Supplementary Information for: Coherent Polariton Dynamics in Coupled Highly Dissipative Cavities

Yong-Chun Liu1, Xingsheng Luan2, Hao-Kun Li1, Qihuang Gong1, Chee Wei Wong2, and Yun-Feng Xiao1

1State Key Laboratory for Mesoscopic Physics and School of Physics, Peking University; Collaborative Innovation Center of Quantum Matter, Beijing 100871, P. R. China and
2Optical Nanostructures Laboratory, Columbia University, New York, NY 10027, USA

(Dated: May 22, 2014)

This Supplementary Information is organized as follows. In Sec. I, we present the procedures for the derivation of the effective Hamiltonian in detail. In Sec. II, we consider the laser driving and derive the effective driving strength. In Sec. III, we discuss the effect of slight anharmonicity by considering the emitter as an anharmonic oscillator. Based on the derived effective parameters, the parameter ranges for reaching effective strong coupling are presented in Sec. IV. In Sec. V, we calculate the system eigenvalues for the first and second excited states by diagonalizing the non-Hermitian Hamiltonian. In Sec. VI, we study the emitter’s spectrum under weak excitation. The discussion on the mode density shaping is presented in Sec. VII.

Contents

I. Derivation of the effective Hamiltonian 1
II. Effective laser driving 3
III. Effect of slight anharmonicity 4
IV. Parameter ranges 4
V. System eigenvalues 5
VI. Weak excitation spectrum 6
VII. Mode density shaping 6

I. DERIVATION OF THE EFFECTIVE HAMILTONIAN

The original Hamiltonian of emitter-(cavity 1)-(cavity 2) coupling system is given by

\[ H = \omega_1 a_1^\dagger a_1 + \omega_2 a_2^\dagger a_2 + \frac{1}{2} \omega_z \sigma_z + g(a_1^\dagger \sigma_+ + a_1 \sigma_-) + J(a_1^\dagger a_2 + a_2^\dagger a_1). \]  

(1)

where \( \Delta_1 \equiv \omega_1 - \omega_e \) and \( \Delta_2 \equiv \omega_2 - \omega_e \) denote the detunings.

Here, \( a_i \) and \( a_i^\dagger \) (i = 1, 2) are the bosonic annihilation and creation operators of the i-th cavity modes satisfying the commutation relations \([a_i, a_j^\dagger] = \delta_{ij} \); \( \sigma_- \equiv |g\rangle \langle e|, \sigma_+ \equiv |e\rangle \langle g| \) and \( \sigma_z \equiv |e\rangle \langle e| - |g\rangle \langle g| \) are the Pauli operators of the two-level dipole quantum emitter, where \( |g\rangle \) and \( |e\rangle \) are the corresponding ground and excited states, respectively; \( \omega_1, \omega_2, \omega_2, \omega_e \) are the resonance frequencies of mode a1, mode a2, and the emitter; \( g \) and \( J \) denotes the coupling strengths between the emitter and mode a1, and between mode a1 and mode a2, respectively. Here \( g \) and \( J \) are assumed to be real numbers by absorbing the phases into the operators. With the unitary transformation \( U = \exp[-i\omega_e (a_1^\dagger a_1 + a_2^\dagger a_2 + \frac{1}{2} \sigma_z)] \), the system Hamiltonian is given by

\[ H \rightarrow U^\dagger H U - \omega_e (a_1^\dagger a_1 + a_2^\dagger a_2 + \frac{1}{2} \sigma_z) \]

\[ = \Delta_1 a_1^\dagger a_1 + \Delta_2 a_2^\dagger a_2 + g(a_1^\dagger \sigma_+ + a_1 \sigma_-) + J(a_1^\dagger a_2 + a_2^\dagger a_1). \]

(2)

where \( \Delta_1 \equiv \omega_1 - \omega_e \) and \( \Delta_2 \equiv \omega_2 - \omega_e \) denote the detunings.
Starting from Eq. (2), the quantum Langevin equations are given by

\[ \begin{align*}
\dot{a}_1 &= (-i\Delta_1 - \frac{\kappa_1}{2})a_1 - ig\sigma_- - iJa_2 - \sqrt{\kappa_1}a_{in,1}, \\
\dot{a}_2 &= (-i\Delta_2 - \frac{\kappa_2}{2})a_2 - iJa_1 - \sqrt{\kappa_2}a_{in,2}, \\
\dot{\sigma}_- &= -\frac{\gamma}{2}\sigma_- + ig\sigma_+ a_1 - \sqrt{\gamma}\sigma_{in,-},
\end{align*} \tag{3a,b,c} \]

where \( \kappa_1, \kappa_2 \) and \( \gamma \) represent the decay rates of mode \( a_1, a_2 \) and the emitter; \( a_{in,1}, a_{in,2} \) and \( \sigma_{in,-} \) are the noise operators associated with these dissipations. These equations can be formally integrated as

\[ \begin{align*}
a_1(t) &= a_1(0) \exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) + \exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) \\
&\times \int_0^t [-ig\sigma_-(\tau) - iJa_2(\tau) - \sqrt{\kappa_1}a_{in,1}(\tau)] \exp(i\Delta_1\tau + \frac{\kappa_1}{2}\tau) d\tau, \\
a_2(t) &= a_2(0) \exp(-i\Delta_2 t - \frac{\kappa_2}{2}t) + \exp(-i\Delta_2 t - \frac{\kappa_2}{2}t) \int_0^t [-iJa_1(\tau) - \sqrt{\kappa_2}a_{in,2}(\tau)] \exp(i\Delta_2\tau + \frac{\kappa_2}{2}\tau) d\tau, \\
\sigma_-(t) &= \sigma_-(0) \exp(-\frac{\gamma}{2}t) + \exp(-\frac{\gamma}{2}t) \int_0^t [ig\sigma_+(\tau)a_1(\tau) - \sqrt{\gamma}\sigma_{in,1-}(\tau)] \exp(\frac{\gamma}{2}\tau) d\tau,
\end{align*} \tag{4a,b,c} \]

We consider the case of \( |\Delta_1|, \kappa_1 \gg \langle g, J \rangle \), i.e., mode \( a_1 \) is highly dissipative. Then the dynamics of mode \( a_2 \) and the emitter is only slightly affected by mode \( a_1 \). From Eqs. (4b) and (4c) we obtain the approximated expressions

\[ \begin{align*}
a_2(t) &\simeq a_2(0) \exp(-i\Delta_2 t - \frac{\kappa_2}{2}t) + A_{in,2}(t), \\
\sigma_-(t) &\simeq \sigma_-(0) \exp(-\frac{\gamma}{2}t) + \Sigma_{in,-}(t),
\end{align*} \tag{5a,b} \]

where \( A_{in,2}(t) \) and \( \Sigma_{in,-}(t) \) denote the noise terms. By plugging Eqs. (5a) and (5b) into Eq. (4a) we obtain

\[ \begin{align*}
a_1(t) &\simeq a_1(0) \exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) + \exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) \\
&\times \int_0^t [-ig\sigma_-(0) \exp(-\frac{\gamma}{2}\tau) - iJa_2(0) \exp(-i\Delta_2\tau - \frac{\kappa_2}{2}\tau)] \exp(i\Delta_1\tau + \frac{\kappa_1}{2}\tau) d\tau + A_{in,1}(t) \\
&= a_1(0) \exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) + \frac{-ig\sigma_-(0) \exp(-\frac{\gamma}{2}t)}{i\Delta_1 + \frac{\kappa_1 - \gamma}{2}} = \frac{iJ}{i\Delta_1 + \frac{\kappa_1 - \gamma}{2}} a_2(t) + A'_{in,1}(t), \tag{6} \end{align*} \]

where the noise term is denoted by \( A_{in,1}(t) \). By using Eqs. (5a) and (5b) again, and with the condition \( |\Delta_1| \gg |\Delta_2|, \kappa_1 \gg (\kappa_2, \gamma) \) we obtain

\[ a_1(t) \simeq a_1(0) \exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) - \frac{ig}{i\Delta_1 + \frac{\kappa_1 - \gamma}{2}} \sigma_-(t) - \frac{iJ}{i\Delta_1 + \frac{\kappa_1 - \gamma}{2}} a_2(t) + A'_{in,1}(t), \tag{7} \]

where \( A'_{in,1}(t) \) describes the modified noise term. Since \( \kappa_1 \) is large, the term containing \( \exp(-\kappa_1 t/2) \) in Eq. (7) is a fast decaying term and thus can be neglected. Therefore \( a_1(t) \) now can be expressed using \( a_2(t) \) and \( \sigma_-(t) \). Plugging the expression back to Eqs. (3b) and (3c) we finally obtain

\[ \begin{align*}
\dot{a}_2 &= [-i(\Delta_2 - \beta^2\Delta_1)] a_2 - \frac{\kappa_2 + \beta^2\kappa_1}{2} a_2 - i\beta e^{i\theta} g\sigma_- - A'_{in,2}(t), \\
\dot{\sigma}_- &= (i\alpha^2\Delta_1 - \frac{\gamma + \alpha^2\kappa_1}{2}) \sigma_- + i\beta e^{i\theta} g\sigma_+ a_2 - \Sigma'_{in,1-}(t),
\end{align*} \tag{8a,b} \]

where \( A'_{in,2}(t) \) and \( \Sigma'_{in,1-}(t) \) denote the modified noise terms and we have defined the parameters

\[ \alpha = \frac{g}{\sqrt{\Delta_1^2 + \frac{\kappa_1}{4}}} \simeq \frac{g}{|\Delta_1|}, \quad \beta = \frac{J}{\sqrt{\Delta_1^2 + \frac{\kappa_1}{4}}} \simeq \frac{J}{|\Delta_1|}, \tag{9} \]
which holds for $|\Delta_1| \gg \kappa_1$. Here $\alpha$ and $\beta$ are real numbers and the phases $\theta_\beta = \arg(-J/\Delta_1)$ can be absorbed into the operators. In the above derivation we have used the relation $\sigma_+\sigma_- = (|e\rangle\langle e| - |g\rangle\langle g|) |g\rangle\langle e| = -\sigma_-$. From Eqs. (8a) and (8b) we obtain the effective system Hamiltonian

$$H_{\text{eff}} = (\Delta_2 - \beta^2 \Delta_1) a_1^\dagger a_2 - \frac{1}{2} \alpha^2 \Delta_1 \sigma_z + \beta g (a_1^\dagger \sigma_- + a_2 \sigma_+),$$

and we can define the following effective parameters

$$g_{\text{eff}} = \beta g,$$

$$\omega_{2,\text{eff}} = \omega_2 - \beta^2 \Delta_1, \quad \omega_{e,\text{eff}} = \omega_e - \alpha^2 \Delta_1,$$

$$\Delta_{2,\text{eff}} = \Delta_2 - \beta^2 \Delta_1, \quad \Delta_{e,\text{eff}} = -\alpha^2 \Delta_1,$$

$$\Delta_{e,\text{eff}} = \omega_{2,\text{eff}} - \omega_{e,\text{eff}} = \Delta_2 + (\alpha^2 - \beta^2) \Delta_1,$$

$$\kappa_{\text{eff}} = \kappa_2 + \beta^2 \kappa_1, \quad \gamma_{\text{eff}} = \gamma + \alpha^2 \kappa_1.$$

II. EFFECTIVE LASER DRIVING

Consider the laser driving of the cavity modes or the emitter, which can be described by the driving Hamiltonian

$$H_d = \Omega \sigma_+ + E_1 a_1^\dagger + E_2 a_2^\dagger + H.c.,$$

where $\Omega$ and $E_{1,2}$ correspond to the direct driving strengths of the emitter and modes $a_{1,2}$, and we have assumed resonant driving. With the total Hamiltonian $H_d + H_4$, the quantum Langevin equations are given by

$$\dot{a}_1 = (-i \Delta_1 - \frac{\kappa_1}{2}) a_1 - i g \sigma_- - i J a_2 - i E_1 - \sqrt{\kappa_1} \dot{\sigma}_\text{in,1},$$

$$\dot{a}_2 = (-i \Delta_2 - \frac{\kappa_2}{2}) a_2 - i J a_1 - i E_2 - \sqrt{\kappa_2} \dot{\sigma}_\text{in,2},$$

$$\dot{\sigma}_- = -\frac{\gamma}{2} \sigma_- + i g \sigma_+ a_1 + i \Omega \sigma_z - \sqrt{\gamma} \dot{\sigma}_\text{in,-}.$$

Perform the same procedure as the previous section and we obtain

$$\dot{a}_2 = [-i(\Delta_2 - \beta^2 \Delta_1) - \frac{\kappa_2 + \beta^2 \kappa_1}{2}] a_2 - i \beta e^{i \theta_\beta} g \sigma_- - i(E_2 + \beta e^{i \theta_\beta} E_1) - \dot{\sigma}_\text{in,2}(t),$$

$$\dot{\sigma}_- = (i \alpha^2 \Delta_1 - \frac{\gamma + \alpha^2 \kappa_1}{2}) \sigma_- + i \beta e^{i \theta_\beta} g \sigma_+ a_2 + i(\Omega + \alpha e^{i \theta_\alpha} E_1) \sigma_z - \dot{\sigma}_\text{in,-}(t).$$

In the above derivation, it requires that the dynamics of the emitter is much slower than mode $a_1$ with the time scale $\sim 1/\sqrt{\Delta_1^2 + \kappa_1^2/4}. Thus the pumping limit of the emitter is given by

$$\Omega \ll \sqrt{\Delta_1^2 + \kappa_1^2/4} \simeq |\Delta_1|.$$  

Under this condition, the effective driving Hamiltonian reads

$$H_{d,\text{eff}} = \Omega_{\text{eff}} \sigma_+ + E_{2,\text{eff}} a_2^\dagger + H.c.,$$

with the effective driving strength

$$E_{2,\text{eff}} = E_2 + \beta e^{i \theta_\beta} E_1,$$

$$\Omega_{\text{eff}} = \Omega + \alpha e^{i \theta_\alpha} E_1,$$

where $e^{i \theta_\beta}$ and $e^{i \theta_\alpha}$ are phase factors with $\theta_\beta = \arg(-J/\Delta_1)$ and $\theta_\alpha = \arg(-g/\Delta_1)$. Therefore, the driving of mode $a_1$ will effectively drive both mode $a_2$ and the emitter after the elimination of mode $a_1$.
III. EFFECT OF SLIGHT ANHARMONICITY

Consider a slightly anharmonic oscillator, in which the higher energy levels should be taken into account. The anharmonic oscillator can be described using the Hamiltonian $H_{\text{osc}} = \omega^2 b^2 b^\dagger + \lambda (b + b^\dagger)^2$, where $\lambda$ is the anharmonicity parameter. In the frame rotating at the frequency $\omega$, the system Hamiltonian is given by

$$H_{\text{an}} = \Delta_1 a_1^\dagger a_1 + \Delta_2 a_2^\dagger a_2 + \lambda (b + b^\dagger)^2 + g(a_1^\dagger b + a_1 b^\dagger) + J(a_1^\dagger a_2 + a_2^\dagger a_1).$$  \hfill (18)

The quantum Langevin equations are given by

$$\dot{a}_1 = (-i\Delta_1 - \frac{\kappa_1}{2}) a_1 - ig b - i J a_2 - \sqrt{\kappa_1} a_{\text{in},1},$$  \hfill (19a)

$$\dot{a}_2 = (-i\Delta_2 - \frac{\kappa_2}{2}) a_2 - i J a_1 - \sqrt{\kappa_2} a_{\text{in},2},$$  \hfill (19b)

$$\dot{b} = -2i \lambda (b + b^\dagger) - \frac{\gamma}{2} b - i g a_1 - \sqrt{\gamma} b_{\text{in}}.$$  \hfill (19c)

From Eq. (19c), by neglecting the effect of highly dissipative mode $a_1$, we obtain

$$b(t) \simeq b(0) \exp(-\frac{\gamma}{2} t) - 2i \lambda [b(0) + b^\dagger(0)] t \exp(-\frac{\gamma}{2} t) + B_{\text{in}}(t),$$  \hfill (20)

where $B_{\text{in}}$ denotes the noise term. By performing the same procedure as the first section, we obtain

$$a_1(t) \simeq -\frac{ig}{\Delta_1 + \frac{\kappa_1}{2}} b(t) - \frac{iJ}{\Delta_1 + \frac{\kappa_1}{2}} a_2(t) + \frac{2\lambda g}{(i\Delta_1 + \frac{\kappa_1}{2})^2} [b(t) + b^\dagger(t)] + A_{\text{in},1}(t).$$  \hfill (21)

Note that the third term originates from the anharmonicity of the oscillator. This term can be neglected under the condition

$$\lambda \ll \sqrt{\Delta_1^2 + \kappa_1^2/4} \simeq |\Delta_1|. \hfill (22)$$

In this case the quantum Langevin equations after eliminating mode $a_1$ are obtained as

$$\dot{a}_2 = [-i(\Delta_2 - \beta^2 \Delta_1) - \frac{\kappa_2 + \beta^2 \kappa_1}{2}] a_2 - i\beta e^{i\theta_0} gb - A_{\text{in},2}(t),$$  \hfill (23a)

$$\dot{b} = -2i \lambda (b + b^\dagger) + (i\alpha^2 \Delta_1 - \frac{\gamma + \alpha^2 \kappa_1}{2}) b - i\beta e^{i\theta_0} ga_2 - B_{\text{in}}(t).$$  \hfill (23b)

Then the effective Hamiltonian is given by

$$H_{\text{an,eff}} = (\Delta_2 - \beta^2 \Delta_1) a_2^\dagger a_2 - \alpha^2 \Delta_1 b^\dagger b + \lambda (b + b^\dagger)^2 + \beta g(a_2^\dagger b + a_2 b^\dagger).$$  \hfill (24)

IV. PARAMETER RANGES

Based on the effective parameters, to obtain effective strong coupling, it requires $g_{\text{eff}} > (\kappa_{\text{eff}}, \gamma_{\text{eff}})$, yielding

$$\beta g > \kappa_2 + \beta^2 \kappa_1,$$  \hfill (25a)

$$\alpha J > \gamma + \alpha^2 \kappa_1.$$  \hfill (25b)

Then we obtain

$$g - \sqrt{g^2 - 4\kappa_1 \kappa_2} < \eta J < g + \sqrt{g^2 - 4\kappa_1 \kappa_2},$$  \hfill (26a)

$$J - \sqrt{J^2 - 4\kappa_1 \gamma} < \eta g < J + \sqrt{J^2 - 4\kappa_1 \gamma},$$  \hfill (26b)

where

$$\eta = \frac{2\kappa_1}{\sqrt{\Delta_1^2 + \kappa_1^2/4}} \simeq \frac{2\kappa_1}{|\Delta_1|}.$$  \hfill (27)
Thus the necessary conditions for effective strong coupling read
\[ g^2 > 4\kappa_1\kappa_2, \]  
\[ J^2 > 4\kappa_1\gamma. \]  

From Eq. (26b) we further obtain
\[ J > \frac{\eta^2 g^2 + 4\kappa_1\gamma}{2\eta g}. \]  
Then the parameter range for \( J \) is determined by Eqs. (26a) and (29). To ensure that the effective strong coupling can be achieved for large ranges of \( J \), it requires
\[ \frac{\eta^2 g^2 + 4\kappa_1\gamma}{2\eta g} < g + \sqrt{g^2 - 4\kappa_1\kappa_2}, \]  
which yields
\[ \eta^2 < 2 - 4\kappa_1\gamma/g^2 + 2\sqrt{1 - 4\kappa_1\kappa_2/g^2}. \]

Then the lower bound for \(|\Delta_1|\) is approximately given by
\[ |\Delta_1| > \kappa_1. \]  
The upper bound for \(|\Delta_1|\) can be obtained by considering \( \beta g > \kappa_2 \) and \( \alpha J > \gamma \) from Eq. (25), which gives
\[ |\Delta_1| < \left( \frac{g}{\kappa_2}, \frac{Jg}{\gamma} \right). \]

V. SYSTEM EIGENVALUES

To obtain the system eigenvalues, we consider the non-Hermitian Hamiltonian
\[ H_{\text{nonH}} = (\Delta_1 - i\frac{\kappa_1}{2})a_1^\dagger a_1 + (\Delta_2 - i\frac{\kappa_2}{2})a_2^\dagger a_2 - i\frac{\gamma}{2}|e\rangle\langle e| + g(a_1^\dagger \sigma_- + a_1 \sigma_+) + J(a_1^\dagger a_2 + a_2^\dagger a_1). \]  

For the first excited state, the uncoupled bases are \(|g\rangle|1\rangle_1|0\rangle_2\), \(|g\rangle|0\rangle_1|1\rangle_2\) and \(|e\rangle|0\rangle_1|0\rangle_2\). Under these bases, the matrix form of the Hamiltonian in the first-excited-state subspace is given by
\[ H_{\text{nonH}}^{(1)} = \begin{pmatrix} \Delta_1 - i\kappa_1 \kappa_2 & J & g \\ J & \Delta_2 - i\kappa_2 & 0 \\ g & 0 & -i\frac{\gamma}{2} \end{pmatrix}. \]  

Then the eigenvalues of \( H_{\text{nonH}}^{(1)} \) can be calculated straightforwardly, but the analytical expressions are tedious and not presented here. From the effective non-Hermitian Hamiltonian
\[ H_{\text{eff, nonH}} = (\Delta_2^{\text{eff}} - i\kappa^{\text{eff}}/2)\sigma_2^\dagger a_2 + (\Delta^{\text{eff}} - i\gamma^{\text{eff}}/2)|e\rangle\langle e| + g^{\text{eff}}(a_1^\dagger \sigma_- + a_2 \sigma_+), \]

with the uncoupled bases \(|g\rangle|1\rangle_2\) and \(|e\rangle|0\rangle_2\), we obtain the matrix form of the effective Hamiltonian in the first-excited-state subspace as
\[ H_{\text{eff, nonH}}^{(1)} = \begin{pmatrix} \Delta^{\text{eff}} - i\kappa^{\text{eff}}/2 & g^{\text{eff}} \\ g^{\text{eff}} & \Delta_{\text{eff}}^\dagger - i\gamma^{\text{eff}}/2 \end{pmatrix}. \]  

The eigenvalues of \( H_{\text{eff, nonH}}^{(1)} \) are given by
\[ E_{\text{eff}}^{(1)} = \Delta_{\text{eff}}^\dagger + \frac{1}{2}(\Delta^{\text{eff}} - i\kappa^{\text{eff}} + \gamma^{\text{eff}}) \pm \frac{1}{2} \sqrt{(\Delta^{\text{eff}} - i\kappa^{\text{eff}} - \gamma^{\text{eff}})^2 + 4g_{\text{eff}}^2} \]
\[ = -\alpha^2\Delta_1 + \frac{1}{2}[\Delta_2 + (\alpha^2 - \beta^2)\Delta_1 - i\kappa_2 + \gamma + (\beta^2 + \alpha^2)\kappa_1] \]
\[ \pm \frac{1}{2} \sqrt{[\Delta_2 + (\alpha^2 - \beta^2)\Delta_1 - i\kappa_2 - \gamma + (\beta^2 - \alpha^2)\kappa_1]^2 + 4\beta^2 g^2}. \]
Now we calculate the shaped mode density of the supermode. The Hamiltonian of the coupled cavity system is given by

\[
H^{(2)}_{\text{nonH}} = \begin{pmatrix}
2\Delta_1 - i\kappa_1 & \sqrt{2}J & \sqrt{2}g & 0 & 0 \\
\sqrt{2}J & \Delta_1 + \Delta_2 - i\kappa_1 + \kappa_2 & \sqrt{2}g & 0 & g \\
\sqrt{2}g & \sqrt{2}J & \Delta_1 - i\kappa_1 + \kappa_2 & 0 & 0 \\
0 & \sqrt{2}J & \Delta_2 - i\kappa_2 & 0 & 0 \\
0 & 0 & 2\Delta_2 - i\kappa_2 & 0 & 0 \\
\end{pmatrix}.
\]

The corresponding effective Hamiltonian is given by

\[
H^{(2)}_{\text{eff, nonH}} = \begin{pmatrix}
2\Delta_{2,\text{eff}} - i\kappa_{\text{eff}} & \sqrt{2g_{\text{eff}}} & \Delta_{2,\text{eff}} + \Delta_{e,\text{eff}} - i\frac{\kappa_{\text{eff}} + \gamma_{\text{eff}}}{2} \\
\sqrt{2g_{\text{eff}}} & \Delta_{2,\text{eff}} + i\kappa_{\text{eff}} - \gamma_{\text{eff}} & 0 & 0 & 0 \\
\Delta_{2,\text{eff}} + \Delta_{e,\text{eff}} - i\frac{\kappa_{\text{eff}} + \gamma_{\text{eff}}}{2} & 0 & 2\Delta_{2,\text{eff}} - i\kappa_{\text{eff}} & 0 & 0 \\
0 & 0 & 2\Delta_{2,\text{eff}} - i\kappa_{\text{eff}} & 0 & 0 \\
0 & 0 & 0 & 2\Delta_{2,\text{eff}} - i\kappa_{\text{eff}} & 0 \\
\end{pmatrix}.
\]

VI. WEAK EXCITATION SPECTRUM

Under weak excitation, the emitter is predominantly in the ground state. Thus \(\sigma_2\) can be substituted for its average value of \(-1\), and the quantum Langevin equations (3a)-(3c) becomes linear. In the frequency domain, the equations are given by

\[
-\omega \tilde{a}_1(\omega) = (-i\Delta_1 - \frac{\kappa_1}{2}) \tilde{a}_1(\omega) - ig\tilde{\sigma}_-(\omega) - iJ \tilde{a}_2(\omega) - \sqrt{\kappa_1} \tilde{a}_{\text{in},1}(\omega),
\]

\[
-\omega \tilde{a}_2(\omega) = (-i\Delta_2 - \frac{\kappa_2}{2}) \tilde{a}_2(\omega) - iJ \tilde{a}_1(\omega) - \sqrt{\kappa_2} \tilde{a}_{\text{in},2}(\omega),
\]

\[
-\omega \tilde{\sigma}_-(\omega) = -\frac{\gamma}{2} \tilde{\sigma}_-(\omega) - ig \tilde{a}_1(\omega) - \sqrt{\gamma} \tilde{\sigma}_{\text{in},-}(\omega).
\]

Then \(\tilde{a}_1(\omega), \tilde{a}_2(\omega)\) and \(\tilde{\sigma}_-(\omega)\) can be solved, and the emitter’s spectrum can be obtained as \(S(\omega) = \int \tilde{\sigma}_+(\omega) \tilde{\sigma}_-(\omega') d\omega'\).

In the effective picture, the equations are

\[
-\omega \tilde{a}_2(\omega) = (-i\Delta_{\text{eff}} - \frac{\kappa_{\text{eff}}}{2}) \tilde{a}_2(\omega) - ig \tilde{\sigma}_-(\omega) - \sqrt{\kappa_{\text{eff}}} \tilde{a}_{\text{in},2}(\omega),
\]

\[
-\omega \tilde{\sigma}_-(\omega) = -\frac{\gamma_{\text{eff}}}{2} \tilde{\sigma}_-(\omega) - ig \tilde{a}_2(\omega) - \sqrt{\gamma_{\text{eff}}} \tilde{\sigma}_{\text{in},-}(\omega),
\]

and the emitter’s effective spectrum can be obtained with \(\tilde{\sigma}_-(\omega)\) solved from these two equations.

VII. MODE DENSITY SHAPING

In the viewpoint of mode density shaping, we can consider the cavity modes \(a_1\) and \(a_2\) as a whole, which form a supermode. Now we calculate the shaped mode density of the supermode. The Hamiltonian of the coupled cavity system is given by

\[
H_{\text{cav}} = \omega_1 a_1^\dagger a_1 + \omega_2 a_2^\dagger a_2 + J(a_1^\dagger a_2 + a_2^\dagger a_1).
\]

The quantum Langevin equations are given by

\[
\dot{a}_1 = (-i\omega - \frac{\kappa_1}{2}) a_1 - iJ a_2 - \sqrt{\kappa_1} a_{\text{in},1},
\]

\[
\dot{a}_2 = (-i\omega - \frac{\kappa_2}{2}) a_2 - iJ a_1 - \sqrt{\kappa_2} a_{\text{in},2}.
\]
Fig. S1: Normalized spectrum of the coupled cavity system for $\kappa_2/\kappa_1 = 10^{-2}$, $J/\kappa_1 = 0.2$ and $\Delta_{12}/\kappa_1 = 2$.

In the frequency domain, the equations are given by

\begin{align}
- i \omega \hat{a}_1(\omega) &= (-i \omega_1 - \frac{\kappa_1}{2})\hat{a}_1(\omega) - i J \hat{a}_2(\omega) - \sqrt{\kappa_1} \hat{a}_{in,1}(\omega), \\
- i \omega \hat{a}_2(\omega) &= (-i \omega_2 - \frac{\kappa_2}{2})\hat{a}_2(\omega) - i J \hat{a}_1(\omega) - \sqrt{\kappa_2} \hat{a}_{in,2}(\omega).
\end{align}

(46a) (46b)

Then we obtain

\begin{equation}
\hat{a}_1(\omega) = \frac{(i \omega - i \omega_2 - \frac{\kappa_2}{2}) \sqrt{\kappa_1} \hat{a}_{in,1}(\omega) + i J \sqrt{\kappa_2} \hat{a}_{in,2}(\omega)}{(i \omega - i \omega_1 - \frac{\kappa_1}{2})(i \omega - i \omega_2 - \frac{\kappa_2}{2}) + J^2}.
\end{equation}

(47)

The spectrum of the coupled-cavity supermode is given by $S_{cav}(\omega) = \int \hat{a}_1^\dagger(\omega)\hat{a}_1(\omega')d\omega'$. In Fig. S1 we plot the normalized spectrum for $\kappa_2/\kappa_1 = 10^{-2}$, $J/\kappa_1 = 0.2$ and $\Delta_{12}/\kappa_1 = 2$. It shows that the spectrum has Fano-type lineshape. Near the Fano lineshape region the mode density is the combined effect of the two cavity modes.