Supplementary Material
Light-matter interaction in open cavities with dielectric stacks

I. SINGLE LAYER AND HAMILTONIAN DERIVATION

The electromagnetic field within a cavity is described by solution of the Helmholtz equation:

$$\left(\frac{d^2}{dx^2}+\varepsilon_r\left(x\right)\frac{\omega^2}{c^2}\right)\Phi_\omega\left(x\right)=0, \tag{S1}$$

where $\varepsilon_r(x)$ is the relative permittivity of the physical medium, $\Phi_\omega(x)$ is the space-dependent field eigenmode of continuous index $\omega=2\pi c/\lambda$, where λ is the wavelength of the travelling wave and c is the speed of light. The orthonormalization condition for these modes is given by:

$$\langle\Phi_\omega,\Phi_{\omega'}\rangle=\int_{-\infty}^{\infty}dx\;\varepsilon_r\left(x\right)\Phi^*_\omega\left(x\right)\Phi_{\omega'}\left(x\right)=\delta\left(\omega-\omega'\right). \tag{S2}$$

For convenience, eigenmodes can be spatially separated into three regions: between the mirrors, within the stack, and outside of the cavity:

$$\Phi_\omega\left(x\right)=\Phi_{\omega,\mathrm{in}}\left(x\right)+\Phi_{\omega,\mathrm{stack}}\left(x\right)+\Phi_{\omega,\mathrm{out}}\left(x\right). \tag{S3}$$

The corresponding quantised electric field can be written in the Schrödinger picture as:

$$\hat{E}\left(x\right)=-i\int_0^{\infty}d\omega\sqrt{\frac{\hbar\omega}{2\varepsilon_0}}\left(\Phi_\omega\left(x\right)\hat{a}_\omega-\Phi^*_\omega\left(x\right)\hat{a}^\dagger_\omega\right), \tag{S4}$$

where

$$\begin{aligned}[\hat{a}_\omega,\hat{a}^\dagger_{\omega'}]&=\delta\left(\omega-\omega'\right),\\ [\hat{a}_\omega,\hat{a}_{\omega'}]&=0.\end{aligned} \tag{S5}$$

We highlight that the above quantisation procedure from first principles does not break the modes into separate parts (as opposed to any approach first quantising the field inside a perfect cavity and subsequently coupling the field modes to the outside using a variety of input-output formalisms[1,2]), but rather considers the global modes occupying all the Hilbert space. These global modes behave as uncoupled quantum harmonic oscillators with a Hamiltonian of the form:

$$\hat{H}=\int_0^{\infty}d\omega\;\hbar\omega\left(\hat{a}^\dagger_\omega\hat{a}_\omega+\frac{1}{2}\right), \tag{S6}$$

where the $1/2$ term is usually removed by renormalizing the zero-point energy of the field.

To take into account the interaction of the field with an atom localized between the mirrors, one would like:

1. To derive an effective Hamiltonian and master equations accounting for the dynamics inside of the cavity;
2. To describe the leakage of the photons from the cavity;
3. To characterize the photons leaking out of the cavity.

For this, we derive the Hamiltonian including the global field and the atom within the cavity. The presence of the atom from the outset makes the calculations relatively simpler than introducing it later, as shown in[2], since the atom, localized in space and interacting with the global modes $\hat{a}_\omega$, leads to a mode-selective behavior of the overall system.

Let us assume that a point-like two-level atom with states $|g\rangle$ and $|e\rangle$ separated in energy by $\hbar\omega_A$, is positioned at $x=x_A$, between a perfect, mirror placed at $x=-\ell_c$ and a partially transparent dielectric mirror positioned at the origin. Here, $-\ell_c<x_A<0$, where ℓ_c is the geometric length of the cavity. The electric field at the position of the atom can be written by evaluating Eq. (S4) at $x=x_A$, and we consider a field-atom dipolar interaction:

$$\hat{V}=-\hat{d}\hat{E}\left(x_A\right), \tag{S7}$$

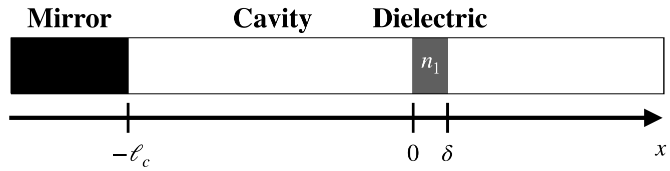

FIG. S1. Description of the single-layered cavity model. A perfect mirror stands at $x = -\ell_c$, delimiting a cavity of length ℓ_c with a partially transparent dielectric material from $x = 0$ to $x = \delta$.

where $\hat{d} = d\hat{\sigma}_+ + d^*\hat{\sigma}_-$ is the dipole moment of the atom, $\hat{\sigma}_+ = |e\rangle\langle g|$ and $\hat{\sigma}_- = |g\rangle\langle e|$. By applying the rotating wave approximation, the interaction terms read:

$$\hat{V} = i\hbar \int_0^{+\infty} d\omega \left(\eta_\omega \hat{\sigma}_+ \hat{a}_\omega - \eta_\omega^* \hat{a}_\omega^\dagger \hat{\sigma}_- \right), \tag{S8}$$

with a coupling strength given by:

$$\eta_\omega = \sqrt{\frac{\omega}{2\hbar\varepsilon_0}}\, d\,\Phi_{\omega,\text{in}}(x_A). \tag{S9}$$

Therefore, the total Hamiltonian of the atom-cavity global system is of the form

$$\hat{H} = \hbar\omega_A \hat{\sigma}_+\hat{\sigma}_- + \int_0^{+\infty} d\omega\, \hbar\omega\, \hat{a}_\omega^\dagger \hat{a}_\omega + i\hbar \int_0^{+\infty} d\omega \Big(\eta_\omega \hat{\sigma}_+ \hat{a}_\omega - \eta_\omega^* \hat{a}_\omega^\dagger \hat{\sigma}_- \Big), \tag{S10}$$

where the first term corresponds to the energy of the atom, the second term to the energy of the global electromagnetic field and the third term the interaction of this field with the atom placed between the mirrors.

In order to derive an effective Hamiltonian describing the dynamics of the open atom-cavity system, we need to derive an effective atom-cavity coupling strength from Eq. (S9). Therefore, it is necessary to find the modes Φ_ω that satisfy the orthonormalization condition Eq. (S2). We perform this calculation, taking into account the structure of the partially transparent mirror of the cavity.

Now, we describe the field modes in Bragg stacks. We first review the propagation of light in a cavity with a thin single-layered mirror and then build upon it to describe the case of multilayered stacks. Although this procedure is standard in the literature, it will prove important to show explicitly the differences that arise from realistic mirror structures.

For the purposes of future reference, we list here the parameters used to characterise our system:

Definition	**Notation**
Mirror stack design wavelength and angular frequency.	$\lambda_0, \omega_0 = \frac{2\pi c}{\lambda_0}$
Propagating light wavelength and angular frequency (continuous).	$\lambda, \omega = \frac{2\pi c}{\lambda}$
Geometric cavity length, cavity resonance wavelength and angular frequency.	$\ell_c, \lambda_m,$ $\omega_m = \frac{2\pi c}{\lambda_m} = \frac{\pi c m}{\ell_c}$
Actual cavity resonance angular frequency, leading to a resonance effective length.	$\omega_{\text{eff}} = \omega_N^{(m)},\ \ell_{\text{eff}} = \frac{\pi c m}{\omega_{\text{eff}}}$
Coupling factor cavity length.	$L_N^{(m)}$

TABLE S1. Relevant notations for the different wavelengths and frequencies within the atom-cavity system. The index m corresponds to the number of anti-nodes of the wave between the mirrors, and N to the $2N-1$ dielectric layers.

Here, we describe the situation arising from a single-layered stack. Let us model a cavity of length ℓ_c. For the sake of simplicity, we assume that such a cavity has a perfect mirror on the left side, positioned at $x = -\ell_c$, and a partially transparent dielectric placed at the origin, with a real refractive index n_1 and a thickness δ. The dielectric layer thickness has been designed to support a target wavelength, λ_0. This structure can be seen in Fig. S1. In this structure, the spatial distribution of the electromagnetic field mode $\Phi_\omega(x)$ with frequency $\omega = 2\pi c/\lambda$ is described by the Helmholtz equation (S1), with a relative permittivity structure of the form

$$\varepsilon_r(x) = \begin{cases} 1 & x \in [-\ell_c, 0] \cup [\delta, \infty) \\ n_1^2 & x \in (0, \delta) \end{cases}. \tag{S11}$$

Under these conditions, Helmholtz's equation is solved by a superposition of plane waves of the form

$$\Phi_\omega(x) = A_+ e^{i\frac{\omega}{c}\sqrt{\varepsilon_r(x)}x} + A_- e^{-i\frac{\omega}{c}\sqrt{\varepsilon_r(x)}x}. \tag{S12}$$

The distinct solutions are then stitched together by the condition that $\Phi_\omega(x)$ and its derivative must ensure continuity throughout the points of discontinuity of $\varepsilon_r(x)$, thus fixing the values of the coefficients A_+ and A_- for every region. For a single-layered stack, the solution has the form

$$\Phi_\omega(x) = \Phi_{\omega,\mathrm{in}}(x)\,\chi_{[-\ell_c,0]} + \Phi_{\omega,\mathrm{stack}}(x)\,\chi_{(0,\delta)} + \Phi_{\omega,\mathrm{out}}(x)\,\chi_{[\delta,\infty)}, \tag{S13}$$

following the convention defined by Eq. (S3). Here, $\chi_{\mathscr{D}}$ represents the indicator function within a domain $\mathscr{D} \subset \mathbb{R}$, i.e., $\chi_{\mathscr{D}}(x) = 1$ for $x \in \mathscr{D}$ and 0 otherwise.

By following the reflection of a monochromatic wave coming from $+\infty$ to the cavity, we derive the complete set of orthonormalized modes, described in the three regions:

$$\begin{aligned}
\Phi_{\omega,\mathrm{in}}(x) &= \frac{2i}{\sqrt{2\pi c\mathscr{A}}} e^{i\frac{\omega}{c}\ell_c} T(\omega) \sin\left[\frac{\omega}{c}(x+\ell_c)\right], \\
\Phi_{\omega,\mathrm{stack}}(x) &= \frac{1}{\sqrt{2\pi c\mathscr{A}}} e^{i\frac{\omega}{c}\ell_c} \frac{T(\omega)}{1+r_1}\left[(e^{i\frac{\omega}{c}\ell_c} - r_1 e^{-i\frac{\omega}{c}\ell_c}) e^{i\frac{\omega}{c}n_1 x} + (r_1 e^{i\frac{\omega}{c}\ell_c} - e^{-i\frac{\omega}{c}\ell_c}) e^{-i\frac{\omega}{c}n_1 x}\right], \\
\Phi_{\omega,\mathrm{out}}(x) &= \frac{1}{\sqrt{2\pi c\mathscr{A}}} \left(e^{2i\frac{\omega}{c}\ell_c} \frac{T(\omega)}{T^*(\omega)} e^{i\frac{\omega}{c}x} - e^{-i\frac{\omega}{c}x} \right),
\end{aligned} \tag{S14}$$

where $\mathscr{A}$ is the transverse area of the mode (a quantity which is calculated when the mode is normalized in three dimensions), and r is the single-layer reflectivity

$$r_1 = \frac{n_1 - 1}{n_1 + 1}, \tag{S15}$$

$T(\omega)$ is the cavity spectral response function, which describes the ratio of intensity between the inside and outside of the cavity, with respect to a particular frequency ω:

$$T(\omega) = \frac{t(\omega)}{1 + r(\omega) e^{2i\frac{\omega}{c}(\ell_c + \frac{\delta}{2})}}. \tag{S16}$$

$T(\omega)$ depends on the single layer spectral transmission response function[3] $t(\omega)$:

$$t(\omega) = \frac{(1-r_1^2) e^{i(n_1-1)\frac{\omega}{c}\delta}}{1 - e^{2in_1\frac{\omega}{c}\delta} r_1^2}, \tag{S17}$$

as well as on the single layer spectral reflection response function $r(\omega)$:

$$r(\omega) = e^{-i\frac{\omega}{c}\delta} \frac{r_1(e^{2in_1\frac{\omega}{c}\delta} - 1)}{1 - e^{2in_1\frac{\omega}{c}\delta} r_1^2} = |r(\omega)|\, e^{i\phi_r(\omega)}. \tag{S18}$$

It is important to stress that both of these parameters are complex and have associated phases. We will use $\phi_r(\omega)$ in later paragraphs. Together, $t(\omega)$ and $r(\omega)$ satisfy the beam splitter relations

$$|t(\omega)|^2 + |r(\omega)|^2 = 1, \tag{S19}$$

$$r^*(\omega)t(\omega) + t^*(\omega)r(\omega) = 0. \tag{S20}$$

The squared norm of the cavity response function can be decomposed as a sum of Lorentzian-like functions[3,4], still having $\tilde{\omega}_m$ and γ_1 depending on ω: (see the details in section III of this Supplementary Material):

$$|T(\omega)|^2 = \sum_{m=-\infty}^{\infty} \frac{c}{2L_1} \frac{\gamma_1(\omega)}{(\omega - \tilde{\omega}_m(\omega))^2 + \left(\frac{\gamma_1(\omega)}{2}\right)^2}, \tag{S21}$$

where

$$\begin{aligned}\gamma_1(\omega) &= -\frac{c}{L_1}\ln|r(\omega)|,\\ \tilde{\omega}_m(\omega) &= m\frac{\pi c}{L_1}+\frac{c}{2L_1}(\pi-\phi_r(\omega)),\\ L_1 &= \ell_c+\frac{\delta}{2},\end{aligned} \tag{S22}$$

and index 1 in L_1 and γ_1 indicates that they are for the case of a single-layer mirror.

In the high-finesse limit (reached when assuming a fictitious dielectric with a high refractive index), and for cavities having $\delta \ll \ell_c$, such that $L_1 \approx \ell_c$, the squared norm of the response function can be approximated as follows:

$$|T(\omega)|^2 \approx \sum_{m=-\infty}^{\infty}\frac{c}{2\ell_c}\frac{\Gamma_m}{(\omega-\tilde{\omega}_m)^2+\left(\frac{\Gamma_m}{2}\right)^2}, \tag{S23}$$

where

$$\Gamma_m = \gamma_1(\omega_m) = -\frac{c}{\ell_c}\ln|r(\omega_m)|, \tag{S24}$$

$$\tilde{\omega}_m = \omega_m+\frac{c}{2\ell_c}(\pi-\phi_r(\omega_m)), \tag{S25}$$

with $\omega_m = m\pi c/\ell_c$.

At this stage, the terms in the sum in Eq. (S23) become Lorentzian, with each term centered around the resonance frequency $\tilde{\omega}_m$ and well separated from each other. Taking this into account we can now write the cavity response function as

$$T(\omega) \approx \sum_{m=-\infty}^{\infty}\sqrt{\frac{c}{2\ell_c}}\frac{\sqrt{\Gamma_m}}{(\omega-\tilde{\omega}_m)+i\frac{\Gamma_m}{2}} = \sum_{m=-\infty}^{\infty}T_m(\omega), \tag{S26}$$

where $T_m(\omega)$ is so narrow that it does not overlap with the other Lorentzians $T_{m'}(\omega)$. With this result we can now write the coupling strength (S9) in the form:

$$\eta_\omega = \sum_{m=-\infty}^{\infty}\left(i\sqrt{\frac{\omega}{\hbar\varepsilon_0\ell_c\mathscr{A}}}d\,e^{i\frac{\omega}{c}\ell_c}\sin\left[\tfrac{\omega}{c}(x_A+\ell_c)\right]\times\sqrt{\frac{\Gamma_m}{2\pi}}\frac{1}{(\omega-\tilde{\omega}_m)+i\frac{\Gamma_m}{2}}\right), \tag{S27}$$

which describes the coupling between the global electric field with the atom localized between the mirrors. The product $\ell_c\mathscr{A}$ appearing in the coupling strength can be interpreted as the cavity mode volume. In the high-finesse case considered here, it happens to be identical to the mode volume determined by the geometric parameters of the cavity: its mirror separation (or geometric length, ℓ_c), and mirror area, $\mathscr{A}$. However, we emphasise that the mode volume was not introduced by normalizing the field in a perfect resonator of length ℓ_c. Instead, it is a result of the finite width of the resonances in Eq. (S23). To this respect, recovering ℓ_c only appears in the case of a long, high-finesse resonator and this may not prevail when such condition is not met, as we shall demonstrate in the following discussion.

II. CALCULATION OF THE MODE STRUCTURE FOR THE MULTILAYER CASE

By following the continuity of the field and its derivative, we solve Helmholtz equation (S1) and find that the mode can be described in terms of the following functions:

$$B_0(\omega,x) = e^{i\frac{\omega}{c}(x+\ell_c)}, \tag{S28}$$

$$B_j(\omega,x) = \begin{cases}\frac{1}{1-r_1}\left(B_{j-1}(\omega,x_2(j))+r_1B^*_{j-1}(\omega,x_2(j))\right)e^{i\frac{\omega}{c}(x-x_2(j))} & j\text{ even},\\ \frac{1}{1+r_1}\left(B_{j-1}(\omega,x_1(j))-r_1B^*_{j-1}(\omega,x_1(j))\right)e^{i\frac{\omega}{c}n_1(x-x_1(j))} & j\text{ odd},\end{cases} \tag{S29}$$

where

$$x_1(j) = \frac{j-1}{2}(\delta+\alpha), \tag{S30}$$

$$x_2(j) = \frac{j}{2}(\delta+\alpha)-\alpha. \tag{S31}$$

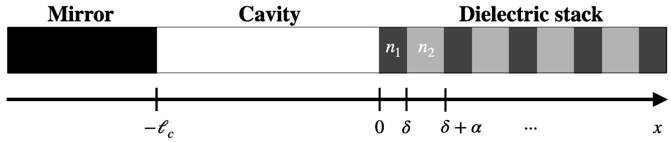

FIG. S2. Description of the considered model. A perfect mirror stands at $x=-\ell_c$, delimiting a cavity of length ℓ with an alternating dielectric stack standing from zero.

The solution to the Helmholtz equation is then given by

$$\Phi_\omega(x)=\sum_{j=0}^{2N} CA_j(\omega,x)\chi_{\Omega_j}, \tag{S32}$$

where j runs over the $2N-1$ possible layers, labelled as $\Omega_j\subset\mathbb{R}$. Here, $\chi_{\mathscr{D}}$ represents the indicator function within a domain $\mathscr{D}\subset\mathbb{R}$, i.e., $\chi_{\mathscr{D}}(x)=1$ for $x\in\mathscr{D}$ and 0 otherwise. The $2N$th term describes the mode exiting the cavity. C is a normalization factor common to all terms and

$$A_j(\omega,x)=B_j(\omega,x)-B_j^*(\omega,x). \tag{S33}$$

In particular, Ω_j has the form

$$\Omega_0=[-\ell_c,0], \tag{S34}$$

$$\Omega_j=\begin{cases}[x_2(j),x_2(j)+\alpha], & j \text{ even},\\ [x_1(j),x_1(j)+\delta], & j \text{ odd}.\end{cases} \tag{S35}$$

Here $A_0(\omega,x)$ corresponds to the mode propagating between the two mirrors and $A_{2N}(\omega,x)$ is the mode corresponding to the outgoing wave.

In the field amplitude ratio of the multilayered cavity, which has the form

$$\mathscr{T}(\omega)=\frac{e^{-i\frac{\omega}{c}(\ell_c+(N-1)(\delta+\alpha))}}{B_{2N-2}^*(\omega)}\frac{t(\omega)}{1+e^{i\frac{\omega}{c}\delta}e^{i\phi_B(\omega)}r(\omega)}, \tag{S36}$$

where $\phi_B(\omega)=\arg\left(B_{2N-2}/B_{2N-2}^*\right)$, we omitted the argument of $B_{2N-2}=B_{2N-2}(\omega,x_1(2N-1))$ for simplicity.

By indexing the response function defined above with the number of layer pairs such that $\mathscr{T}(\omega)=\mathscr{T}_N(\omega)$, it can be shown that

$$\frac{e^{-i\frac{\omega}{c}(\ell_c+(N-1)(\delta+\alpha))}}{B_{2N-2}^*(\omega)}=\mathscr{T}_{N-1}(\omega), \tag{S37}$$

where $\mathscr{T}_{N-1}(\omega)$ is the response function of a cavity having a dielectric stack with $2N-3$ dielectric layers.

III. LORENTZIAN STRUCTURE OF THE CAVITY SPECTRAL RESPONSE FUNCTION

In this supplementary section we derive the Lorentzian structure of the cavity response function, shown in Eq. (S21) [2]:

$$T(\omega)=\frac{t(\omega)}{1+r(\omega)e^{2i\frac{\omega}{c}L_1}}. \tag{S38}$$

Writing the square modulus of $T(\omega)$, and using the conditions of equations (S19) and (S20), we get

$$|T(\omega)|^2=\frac{1-|r|^2}{|1+|r|e^{i\Phi}|^2}=\left[1-\frac{|r|e^{i\Phi}}{1+|r|e^{i\Phi}}-\frac{|r|e^{-i\Phi}}{1+|r|e^{-i\Phi}}\right], \tag{S39}$$

where for simplicity we have not written the dependence of $r(\omega)$ on ω, and we have used the following notations $r(\omega)=|r|e^{i\Phi_r(\omega)}$, $\Phi=2\frac{\omega}{c}L_1+\Phi_r(\omega)$. Using the geometric series formula

$$\sum_{n=1}^{+\infty}q^n=\frac{q}{1-q},\quad |q|<1 \tag{S40}$$

we can write the above expression for the response function as follows

$$|T(\omega)|^2 = 1 + \sum_{n=1}^{+\infty} |r|^n \left(e^{in(\Phi+\pi)} + e^{-in(\Phi+\pi)} \right)$$
$$= \sum_{n=-\infty}^{+\infty} |r|^{|n|} e^{in(\Phi+\pi)}. \tag{S41}$$

We further apply the Poisson summation formula, which states

$$\sum_{n=-\infty}^{+\infty} f(n) = \sum_{m=-\infty}^{+\infty} \int_{-\infty}^{+\infty} dx\, f(x) e^{-i2\pi mx} = \sum_{m=-\infty}^{+\infty} \tilde{f}(m) \tag{S42}$$

where $\tilde{f}(m) = \mathbf{F}_m\left[f(x)\right]$ is the Fourier transform of $f(x)$. We apply the Fourier transform to the function

$$f(x) = |r|^{|x|} e^{ix(\Phi+\pi)} = e^{|x|\ln|r| + ix(\Phi+\pi)}$$

and obtain

$$\mathbf{F}_m\left[f(x)\right] = \frac{2a}{a^2+\alpha^2}, \tag{S43}$$

where $a = -\ln|r|$, $\alpha = \Phi + \pi - 2\pi m$. Hence,

$$\tilde{f}(m) = \frac{-2\ln|r|}{(\ln|r|)^2 + ((\Phi+\pi) - 2\pi m)^2}. \tag{S44}$$

We further expand this expression by writing the explicit expression for Φ:

$$\tilde{f}(m) = \frac{-2\ln|r|}{(\ln|r|)^2 + ((2\frac{\omega}{c}L_1 + \Phi_r + \pi) - 2\pi m)^2}, \tag{S45}$$

and multiplying both the numerator and the denominator by $(c/2L_1)^2$ we find the Lorentzian structure of the cavity spectral response function, still having the parameters $\tilde{\omega}_m$ and γ_1 depend on ω:

$$|T(\omega)|^2 = \sum_{n=-\infty}^{+\infty} \frac{c}{2L_1} \frac{\gamma_1(\omega)}{(\omega - \tilde{\omega}_m(\omega))^2 + \left(\frac{\gamma_1(\omega)}{2}\right)^2}, \tag{S46}$$

$$\gamma_1(\omega) = -\frac{c}{L_1}\ln|r(\omega)|, \tag{S47}$$

$$\tilde{\omega}_m = \frac{\pi c}{L_1} m - \frac{c}{2L_1}(\Phi_r(\omega) + \pi), \tag{S48}$$

In the limit of large refractive indices, the reflection coefficient becomes $r(\omega) = r_1 e^{-i(\pi + \frac{\omega}{c}\delta)}$, i.e $\Phi_r(\omega) = -(\pi + \frac{\omega}{c}\delta)$, which tends to $-\pi$ in the limit of a negligible width δ of the dielectric, and we recover the expression for a perfect cavity.

IV. EFFECTIVE HAMILTONIAN

In order to extract the description of the atom-cavity open system, we first solve the Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} |\psi(t)\rangle = \hat{H} |\psi(t)\rangle. \tag{S49}$$

If we put the Hamiltonian of Eq. (3) and the following wavefunction

$$|\psi(t)\rangle = \int_0^{+\infty} d\omega e^{-i\omega t} c_{g,1}(\omega,t) |g, 1_\omega\rangle + e^{-i\omega_A t} c_{e,0}(t) |e, 0\rangle \tag{S50}$$

into Eq. (S49), where $|1_\omega\rangle$ corresponds to the single excitation of a mode Φ_ω, i.e. $|1_\omega\rangle = \hat{a}^\dagger_\omega|\mathbf{0}\rangle$, we obtain the dynamical equations for the coefficients $c_{g,1}(\omega,t)$, $c_{e,0}(t)$[4]:

$$\dot{c}_{g,1}(\omega,t) = -\sum_m \eta^*_{\omega,m} e^{i(\omega-\omega_A)t} c_{e,0}(t), \tag{S51}$$

$$\dot{c}_{e,0}(t) = \int_0^{+\infty} d\omega \sum_m \eta_{\omega,m} e^{-i(\omega-\omega_A)t} c_{g,1}(\omega,t). \tag{S52}$$

In order to trace out the continuous degrees of freedom from the dynamics, we define the integrated, mode-selective probability amplitude:

$$c^{(m)}_{g,1}(t) = \frac{1}{g_m}\int_0^{+\infty} d\omega\, \eta_{\omega,m} e^{-i(\omega-\omega_A)t} c_{g,1}(\omega,t). \tag{S53}$$

We now write the time derivative of this quantity, using the property

$$\eta^*_{\omega,m}\eta_{\omega,m'} = \delta_{mm'}|\eta_{\omega,m}|^2, \tag{S54}$$

which is a consequence of the fact that the response function is a sum of narrow peaked and well separated Lorentzians. Using this property we get the equation of motion for the m-th mode, using the integration of Eq. (S51) and inverting the order of time and frequency integration:

$$\begin{aligned}\dot{c}^{(m)}_{g,1}(t) = \dot{c}^{(m,0)}_{g,1}(t) &- \frac{1}{g_m}\int_0^{+\infty} d\omega\, |\eta_{\omega,m}|^2 c_{e,0}(t)\\ &+ \frac{i}{g_m}\int_0^t dt' c_{e,0}(t')\int_0^{+\infty} d\omega\, |\eta_{\omega,m}|^2(\omega-\omega_A)e^{-i(\omega-\omega_A)(t-t')}\end{aligned} \tag{S55}$$

where we introduce the time derivative of the initial time defined as follows:

$$c^{(m,0)}_{g,1} = \frac{1}{g_m}\int_0^{+\infty} d\omega\, \eta_{\omega,m} e^{-i(\omega-\omega_A)t} c_{g,1}(\omega,0). \tag{S56}$$

Two integrals appear in (S55):

$$\mathscr{I}_1 = \int_0^{+\infty} d\omega\, |\eta_{\omega,m}|^2, \tag{S57}$$

$$\mathscr{I}_2 = \int_0^{+\infty} d\omega\, |\eta_{\omega,m}|^2(\omega-\omega_A)e^{-i(\omega-\omega_A)(t-t')}, \tag{S58}$$

which can be evaluated using complex contour methods, and in the limit of $\varepsilon = \gamma^{(m)}_N \frac{x_A+\ell}{c} \ll 1$, we obtain (here after in the parameters $\gamma^{(m)}_N$, $L^{(m)}_N$ and $\omega^{(m)}_N$ we omit the index indicating the number of layers)

$$\mathscr{I}_1 = \frac{\omega^{(m)}|d|^2}{\hbar\varepsilon_0\mathscr{A}L^{(m)}}\sin^2\left[2\frac{\omega^{(m)}}{c}(x_A+\ell_c)\right], \tag{S59}$$

$$\mathscr{I}_2 = \frac{\omega^{(m)}|d|^2}{\hbar\varepsilon_0\mathscr{A}L^{(m)}}\sin^2\left[2\frac{\omega^{(m)}}{c}(x_A+\ell_c)\right]\left(\Delta_m - i\frac{\gamma^{(m)}}{2}\right)e^{-i(\Delta_m - i\frac{\gamma^{(m)}}{2})(t-t')}, \tag{S60}$$

where $\Delta_m = \omega^{(m)} - \omega_A$. We note that, since we defined g_m as the normalization coefficient of (S53), then the integrals $\mathscr{I}_1$ and $\mathscr{I}_2$ become

$$\mathscr{I}_1 = |g_m|^2, \tag{S61}$$

$$\mathscr{I}_2 = |g_m|^2\left(\Delta_m - i\frac{\gamma^{(m)}}{2}\right)e^{-i(\Delta_m - i\frac{\gamma^{(m)}}{2})(t-t')}, \tag{S62}$$

with

$$g_m = i\sqrt{\frac{\omega^{(m)}}{\hbar\varepsilon_0\mathscr{A}L^{(m)}}}\, d e^{i\frac{\omega^{(m)}}{c}\ell_c}\sin\left[\frac{\omega^{(m)}}{c}(x_A+\ell_c)\right] + \mathscr{O}\left(\varepsilon^2\right). \tag{S63}$$

Having obtained the expression for g_m, we can now rewrite the dynamical equation (S55) as follows:

$$\dot{c}_{g,1}^{(m)} = \dot{c}_{g,1}^{(m,0)} - g_m^* c_{e,0}(t) + i g_m^* \int_0^t dt' c_{e,0}(t') \left(\Delta_m - i\frac{\gamma^{(m)}}{2} \right) \mathrm{e}^{-i(\Delta_m - i\frac{\gamma^{(m)}}{2})(t-t')} \tag{S64}$$

If we formally integrate equation (S51) and put the result in (S53), it can be shown that

$$c_{g,1}^{(m)}(t) - c_{g,1}^{(m,0)} = -g_m^* \int_0^t dt' c_{e,0}(t') e^{-i(\Delta_m - i\frac{\gamma^{(m)}}{2})(t-t')}, \tag{S65}$$

hence Eq. (S64) becomes

$$\dot{c}_{g,1}^{(m)} = \dot{c}_{g,1}^{(m,0)}(t) - g_m^* c_{e,0}(t) - i \left(\Delta_m - i\frac{\gamma^{(m)}}{2} \right) (c_{g,1}^{(m)}(t) - c_{g,1}^{(m,0)}(t)). \tag{S66}$$

Considering that at initial time $t = 0$ the atom is in its excited state and there is no photon in the cavity, we finally get the following set of dynamical equations

$$\begin{aligned} \dot{c}_{g,1}^{(m)} &= -g_m^* c_{e,0}(t) - i \left(\Delta_m - i\frac{\gamma^{(m)}}{2} \right) c_{g,1}^{(m)}, \\ \dot{c}_{e,0} &= \sum_m g_m c_{g,1}^{(m)}(t), \end{aligned} \tag{S67}$$

which then yields that for the state described by the wavefunction

$$|\psi(t)\rangle = \sum_m c_{g,1}^{(m)} |g, 1_m\rangle + c_{e,0}(t)|e, 0\rangle \tag{S68}$$

the Hamiltonian can be written as

$$\hat{H}_{\text{eff}} = \hbar \sum_m \left(\Delta_m - i\frac{\gamma^{(m)}}{2} \right) \widehat{a}_m^\dagger \widehat{a}_m + i g_m \widehat{\sigma}_+ \widehat{a}_m - i g_m^* \widehat{\sigma}_- \widehat{a}_m^\dagger, \tag{S69}$$

where $\hat{a}_m$ is defined in Eq. (20). Finally, we can write the effective Hamiltonian back from the rotating frame, via the transformation:

$$R = e^{i\omega_A t(\widehat{\sigma}_+ \widehat{\sigma}_- + \sum \widehat{a}_m^\dagger \widehat{a}_m)},$$

leading to an effective Hamiltonian describing the dynamics restricted between the mirrors:

$$\hat{H}_{\text{eff}} = \hbar\omega_A \widehat{\sigma}_+ \widehat{\sigma}_- + \hbar \sum_m \left(\omega^{(m)} - i\frac{\gamma^{(m)}}{2} \right) \widehat{a}_m^\dagger \widehat{a}_m + i g_m \widehat{\sigma}_+ \widehat{a}_m - i g_m^* \widehat{\sigma}_- \widehat{a}_m^\dagger. \tag{S70}$$

For the more general case, when the atom is not necessarily on resonance with the cavity, (i.e. we consider the states $|e, 0\rangle$, $|g, 1\rangle$, as well as $|e, 1\rangle$ and $|g, 0\rangle$), it can be shown that the Hamiltonian (Eq. (S70)) becomes:

$$\hat{H}_{\text{eff}} = \hbar\omega_A \widehat{\sigma}_+ \widehat{\sigma}_- + \hbar \sum_m \left(\omega^{(m)} - i\frac{\gamma^{(m)}}{2} \right) \widehat{a}_m^\dagger \widehat{a}_m + i g_m \widehat{\sigma}_+ \widehat{a}_m - i g_m^* \widehat{\sigma}_- \widehat{a}_m^\dagger + i \underline{g}_m \widehat{\sigma}_- \widehat{a}_m - i \underline{g}_m^* \widehat{\sigma}_+ \widehat{a}\dagger_m, \tag{S71}$$

where

$$\underline{g}_m = i\sqrt{\frac{\omega^{(m)}}{\hbar\varepsilon_0 L^{(m)} \mathscr{A}}} d^* e^{i\frac{\omega^{(m)}}{c}\ell_c} \sin\left[\frac{\omega^{(m)}}{c}(x_A + \ell_c) \right] + \mathscr{O}\left(\varepsilon^2\right). \tag{S72}$$

This non-Hermitian Hamiltonian is a consequence of tracing out the environment to stay only with the dynamics of the cavity.

The expression for g_m (Eq. (S63)) looks similar to the atom-cavity coupling for a perfect cavity, i.e., if we write the perfect cavity Hamiltonian with zero boundary conditions at the mirrors we obtain:

$$g_m = i\sqrt{\frac{\omega_m}{\hbar\varepsilon_0 \ell_c \mathscr{A}}} d\, e^{i\frac{\omega_m}{c}\ell_c} \sin\left[\tfrac{\omega_m}{c}(x_A + \ell_c) \right], \tag{S73}$$

which is different from Eq. (S63) by an error factor $\mathscr{O}\left(\varepsilon^2\right)$, and due to the mismatch between ℓ_c and $L_N^{(m)}$ and ω_m and $\omega_N^{(m)}$.

V. COMPARISON TO USUAL SYSTEMS

The standard model for light-matter interaction in optical cavities considers the mode volume of the cavity to be the product between the longitudinal extent of the mode (considered to be only the separation between the mirrors) and a factor accounting for the transverse behavior of the mode[5]. However, here we have shown that this is only valid if the mirrors can be seen as hard mode-delimiting boundaries. Any physical cavity is very different to that respect. The mode penetrates into the dielectric mirror stack, couples to the outside and the light frequency is not quantised in these open systems. However, even though we don't explicitly consider a mode volume, we show that the atom-cavity coupling involves a factor having the unit of volume. Only in the case of a perfect cavity having hard boundaries at the positions of the mirrors, this factor coincides with the geometric definition of the mode volume. In the multilayered case, however, this factor does not necessarily correspond to the geometric volume of the cavity, particularly due to the discrepency between the geometric length ℓ_c, the effective length ℓ_{eff} and $L_N^{(m)}$. More specifically, for a cavity with a mirror spacing $\ell_c = \lambda_0/2$, the effective cavity length ℓ_{eff} coincides with ℓ_c, however the coupling factor cavity length does not: $L_{19}^{(1)} \approx 3.06\ell_c$. If we increase the mirror spacing, such that $\ell_c = 1.8\lambda_0/2$, then for the other parameters we obtain: $\ell_{\text{eff}} \approx 0.53\ell_c$ and $L_{19}^{(1)} \approx 2.4\ell_c$. As expected, for longer cavities the discrepancy between these terms decreases, e.g., when $\ell_c = 30.8\lambda_0/2$ we obtain $\ell_{\text{eff}} \approx 0.97\ell_c$ and $L_{19}^{(q)} \approx 1.09\ell_c$, where q is the number of the mode with highest amplitude. This discrepancy between the coupling factor cavity length $L_N^{(m)}$ and geometric cavity length ℓ_c can lead to a Purcell factor different from the one calculated by using a standard approach, especially with short mirror spacings.

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