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Excitation of single-photon embedded eigenstates in coupled cavity-atom systems: supplementary material

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A. Derivation of the EE condition for the atom-cavity system

In the main text, we considered the full Hamiltonian of the atom-cavity-waveguide system,

$$\hat{H} = \hat{H}_{AC} + \int_{-\infty}^{+\infty} dk\ \omega_k \hat{c}_k^\dagger \hat{c}_k + \int_{-\infty}^{+\infty} dk \left[\hat{c}_k^\dagger (V_C \hat{a} + V_A \hat{\sigma}_-) + h.c.\right] \quad \textbf{(S1)}$$

where $\hat{H}_{AC} = \omega_c \hat{a}^\dagger \hat{a} + \omega_A \hat{\sigma}_+ \hat{\sigma}_- + J\left(\hat{a}\hat{\sigma}_+ + h.c.\right)$, and we showed that a single-excitation state $|\Psi_{EE}^{(1)}\rangle = C^{(1)}|1,g\rangle + A^{(1)}|0,e\rangle$ is an embedded eigenstate (EE) if $V_C C^{(1)} + V_A A^{(1)} = 0$. In this section we show that this latter condition is equivalent to the EE condition in eq. 1 of the main text (with the parameters relabeled accordingly). In the one-excitation sector, we can rewrite the time-independent Schroedinger equation $\hat{H}_{AC}|\Psi_{EE}^{(1)}\rangle = \omega_{EE}^{(1)}|\Psi_{EE}^{(1)}\rangle$ as

$$\begin{pmatrix} \omega_A & J \\ J & \omega_C \end{pmatrix}\begin{pmatrix} A^{(1)} \\ C^{(1)} \end{pmatrix} = \omega_{EE}^{(1)}\begin{pmatrix} A^{(1)} \\ C^{(1)} \end{pmatrix}. \quad \textbf{(S2)}$$

Replacing $C^{(1)} = -V_A / V_C A^{(1)}$ we obtain two equations for $\omega_{EE}^{(1)}$, and by requiring that they both hold we obtain

$$\frac{\omega_A V_c - J V_A}{V_C} = \frac{\omega_C V_A - J V_C}{V_C} \Rightarrow J(V_A^2 - V_C^2) = (\omega_A - \omega_C) V_A V_C \quad \textbf{(S2)}$$

B. Derivation of the Input-Output formalism for the atom-cavity Hamiltonian

In this section we derive the input-output formalism for the system considered in the main text. The derivation follows closely the one outlined in ref. [1]. We start by considering the Hamiltonian in eq. (S1). We can split the waveguide modes into right- and left-propagating modes

$$\int_{-\infty}^{+\infty} dk\ \omega_k \hat{c}_k^\dagger \hat{c}_k = \int_0^{+\infty} dk\ \omega_k \hat{c}_k^\dagger \hat{c}_k + \int_{-\infty}^0 dk\ \omega_k \hat{c}_k^\dagger \hat{c}_k \quad \textbf{(S3)}$$

Next, we assume that the waveguide dispersion relation can be linearized in a narrow frequency range centered on a frequency of interest ω_0. This allows us to write $\omega_k = \omega_0 + v_g(k - k_0)$ for $k > 0$, and $\omega_k = \omega_0 - v_g(k + k_0)$ for $k < 0$, where $\pm k_0$ is the wavevector at ω_0 and v_g is the group velocity. We define $\omega \equiv v_g(k \mp k_0)$ for right- and left-propagation modes respectively. By introducing the operators $\hat{c}_{R,\omega} \equiv \hat{c}_{k+k_0} / \sqrt{v_g}$, $\hat{c}_{L,\omega} \equiv \hat{c}_{k-k_0} / \sqrt{v_g}$, and extending the limits of integration to $\pm\infty$, the free evolution Hamiltonian of the waveguide modes becomes

$$H_{WG} = \int_{-\infty}^{+\infty} d\omega\ \omega\, (\hat{c}_{R,\omega}^\dagger \hat{c}_{R,\omega} - \hat{c}_{L,\omega}^\dagger \hat{c}_{L,\omega}) \quad \textbf{(S4)}$$

where the frequency ω is now defined with respect to ω_0. By substituting the new operators in the full Hamiltonian in eq. (S1) we get

$$\hat{H}=\hat{H}_{AC}+\int_{-\infty}^{+\infty} d\omega\,\omega\,(\hat{c}^{\dagger}_{R,\omega}\hat{c}_{R,\omega}-\hat{c}^{\dagger}_{L,\omega}\hat{c}_{L,\omega})+ \frac{1}{\sqrt{v_g}}\int_{-\infty}^{+\infty} d\omega\left[(\hat{c}^{\dagger}_{R,\omega}+\hat{c}^{\dagger}_{L,\omega})(V_C\hat{a}+V_A\hat{\sigma}_-)+h.c.\right] \quad \textbf{(S5)}$$

where we also subtracted the frequency ω_0 in the atom-cavity Hamiltonian, $\hat{H}_{AC}=(\omega_c-\omega_0)\hat{a}^{\dagger}\hat{a}+(\omega_A-\omega_0)\hat{\sigma}_+\hat{\sigma}_-+J\left(\hat{a}\hat{\sigma}_++h.c.\right)$. From the Hamiltonian in eq. (S5) we can derive the Heisenberg equations of motion of the different operators,

$$\dot{\hat{c}}_{R,\omega}=-i\omega\hat{c}_{R,\omega}-i\frac{1}{\sqrt{v_g}}\left(V_A\hat{\sigma}_-+V_C\hat{a}\right) \quad \textbf{(S6)}$$

$$\dot{\hat{c}}_{L,\omega}=i\omega\hat{c}_{L,\omega}-i\frac{1}{\sqrt{v_g}}\left(V_A\hat{\sigma}_-+V_C\hat{a}\right) \quad \textbf{(S7)}$$

$$\dot{\hat{\sigma}}_-=-i(\omega_A-\omega_0)\hat{\sigma}_-+iJ\hat{a}\hat{\sigma}_z+i\frac{V_A^*}{\sqrt{v_g}}\hat{\sigma}_z\int d\omega(\hat{c}_{R,\omega}+\hat{c}_{L,\omega}) \quad \textbf{(S8)}$$

$$\dot{\hat{a}}=-i(\omega_C-\omega_0)\hat{a}-iJ\hat{\sigma}_--i\frac{V_C^*}{\sqrt{v_g}}\int d\omega(\hat{c}_{R,\omega}+\hat{c}_{L,\omega}) \quad \textbf{(S9)}$$

We integrate eqs. (S6) and (S7),

$$\hat{c}_{R/L,\omega}(t)=\hat{c}_{R/L,\omega}(t_0)e^{\mp i\omega(t-t_0)}- -\frac{i}{\sqrt{v_g}}\int_{t_0}^{t}dt'\,e^{\mp i\omega(t-t')}[V_A\hat{\sigma}_-(t')+V_C\hat{a}(t')] \quad \textbf{(S10)}$$

and substitute them into eqs. (S8) and (S9), obtaining

$$\dot{\hat{\sigma}}_-=-i\left(\omega_A-\omega_0-i2\pi\frac{|V_A|^2}{v_g}\right)\hat{\sigma}_-+ +i\left(J-i2\pi\frac{V_A^*V_C}{v_g}\right)\hat{a}\hat{\sigma}_z+i\sqrt{\frac{2\pi}{v_g}}V_A^*\hat{\sigma}_z[\hat{c}_{R,IN}(t)+\hat{c}_{L,IN}(t)] \quad \textbf{(S11)}$$

$$\dot{\hat{a}}=-i\left(\omega_C-\omega_0-i2\pi\frac{|V_C|^2}{v_g}\right)\hat{a}- -i\left(J-i2\pi\frac{V_C^*V_A}{v_g}\right)\hat{\sigma}_--i\sqrt{\frac{2\pi}{v_g}}V_C^*[\hat{c}_{R,IN}(t)+\hat{c}_{L,IN}(t)] \quad \textbf{(S12)}$$

where we have reversed the order of integration and we have used $\int d\omega\exp[\pm i\omega(t-t')]=2\pi\delta(t-t')$. Following a well-known procedure [1], we have defined the input fields as

$$\hat{c}_{R/L,IN}(t)=\frac{1}{\sqrt{2\pi}}\int d\omega\,\hat{c}_{R/L,\omega}(t_0)e^{\mp i\omega(t-t_0)} \quad \textbf{(S13)}$$

where t_0 is an arbitrary initial time. Equations (S11) and (S12) show explicitly that the presence of the waveguide, besides providing an input channel for the system, leads to finite decay rates for the atom and cavity amplitudes, $\Gamma_{A,C}=2\pi|V_{A/C}|^2/v_g$, and to a modification of the bare coupling rate J.

C. Eigenstates of the atom-cavity system in the one- and two-excitation sector

Equations (S11)-(S12) allows us to calculate the eigenstates and complex eigenvalues of the atom-cavity system in absence of external pumping. Indeed, the obtained equations of motion are equivalent, for a single excitation, to a non-Hermitian effective Hamiltonian

$$\hat{H}_{AC}^{(eff)}=(\omega_c-i\Gamma_C)\hat{a}^{\dagger}\hat{a}+(\omega_A-i\Gamma_A)\hat{\sigma}_+\hat{\sigma}_-+(J-i\sqrt{\Gamma_C\Gamma_A})\left(\hat{a}\hat{\sigma}_++h.c.\right) \quad \textbf{(S14)}$$

where we assumed the couplings $V_{A/C}$ to be real, and we have omitted the common frequency ω_0. Therefore, we can find the eigenstates and eigenvalues in the one-excitation subspace by diagonalizing the matrix

$$\begin{pmatrix}\omega_A-i\Gamma_A & J-i\sqrt{\Gamma_C\Gamma_A}\\ J-i\sqrt{\Gamma_C\Gamma_A} & \omega_C-i\Gamma_C\end{pmatrix} \quad \textbf{(S15)}$$

with the additional requirement given by eq. (S2). A direct calculation gives the frequencies of the EE and of the bright mode

$$\omega_{EE}^{(1)}=\omega_C-J\frac{V_C}{V_A}=\omega_A-J\frac{V_A}{V_C}\ , \quad \textbf{(S16)}$$

$$\tilde{\omega}_B^{(1)}=\omega_A+J\frac{V_C}{V_A}-i\pi\frac{V_A^2+V_C^2}{v_g}=\omega_C+J\frac{V_A}{V_C}-i\pi\frac{V_A^2+V_C^2}{v_g} \quad \textbf{(S17)}$$

We note that, according to the sign of $J\,V_C/V_A$, the EE will be the lower ($JV_C/V_A>0$) or upper ($J\,V_C/V_A<0$) resonance, in agreement with the original discussion of Friedrich and Wintgen [2]. In the following, and in the main text, we indicate the complex frequency of the single-photon bright mode as $\tilde{\omega}_B^{(1)}\equiv\omega_B^{(1)}-i\Gamma$. The corresponding eigenstates are $|\Psi_{EE}^{(1)}\rangle=N\,(V_C\hat{\sigma}_+-V_A\hat{a}^{\dagger})|0\rangle$ and $|\Psi_B^{(1)}\rangle=N\,(V_A\hat{\sigma}_++V_C\hat{a}^{\dagger})|0\rangle$, with the normalization constant $N=1/\sqrt{V_A^2+V_C^2}$.

Similarly, within the validity of the effective Hamiltonian in eq. (S15) we can look for the eigenstates of the system in the two-excitation sector, where a general state can be written as $|\Psi^{(2)}\rangle=\left[E_{2C}(\hat{a}^{\dagger 2}/\sqrt{2})+E_{AC}\hat{a}^{\dagger}\hat{\sigma}_+\right]|0\rangle$. The eigenvalues and eigenstates are therefore obtained by diagonalizing the matrix

$$\begin{pmatrix}\omega_A+\omega_C-i(\Gamma_A+\Gamma_C) & \sqrt{2}\left[J-i\sqrt{\Gamma_C\Gamma_A}\right]\\ \sqrt{2}\left[J-i\sqrt{\Gamma_C\Gamma_A}\right] & 2(\omega_C-i\Gamma_C)\end{pmatrix} \quad \textbf{(S18)}$$

The eigenvalues and eigenstates do not have a simple form in this case, and they are not reported here. We note that both eigenvalues $\tilde{\omega}_{1/2}^{(2)}=\omega_{1/2}^{(2)}-i\Gamma_{1/2}^{(2)}$ have a nonzero imaginary component (as expected since no EE exists in the two-excitation sector), and their ratio reads

$$\frac{\Gamma_1^{(2)}}{\Gamma_2^{(2)}}=\frac{5}{4}+\frac{3}{2\alpha^2}+\frac{1}{4\alpha^4}+\frac{1+3\alpha^2}{4\alpha^4}\sqrt{\alpha^4+6\alpha^2+1}, \quad \textbf{(S19)}$$

where we have set $\alpha=V_C/V_A$. We note that for $\alpha=O(1)$ the two decay rates are quite different. For example, for $\alpha=1$, $\Gamma_1^{(2)}/\Gamma_2^{(2)}\approx 5.8$, and for $\alpha = 0.5$ (the value used in figs. 1d, 2(a,b,d), 3 and 4a of the main text), $\Gamma_1^{(2)} / \Gamma_2^{(2)} \approx 22$. Therefore, also in the two-excitation sector we can identify a "bright" and a "dark" eigenstate, whose real frequencies are denoted $\omega_B^{(2)}$ and $\omega_D^{(2)}$, respectively, in fig. 2d of the main text.

D. Existence of embedded eigenstates when atom and cavity are not co-located

When cavity and atom are not co-located, it is necessary to take in account a phase-delay when light propagates between them. We assume that the atom is located at $x = 0$, and the cavity is at $x = d$. The last term of the Hamiltonian in eq. (S1) is modified into

$$\hat{H}_k = \int_{-\infty}^{+\infty} dk \left[V_C (e^{-ikd} \hat{c}_k^\dagger \hat{a} + e^{ikd} \hat{c}_k \hat{a}^\dagger) + V_A (\hat{c}_k^\dagger \hat{\sigma}_- + \hat{c}_k \hat{\sigma}_+) \right] \tag{S20}$$

By repeating the same analysis of sec. 2 we obtain the modified equations of motion for the atom and cavity operators,

$$\dot{\hat{\sigma}}_-(t) = -i\left(\omega_A - \omega_0 - i2\pi \frac{|V_A|^2}{v_g} \right) \hat{\sigma}_-(t) + i\hat{\sigma}_z(t) \left[J\, \hat{a}(t) - i2\pi \frac{V_A V_C}{v_g} \hat{a}(t - d/v_g) \right] \tag{S21}$$

$$\dot{\hat{a}}(t) = -i\left(\omega_C - \omega_0 - i2\pi \frac{|V_C|^2}{v_g} \right) \hat{a}(t) - i\left[J\hat{\sigma}_-(t) - i2\pi \frac{V_C^* V_A}{v_g} \hat{\sigma}_-(t - d/v_g) \right] \tag{S22}$$

where we ignored the pumping terms for brevity (see, e.g., ref. [3] for a derivation of these equations for the case of two atoms) . We note that eqs. (S21)-(S22) are now non-local in time. That is, the value of one operator at a time t depends on the value of another operator at a time $t - d/v_g$. Within the Markov approximation, we can replace $\hat{a}(t - d/v_g) = \hat{a}(t) e^{i\omega_C \tau}$ and $\hat{\sigma}(t - d/v_g) = \hat{\sigma}(t) e^{i\omega_A \tau}$, where $\tau = d/v_g$. This is justified if d/v_g is much smaller than the typical timescale over which the system evolves, which is set by $\min\{J^{-1}, \Gamma_A^{-1}, \Gamma_C^{-1}\}$ [3]–[5]. Under this approximation, and in the single-excitation sector, the equations of motion become local and they read

$$\begin{pmatrix} \dot{\hat{\sigma}}_-(t) \\ \dot{\hat{a}}(t) \end{pmatrix} = -i \begin{pmatrix} \omega_A - i\Gamma_A & J - i\sqrt{\Gamma_C \Gamma_A} e^{i\omega_C \tau} \\ J - i\sqrt{\Gamma_C \Gamma_A} e^{i\omega_A \tau} & \omega_C - i\Gamma_C \end{pmatrix} \begin{pmatrix} \hat{\sigma}_-(t) \\ \hat{a}(t) \end{pmatrix}, \tag{S23}$$

and the frequencies of the single-photon eigenstates can be obtained by diagonalizing the matrix in eq. (S23). In particular, an EE is obtained with the imaginary part of the one of the two complex eigenfrequencies is zero. This leads to the condition

$$\Delta^2 + 4J^2 + 4\Gamma_A \Gamma_C (1 - \cos[(\omega_A + \omega_C)\tau]) + 4J\sqrt{\Gamma_A \Gamma_C}[\sin(\omega_A \tau) + \sin(\omega_C \tau)] = \left[\frac{\Delta(\Gamma_A - \Gamma_C) + 2\sin[(\omega_A + \omega_C)\tau]\Gamma_A \Gamma_C + 2J\sqrt{\Gamma_A \Gamma_C}[\cos(\omega_A \tau) + \cos(\omega_C \tau)]}{\Gamma_A + \Gamma_C} \right]^2, \tag{S24}$$

where $\Delta \equiv \omega_A - \omega_C$. For a given set of parameters $\{\omega_A, \omega_C, \Gamma_A, \Gamma_C, J\}$ one can look for values of τ that satisfies this relation. We note that for $\tau = 0$ we re-obtain the EE condition for the co-located case (eq. 1 of main text). A careful inspection reveals that eq. (S24) can be satisfied only when the condition for the co-located case, $\Delta\sqrt{\Gamma_A \Gamma_C} = J(\Gamma_A - \Gamma_C)$, holds, and τ is chosen such that ω_A, ω_C and $(\omega_A + \omega_C)$ are integer multiple of $2\pi/\tau$. When ω_A and ω_C are close but not equal, this requirement leads to very large values of τ, where the Markov approximation may break down [5]. On the other side, using very large detuning Δ can decrease the value of τ, but it will likely break the approximation that the coupling rate between cavity/atom and waveguide is independent of the frequency. The most feasible situation is therefore obtained when $\omega_A = \omega_C \equiv \omega_0$: satisfying equation (S24) in this case requires that $J = 0$ (i.e., atom and cavity interact only through the waveguide) and $\omega_0 \tau = n\pi,\ n \in \mathbb{N}$. In this case the EE coincides with the symmetric or anti-symmetric state, $|\psi\rangle \propto (|0,1\rangle \pm |1,0\rangle)$. The existence of this particular EE has been studied in systems composed of two separated atoms in a waveguide (see also next section), and it has been linked to non-reciprocal transmission [6].

E. Replacing both cavities with atoms

In the main text we considered the case where only one of the two cavities is replaced by a two-level atom. This was done in order to show the minimum requirement, in terms of atomic nonlinearities, to obtain nonlinear trapping into an embedded eigenstate. In general, a single-photon EE can be also obtained when both cavities are replaced by atoms.

Since the quantum descriptions of a cavity and an atom are identical in the single-excitation sector, several formulas presented for the atom-cavity case apply to the atom-atom case too. In particular, if the two atoms interact with the waveguide at the same point, a single-photon EE exists when $(\omega_1 - \omega_2)V_1 V_2 = J(V_1^2 - V_2^2)$ (where now $\omega_{1(2)}$ and $V_{1(2)}$ denote the frequency of atom 1(2) and its coupling to the waveguide, respectively, and J is the atom-atom coupling), and the EE is characterized by the destructive interference condition $V_1 A_1 + V_2 A_2 = 0$ (where $A_{1(2)}$ is the amplitude probability of having atom 1(2) excited). The frequencies of the single-photon EE and bright mode are the same as the ones obtained in eqs. (S16)-(S17) (with the proper re-labeling of the parameters).

The systems differ instead in the two-excitation sector. The two-excitation state is given only by the term $|\Psi^{(2)}\rangle = |ee\rangle$ (describing both atoms in the excited state), since now none of the two resonators can host two excitations. All the equations shown in section 7 (below) can be straightforwardly adapted to the case of two atoms by removing all the terms that describe the presence of two photons in one of the two resonators (i.e., removing the term $E_{2C}(t)$ from eqs. (S29) and following equations). As described in the previous section, an EE can also be obtained if the two atoms are resonant and they are displaced by a distance that is an integer multiple of the halved wavelength. The existence of these EEs have been investigated also in other works [7], [8].

F. Derivation of a density-matrix master equation from the input-output theory

In order to calculate numerically the dynamics of the system, it is useful to recast Eqs. (S11) and (S12) into a master equation for the density matrix (in Schrödinger representation) of the atom-cavity system. We start from a standard form of the master equation

$$\dot{\rho} = -i[\hat{H}_{AC} + \hat{H}_{input}, \rho] + \mathcal{L}\rho\mathcal{L}^{\dagger} - \{\mathcal{L}^{\dagger}\mathcal{L}, \rho\}/2 \qquad \textbf{(S25)}$$

where ρ is the density matrix of the atom-cavity system, $\hat{H}_{input} = \sqrt{2\pi / v_g}[(V_A\hat{\sigma}_- + V_C\hat{a})(\hat{c}_{R,IN}(t) + \hat{c}_{L,IN}(t)) + h.c.]$ and $\mathcal{L}$ is a Lindblad operator to be determined. The operator $\mathcal{L}$ describes the non-unitary evolution introduced by the waveguide through the decay rates $\Gamma_{A,C}$ and the modification of the coupling J. To determine the exact form of $\mathcal{L}$ we use the fact that the master equation in Schrödinger representation [eq. (S25)] can be expressed in Heisenberg representation for a general observable $\hat{x}$ as

$$\dot{\hat{x}} = i[\hat{H}_{AC} + \hat{H}_{input}, \hat{x}] + \mathcal{L}^{\dagger}x\mathcal{L} - \{\mathcal{L}^{\dagger}\mathcal{L}, \hat{x}\}/2\,. \qquad \textbf{(S26)}$$

Therefore, the operator $\mathcal{L}$ must be chosen such that, when eq. (S26) is applied to the operators $\hat{\sigma}_-$ and $\hat{a}$, the equations of motions (S11) and (S12) are re-obtained. By starting with the Ansatz $\mathcal{L} = A\hat{a} + B\hat{\sigma}_-$, it is straightforward to find that $\mathcal{L} = \sqrt{4\pi / v_g}(V_A\hat{a} + V_C\hat{\sigma}_-)$.

The results shown in fig. 1d and 4c of the main paper have been obtained by solving numerically the master equation (S25) with the aid of the open-source Python framework QuTip [9]. Similarly to what we do in the main text for the two-photon excitation, we assumed an identical excitation in the right and left channel, and we define the even input mode, $\hat{c}_{e,IN}(t) \equiv \left[\hat{c}_{R,IN}(t) + \hat{c}_{L,IN}(t)\right]/\sqrt{2}$. Moreover, we assume that the input field is a wave-packet in a coherent state with an average photon number $\langle N\rangle$. This allows us to replace the operator $\hat{c}_{e,IN}(t)$ by $\sqrt{\langle N\rangle}\,\xi(t)$ where $\xi(t)$ is a square-normalized scalar function. In particular, for the calculations in fig. 1d and 4c of the main paper we considered a Gaussian wave-packet $\xi(t) = (\pi\sigma^2)^{1/4}\exp[-(t-t_0)^2/(2\sigma^2) + i\omega t]$.

G. Dynamical equations in the two-photon sector

In this section we derive the differential equations that describe the time-evolution of an arbitrary two-excitation initial state of the atom-cavity-waveguide system, and we subsequently recast them in a form that facilitates the analytical and numerical calculations. We note that a similar technique has been used in ref. [10] for the case of a single atom. We start from the total Hamiltonian of the system (eq. 3 of the main text) in real-space formulation, and we introduce the even and odd modes, $\hat{c}_{e/o}(x) \equiv (1/\sqrt{2})[\hat{c}_R(x) \pm \hat{c}_L(-x)]$. The odd mode does not interact with the system and can be neglected, leading to

$$\hat{H}_e = \hat{H}_{AC} - i\int dx\, \hat{c}_e^{\dagger}(x)\,\partial_x\,\hat{c}_e(x) + \int dx\delta(x)\left[\hat{c}_e^{\dagger}(x)(\tilde{V}_C\hat{a} + \tilde{V}_A\hat{\sigma}_-) + h.c.\right] \qquad \textbf{(S27)}$$

where $\tilde{V}_{A/C} \equiv 2\sqrt{\pi}\,V_{A/C}$. The general form of a two-photon state is

$$\begin{aligned}|\psi(t)\rangle = \Big[&\int dx_1 dx_2\,\chi(x_1,x_2,t)\hat{c}_e^{\dagger}(x_1)\hat{c}_e^{\dagger}(x_2)/\sqrt{2} + \\ &+\int dx\left(\phi_A(x,t)\hat{\sigma}_+ + \phi_C(x,t)\hat{a}^{\dagger}\right)\hat{c}_e^{\dagger}(x) + \\ &+ E_{AC}(t)\hat{a}^{\dagger}\hat{\sigma}_+ + E_{2C}(t)(\hat{a}^{\dagger})^2/\sqrt{2}\Big]|0,g\rangle\end{aligned} \qquad \textbf{(S28)}$$

where the meaning of all functions is defined in the main text. Consistently with the main text we have set $v_g = 1$. We now apply the time-dependent Schrödinger equation $i\partial_t|\psi\rangle = \hat{H}_e|\psi\rangle$ and we equate the coefficients of each vector state. This leads to five differential equations,

$$\begin{cases}\partial_t\,\chi(x_1,x_2,t) = -(\partial_{x_1} + \partial_{x_2})\chi(x_1,x_2,t) - \\ \quad -i\dfrac{\tilde{V}_A}{\sqrt{2}}\left[\phi_A(x_1,t)\delta(x_2) + \phi_A(x_2,t)\delta(x_1)\right] - i\dfrac{\tilde{V}_C}{\sqrt{2}}\left[\phi_C(x_1,t)\delta(x_2) + \phi_C(x_2,t)\delta(x_1)\right], \\ \partial_t\phi_A(x,t) = -\partial_x\phi_A(x,t) - i(\omega_A - \omega_0)\phi_A(x,t) - iJ\phi_C(x,t) - \\ -i\dfrac{\tilde{V}_A^{\,*}}{\sqrt{2}}[\chi(x,0,t) + \chi(0,x,t)] - i\delta(x)\tilde{V}_C E_{AC}(t), \\ \partial_t\phi_C(x,t) = -\partial_x\phi_C(x,t) - i(\omega_C - \omega_0)\phi_C(x,t) - iJ\phi_A(x,t) - \\ \quad -i\dfrac{\tilde{V}_C^{\,*}}{\sqrt{2}}[\chi(x,0,t) + \chi(0,x,t)] - i\delta(x)\left(\tilde{V}_A E_{AC}(t) + \sqrt{2}\tilde{V}_C E_{2C}(t)\right), \\ \partial_t E_{2C}(t) = -2i(\omega_C - \omega_0)E_{2C}(t) - i\sqrt{2}JE_{AC}(t) - i\sqrt{2}\tilde{V}_C^{*}\phi_C(0,t), \\ \partial_t E_{AC}(t) = -i(\omega_C + \omega_A - 2\omega_0)E_{AC}(t) - i\sqrt{2}JE_{2C}(t) - i\left[\tilde{V}_C^{*}\phi_A(0,t) + \tilde{V}_A^{*}\phi_C(0,t)\right].\end{cases} \qquad \textbf{(S29)}$$

In principle, these five inter-dependent differential equations must be solved simultaneously to describe correctly the system dynamics. However, the system can be largely simplified by formally integrating the equation for $\partial_t\chi(x_1,x_2,t)$ with the aid of the Fourier transform, which leads to

$$\begin{aligned}&\chi(x_1,x_2,t) = \chi(x_1 - t, x_2 - t, 0) \\ &-i\frac{\tilde{V}_A}{\sqrt{2}}\left[\phi_A(x_1 - x_2, t - x_2)\theta(x_2)\theta(t - x_2) + \phi_A(x_2 - x_1, t - x_1)\theta(x_1)\theta(t - x_1)\right] \\ &+A \leftrightarrow C\end{aligned} \qquad \textbf{(S30)}$$

that is, the two-photon function $\chi(x_1,x_2,t)$ is given by its value at $t = 0$ (shifted by the photon propagation time) plus a term that depends on the values of ϕ_A and ϕ_C at different positions and times. By replacing eq. (S30) in the differential equations for $\partial_t\phi_A$ and $\partial_t\phi_C$ we get

$$\begin{aligned}&\partial_t\phi_A(x,t) = -\partial_x\phi_A(x,t) - i\left(\omega_A - \omega_0 - i\frac{|\tilde{V}_A|^2}{2}\right)\phi_A(x,t) \\ &-i\left(J - i\frac{\tilde{V}_A^{\,*}\tilde{V}_C}{2}\right)\phi_C(x,t) - \\ &-\theta(x)\theta(t-x)\left[|\tilde{V}_A|^2\,\phi_A(-x,t-x) + \tilde{V}_A^{*}\tilde{V}_C\phi_C(-x,t-x)\right] \\ &-i\frac{\tilde{V}_A^{\,*}}{\sqrt{2}}[\chi(x-t,-t,0) + \chi(-t,x-t,0)] - i\delta(x)\tilde{V}_C E_{AC}(t),\end{aligned} \qquad \textbf{(S31a)}$$

and

$$\partial_t\phi_C(x,t)=-\partial_x\phi_C(x,t)-i\left(\omega_C-\omega_0-i\frac{|\tilde{V}_C|^2}{2}\right)\phi_C(x,t)$$
$$-i\left(J-i\frac{\tilde{V}_C^*\tilde{V}_A}{2}\right)\phi_A(x,t)-$$
$$-\theta(x)\theta(t-x)\left[|\tilde{V}_C|^2\phi_C(-x,t-x)+\tilde{V}_C^*\tilde{V}_A\phi_C(-x,t-x)\right]$$
$$-i\frac{\tilde{V}_C^*}{\sqrt{2}}[\chi(x-t,-t,0)+\chi(-t,x-t,0)]-i\delta(x)\left(\tilde{V}_A E_{AC}(t)+\sqrt{2}\tilde{V}_C E_{2C}(t)\right) \quad \textbf{(S31b)}$$

By comparing eqs. (S31) with eqs. (S29) we note that the time evolution of $\phi_A(x,t)$ and $\phi_C(x,t)$ now depends only on the values of $\chi(x_1,x_2,t)$ at $t=0$, but additional terms appeared: in the first row of both eqs. (S31) the frequencies and coupling strength J have been modified, similarly to what happened in eqs. (S11) and (S12). Moreover, the second lines of both eqs. (S31) contain nonlocal terms that are nonzero for $0<x<t$ and depend on the values of $\phi_A(x,t)$ and $\phi_C(x,t)$ in the region $x<0$. These terms describe processes where the cavity or the atom first emits a photon and later absorbs another photon. An example of these processes is shown in fig. S1: at a certain time, a photon is in the waveguide at $x<0$ and travelling towards $+x$ (black dashed arrow), and the atom is excited; the atom can emit a photon that starts travelling towards $+x$ (red dashed arrow), thus feeding the two photon state. When the first photon arrives at $x=0$ it excites again the atom, and therefore it re-transfers the excitation from the two-photon state to the $\hat{c}_e^\dagger(x)\hat{\sigma}_+|0,g\rangle$ state.

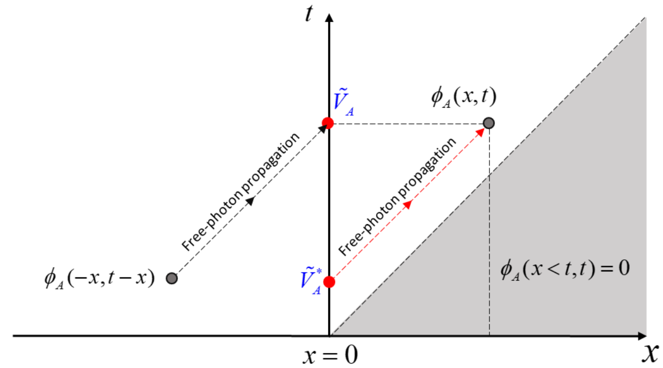

Fig. S1. Schematic description of the nonlocal term $|\tilde{V}_A|^2\phi_A(-x,t-x)$ in the first of eqs. (S31), which makes the field ϕ_A at (x,t) dependent on the value of the same field at $(-x,t-x)$. At time $t-x$, there is an amplitude probability equal to $\phi_A(-x,t-x)$ to have one photon at $-x$ and the atom excited. The atom decays into the waveguide with amplitude probability weighted by $\tilde{V}_A^*$, creating a photon that propagates freely (red dashed arrow) to the point (x,t). This process feeds the two-photon amplitude probability. Simultaneously, the photon that was at position $-x$ propagates freely (black dashed arrow) to the point $(0,t)$, where it is absorbed by the atom with an amplitude probability weighted by $\tilde{V}_A$. The overall process creates a state with the atom excited and a photon in the waveguide at position x, thus "feeding" the field $\phi_A(x,t)$ with a probability amplitude weighted by $|\tilde{V}_A|^2$. Similar explanations hold for the other nonlocal terms in eqs. (S31).

A further simplification of the dynamical equations (S29) is obtained by noticing that the equations for $\partial_t E_{2C}(t)$ and $\partial_t E_{AC}(t)$ depend only on the values of $\phi_A(x,t)$ and $\phi_C(x,t)$ at $x=0$. Since these two functions are discontinuous at $x=0$, we set $\phi_{A/C}(0,t)=[\phi_{A/C}(0^+,t)+\phi_{A/C}(0^-,t)]/2$. By integrating in space the eqs. for $\phi_A(x,t)$ and $\phi_C(x,t)$ in (S29) in the interval $[0-\epsilon,0+\epsilon]$, and taking the limit $\epsilon\to 0^+$, we find the boundary conditions

$$\begin{aligned}&\phi_A(0^+,t)=\phi_A(0^-,t)-i\tilde{V}_C E_{AC}(t),\\&\phi_C(0^+,t)=\phi_C(0^-,t)-i\left(\tilde{V}_A E_{AC}(t)+\sqrt{2}\tilde{V}_C E_{2C}(t)\right).\end{aligned} \quad \textbf{(S32)}$$

Plugging these results into the eqs. for $\partial_t E_{2C}(t)$ and $\partial_t E_{AC}(t)$ we get

$$\begin{cases}\partial_t E_{2C}(t)=-2i\left(\omega_C-\omega_0-i\frac{|\tilde{V}_C|^2}{2}\right)E_{2C}(t)\\ \quad -i\sqrt{2}\left(J-i\frac{\tilde{V}_C^*\tilde{V}_A}{2}\right)E_{AC}(t)-i\sqrt{2}\tilde{V}_C^*\phi_C(0^-,t),\\ \partial_t E_{AC}(t)=-i\left(\omega_C+\omega_A-2\omega_0-i\frac{|\tilde{V}_C|^2+|\tilde{V}_A|^2}{2}\right)E_{AC}(t)\\ -i\sqrt{2}\left(J-i\frac{\tilde{V}_C^*\tilde{V}_A}{2}\right)E_{2C}(t)-i\left[\tilde{V}_C^*\phi_A(0^-,t)+\tilde{V}_A^*\phi_C(0^-,t)\right],\end{cases} \quad \textbf{(S33)}$$

where, again, the influence of the waveguide on the decay rates and on the atom-cavity coupling is now shown explicitly.
The form of the equations (S31) and (S33) allows to apply a straightforward (analytical or numerical) method of solution (as also discussed in [10]). First, equations (S31) are solved for $x<0$ and arbitrary $t\geq 0$. In this region, the terms proportional to $\theta(x)\theta(t-x)$ and $\delta(x)$ do not contribute. Next, we use the values of $\phi_A(x,t)$ and $\phi_C(x,t)$ at $x=0^-$ to solve for $E_{2C}(t)$ and $E_{AC}(t)$ by (S33). Finally, we solve for $\phi_A(x,t)$ and $\phi_C(x,t)$ in the region $0<x<t$ by using the boundary conditions in eqs. (S32). In this step, the terms proportional to $\theta(x)\theta(t-x)$ in eqs. have been fully calculated in the first step. We also note that, by causality, $\phi_A(x,t)=\phi_C(x,t)=0$ for $x>t$ (if we assume that at $t=0$ no photon was present in the waveguide for $x>0$).
The calculations shown in the main text (figs. 2, 3 and 4a) have been obtained by solving numerically eqs. (S31) and (S33) with a home-built leapfrog FDTD algorithm, similar to the one outlined in ref. [11] (but generalized to the case of one atom and a cavity).

H. Efficiency of the release process, analytical results

In this section we first derive analytical results for the process in which the EE is released, and later we seek for conditions to achieve a perfect release. To simplify the notation, in this section we re-label $\tilde{V}_{A/C}\to V_{A/C}$. We consider an initial state in which the atom-cavity system contains one excitation in the EE and one photon is in the waveguide in the region $x<0$. The initial state is therefore

$$|\psi(t=0)\rangle=\int dxF(x)\hat{c}_e^\dagger(x)|\Psi_{EE}^{(1)}\rangle, \quad \textbf{(S34)}$$

where $|\Psi_{EE}^{(1)}\rangle = (V_C^2+V_A^2)^{-1/2}\cdot(V_c\hat{\sigma}_+ - V_A\hat{a}^\dagger)|0,g\rangle$ and $F(x)$ is a square-normalized function that describes the spatial shape of the single-photon pulse, with $F(x>0)=0$. In order to obtain the system evolution we need to solve eqs. (S31) and (S33) with the initial conditions $\phi_A(x,0)=\left(V_C/\sqrt{V_C^2+V_A{}^2}\right)F(x)$ and $\phi_C(x,0)=-\left(V_A/\sqrt{V_C^2+V_A{}^2}\right)F(x)$. For $x<0$, and assuming that the system is set in the EE condition, the evolution of the fields ϕ_A and ϕ_C is simply given by

$$\phi_A(x<0,t)=\frac{V_C}{\sqrt{V_C^2+V_A{}^2}}e^{-i\omega_{EE}t}F(x-t)$$
$$\phi_C(x<0,t)=-\frac{V_A}{\sqrt{V_C^2+V_A{}^2}}e^{-i\omega_{EE}t}F(x-t) \tag{S35}$$

where ω_{EE} is the frequency of the embedded eigenstate. The time evolution of $E_{2C}(t)$ and $E_{AC}(t)$ is then obtained from eqs. (S33), resulting in the linear system

$$\partial_t\begin{pmatrix}E_{2C}\\E_{AC}\end{pmatrix}=-i\begin{pmatrix}2(\omega_C-\omega_0)-iV_C^2 & \sqrt{2}\tilde{J}\\ \sqrt{2}\tilde{J} & \omega_C+\omega_A-2\omega_0-i(V_A^2+V_C^2)/2\end{pmatrix}+i\begin{pmatrix}\sqrt{2}V_CV_A\\V_A^2-V_C^2\end{pmatrix}\frac{F(-t)e^{-i\omega_{EE}t}}{\sqrt{V_A^2+V_C^2}} \tag{S36}$$

which can be solved with standard techniques (for brevity we have defined $\tilde{J}=J-iV_AV_C/2$). The general solution (assuming $E_{2C}(0)=E_{AC}(0)=0$) has the form

$$\begin{pmatrix}E_{2C}(t)\\E_{AC}(t)\end{pmatrix}=\begin{pmatrix}\alpha_1\\\beta_1\end{pmatrix}\mathrm{e}^{-ir_1t}\int_0^t dt'F(-t')e^{i(r_1-\omega_{EE})t'}+\begin{pmatrix}\alpha_2\\\beta_2\end{pmatrix}\mathrm{e}^{-ir_2t}\int_0^t dt'F(-t')e^{i(r_2-\omega_{EE})t'} \tag{S37}$$

where r_1 and r_2 are the eigenvalues of the matrix in eq. (S36) and $\alpha_1,\alpha_2,\beta_1$ and β_2 are constants that depend on the system parameters. We can now solve for $\phi_A(x,t)$ and $\phi_C(x,t)$ for $t>x>0$. Note that, when starting in the state described in eq. (S35), the terms proportional to $\theta(-x)\theta(t-x)$ in eqs. (S31) cancel each other. Therefore, for $t>x>0$ we need to solve the same equations that we had for $x<0$,

$$\begin{cases}\partial_t\phi_A(x,t)=-\partial_x\phi_A(x,t)-i\left(\omega_A-iV_A^2/2\right)\phi_A(x,t)-i\tilde{J}\phi_C(x,t),\\ \partial_t\phi_C(x,t)=-\partial_x\phi_C(x,t)-i\left(\omega_C-iV_C^2/2\right)\phi_C(x,t)-i\tilde{J}\phi_A(x,t),\end{cases} \tag{S38}$$

but with the boundary conditions given by eq. (S32), where $\phi_{A/C}(0^-,t)=\frac{\pm V_{C/A}}{\sqrt{V_C^2+V_A^2}}e^{-i\omega_{EE}t}F(-t)$. By applying the Fourier transform to the system in eq. (S38), it can be easily shown that the general solution has the form

$$\phi_A(x>0,t)=\frac{1}{\sqrt{V_C^2+V_A^2}}\left[V_CC_1(x-t)e^{-i\omega_{EE}t}+V_AC_2(x-t)e^{-i\omega_Bt}e^{-\Gamma t}\right]$$
$$\phi_C(x>0,t)=\frac{1}{\sqrt{V_C^2+V_A^2}}\left[-V_AC_1(x-t)e^{-i\omega_{EE}t}+V_CC_2(x-t)e^{-i\omega_Bt}e^{-\Gamma t}\right] \tag{S39}$$

where $C_1(x-t)$ and $C_2(x-t)$ are functions to be determined in order to satisfy the boundary conditions in eqs. (S32), and ω_B and Γ are the frequency and the decay rate of the bright single-photon eigenstate of the atom-cavity system.

We now seek for conditions for which, when a single photon impinges on an excited EE, the EE is completely released. That is, only the two-photon probability amplitude $\chi(x_1,x_2,t)$ is nonzero for $t\to+\infty$. The functions $E_{2C}(t)$ and $E_{AC}(t)$ will always decay to zero for $t\to+\infty$, because an EE does not exist in the two-excitation sector of the atom-cavity system. Therefore, we only need to ensure that $\phi_{A/C}(x,t)\to 0$ for $t\to+\infty$. The terms proportional to $C_2(x-t)$ in eqs. (S39) decay to zero for $t\to+\infty$ because $\Gamma\in\mathbb{R}$ and $\Gamma>0$. We therefore conclude that a necessary condition to ensure that $\phi_{A/C}(x,t)\to 0$ is that $C_1(z)=0$ for every $z\equiv x-t<0$. We note that it is not sufficient to require that $C_1(z)\propto e^{\gamma z}$ with $\gamma>0$, because all the points $x\approx t$ will not experience any decay for $t\to+\infty$.
By applying the boundary conditions in eqs. (S32) to eqs. (S39) we find that

$$C_1(z)=\sqrt{V_C^2+V_A^2}F(z)+\int_0^z dz'\left(\alpha e^{i(r_1-\omega_{EE})(z-z')}+\beta e^{i(r_2-\omega_{EE})(z-z')}\right)F(z') \tag{S40}$$

where α and β are constants that depend on the system parameters. Requiring that $C_1(z)=0$ leads to the Volterra homogeneous equation

$$F(z)=\int_0^z dz'K(z-z')F(z') \tag{S41}$$

which, for a continuous kernel $K(z-z')$ (as in this case), admits only the trivial solution $F(z)=0$ [12]. We therefore conclude that it is not possible to find a pulse shape $F(x)$ for the initial single-photon pulse such that only a two-photon state is present for $t\to\infty$. However, it can be easily shown that if, for example, $F(x)\propto\exp[-(x-x_0)^2/(2\sigma^2)+ikx]$, equation (S41) can be satisfied with arbitrarily high accuracy as $\sigma\to\infty$. That is, we can obtain final states arbitrarily close to a two-photon state by working with broader and broader impinging pulses.

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