(t - \frac{x'_1}{v_g}) \left[\int_{t-\frac{\min(x_1,x'_1)}{v_g}}^{t} dt' h^2(t')\right]^{n-1}, \quad \text{(S41)} \end{aligned}$$

which generalizes Eqs. (9,11) in the main text to cases when the pulse has only partially entered the medium.

IV. EIGENVECTORS OF THE SINGLE-PHOTON DENSITY MATRIX

In this Section, we study the eigenvectors and eigenvalues of the single-photon density matrix, Eqs. (11) and (16) in the main text, obtained via single-photon filtering from Fock-state and coherent-state inputs, respectively.

We first study the eigenvectors ϕ_i and eigenvalues p_i of the single-photon density matrix $\phi(x,y)$ given in Eq. (11) in the main text. Defining $\tilde{x}=\int_{-\infty}^{x} dz h^2(-z)$, we obtain $\rho=\int_0^1 d\tilde{x}d\tilde{y}\tilde{\phi}(\tilde{x},\tilde{y})\hat{\tilde{\mathcal{E}}}^\dagger(\tilde{y})|0\rangle\langle 0|\hat{\tilde{\mathcal{E}}}(\tilde{x})$, where $\tilde{\phi}(\tilde{x},\tilde{y})=n\left[\min(\tilde{x},\tilde{y})\right]^{n-1}$, $\hat{\tilde{\mathcal{E}}}(\tilde{x})=\hat{\mathcal{E}}(x)/h(-x)$, $[\hat{\tilde{\mathcal{E}}}(\tilde{x}),\hat{\tilde{\mathcal{E}}}^\dagger(\tilde{y})]=\delta(\tilde{x}-\tilde{y})$. The eigenvalues p_i are then the solutions of the characteristic equation $J_{-1/n}\left[2\sqrt{(n-1)/(np)}\right]=0$. In particular, in the limit $n\to\infty$, p_i are the roots of $J_0[2/\sqrt{p}]=0$. The eigenvectors of $\tilde{\phi}(\tilde{x},\tilde{y})$ are $\tilde{\phi}_i(\tilde{x})\propto\tilde{x}^{(n-1)/2}J_{1-1/n}\left[2\sqrt{(n-1)/(np_i)}\tilde{x}^{n/2}\right]$. In particular, for $n=2$, $p_i=2\pi^{-2}\left(i-\frac{1}{2}\right)^{-2}$, $\tilde{\phi}_i(\tilde{x})=\sqrt{2}\sin\left[\pi\left(i-\frac{1}{2}\right)\tilde{x}\right]$. While $\tilde{\phi}(\tilde{x},\tilde{x})$ and $\tilde{\phi}_i(\tilde{x})$ shorten as $1/n$ with increasing n, $\phi(x,x)$ and $\phi_i(x)=h(-x)\tilde{\phi}_i(\tilde{x})$ shorten much slower as $1/\sqrt{\log n}$ for a Gaussian $h(x)$.

We now study the eigenvectors of the single-photon density matrix $\phi(x,y)$ given in Eq. (16) in the main text. Following the same change of variables, we obtain $\tilde{\phi}(\tilde{x},\tilde{y})=|\alpha|^2\exp\left[-|\alpha|^2(1-\min(\tilde{x},\tilde{y}))\right]$, which, for $|\alpha|^2\gg 1$, agrees with the above $\phi(\tilde{x},\tilde{y})$ provided one identifies n with $|\alpha|^2$. For general $|\alpha|^2$, the eigenstates $\tilde{\phi}_i$ of $\tilde{\phi}(\tilde{x},\tilde{y})$ are linear combinations of $e^{-|\alpha|^2(1-\tilde{x})/2}J_1[2e^{-|\alpha|^2(1-\tilde{x})/2}/\sqrt{p_i}]$ and $e^{-|\alpha|^2(1-\tilde{x})/2}Y_1[2e^{-|\alpha|^2(1-\tilde{x})/2}/\sqrt{p_i}]$, where p_i are the eigenvalues.

V. SINGLE-PHOTON SUBTRACTION

In this Section, we present a formal derivation of Eq. (17) in the main text, which describes the output of a single-photon subtractor [S7] and demonstrates the necessity of going beyond the single-mode treatment of Ref. [S7]. In addition to verifying Eq. (17), this method allows one to treat deviations from the ideal result.

Following Ref. [S7], the single-photon subtractor is constructed by introducing a large single-photon detuning to the level diagram in Fig. 1 of the main text. The atoms can then be in one of two collective states $|G\rangle$ (all atoms in the ground state $|g\rangle$) and $|E\rangle$ (a single atom in a Rydberg state $|r\rangle$). Inhomogeneous dephasing of the $|g\rangle$-$|r\rangle$ coherence drives the process (with rate Γ), in which a single photon is absorbed into state $|r\rangle$, thus, transferring the atoms from state $|G\rangle$ to state $|E\rangle$. After this irreversible process takes places, all Rydberg states $|r\rangle$ can be ignored since they are strongly shifted by the single atom in state $|r\rangle$. Therefore, the remaining photons propagate unhindered through the far-off-resonant two-level $|g\rangle$-$|e\rangle$ medium. This situation is described by the following master equation for the density matrix:

$$\dot{\rho}=-i[\hat{H}_0,\rho]+\Gamma\int_0^\infty dx\Big[2\hat{\mathcal{E}}(x)|E\rangle\langle G|\rho|G\rangle\langle E|\hat{\mathcal{E}}^\dagger(x) -\hat{\mathcal{E}}^\dagger(x)\hat{\mathcal{E}}(x)|G\rangle\langle G|\rho-\rho|G\rangle\langle G|\hat{\mathcal{E}}^\dagger(x)\hat{\mathcal{E}}(x)\Big]. \quad \text{(S42)}$$

$\hat{H}_0$ here describes simple propagation of light in vacuum. The photon is subtracted within a few absorption lengths Γ^{-1} of $x=0$, so the remainder of the medium plays no role provided $z_b>L$; hence we assumed $L\to\infty$.

Here, for simplicity, we only present the derivation for two incoming photons $|2\rangle$. Generalization to an arbitrary incoming state $|\psi\rangle=\sum_n c_n|n\rangle$ is straightforward.

Therefore, the full density matrix

$$\rho=\rho_1+|\psi_2\rangle\langle\psi_2| \quad \text{(S43)}$$

consists of the two-photon wavefunction

$$|\psi_2\rangle=\frac{1}{2}\int dxdyEE(x,y)\hat{\mathcal{E}}^\dagger(x)\hat{\mathcal{E}}^\dagger(y)|0\rangle|G\rangle \quad \text{(S44)}$$

and of the single-photon density matrix

$$\rho_1=\int dxdyee(x,y)\hat{\mathcal{E}}^\dagger(y)|0\rangle|E\rangle\langle E|\langle 0|\hat{\mathcal{E}}(x). \quad \text{(S45)}$$

One then finds the following equations of motion:

$$\partial_t EE(x,y)=-\partial_x EE-\partial_y EE-\Gamma[H(x)+H(y)]EE,$$
$$\partial_t ee(x,y)=-\partial_x ee-\partial_y ee+2\Gamma\int_0^\infty dzEE(y,z)EE^*(x,z),$$

where $H(x)$ is the Heaviside step function.

Starting with the boundary conditions $EE(x,y,t)=\sqrt{2}h(t-x)h(t-y)$ for $x,y\leq 0$, we solve for EE:

$$EE(x,y,t)=\sqrt{2}h(t-x)h(t-y)e^{-\Gamma[H(x)x+H(y)y]}. \quad \text{(S46)}$$

Inserting this into the equation of motion for ee and using the fact that the absorption length is much shorter than the (now uncompressed) pulse duration, we obtain

$$\partial_t ee(x,y,t)=-\partial_x ee-\partial_y ee +2h^2(t)h(t-x)h(t-y)[1-H(x)][1-H(y)]. \quad \text{(S47)}$$

For $x,y<0$, this can be integrated to give

$$ee(x,y,t)=2h(t-x)h(t-y)\int_{-\infty}^{t}h^2(t'), \quad \text{(S48)}$$

so that, in the remaining three quadrants of the xy plane,

$$ee(x,y,t)=2h(t-x)h(t-y)\int_{-\infty}^{t-\max(x,y)}dt'h^2(t'), \quad \text{(S49)}$$

which is a special case of Eq. (17) in the main text.

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