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

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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 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

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

$$H = \omega_1 a_1^\dagger a_1 + \omega_2 a_2^\dagger a_2 + \frac{1}{2}\omega_\mathrm{e}\sigma_\mathrm{z} + g(a_1^\dagger \sigma_- + a_1\sigma_+) + J(a_1^\dagger a_2 + a_2^\dagger a_1). \tag{1}$$

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 $[a_i, a_j^\dagger] = \delta_{ij}$; $\sigma_- \equiv |\mathrm{g}\rangle\langle \mathrm{e}|$, $\sigma_+ \equiv |\mathrm{e}\rangle\langle \mathrm{g}|$ and $\sigma_\mathrm{z} \equiv |\mathrm{e}\rangle\langle \mathrm{e}| - |\mathrm{g}\rangle\langle \mathrm{g}|$ are the Pauli operators of the two-level dipole quantum emitter, where $|\mathrm{g}\rangle$ and $|\mathrm{e}\rangle$ are the corresponding ground and excited states, respectively; ω_1, ω_2 and ω_e are the resonance frequencies of mode a_1, mode a_2 and the emitter; g and J denotes the coupling strengths between the emitter and mode a_1, and between mode a_1 and mode a_2, respectively. Here g and J are assumed to be real numbers by absorbing the phases into the operators. With the unitary transformation $U = \exp[-i\omega_\mathrm{e}(a_1^\dagger a_1 + a_2^\dagger a_2 + \frac{1}{2}\sigma_\mathrm{z})t]$, the system Hamiltonian is given by

$$\begin{aligned} H &\to U^\dagger H U - \omega_\mathrm{e}(a_1^\dagger a_1 + a_2^\dagger a_2 + \frac{1}{2}\sigma_\mathrm{z}) \\ &= \Delta_1 a_1^\dagger a_1 + \Delta_2 a_2^\dagger a_2 + g(a_1^\dagger \sigma_- + a_1\sigma_+) + J(a_1^\dagger a_2 + a_2^\dagger a_1), \end{aligned} \tag{2}$$

where $\Delta_1 \equiv \omega_1 - \omega_\mathrm{e}$ and $\Delta_2 \equiv \omega_2 - \omega_\mathrm{e}$ denote the detunings.

Starting from Eq. (2), the quantum Langevin equations are given by

$$\dot{a}_1 = (-i\Delta_1 - \frac{\kappa_1}{2})a_1 - ig\sigma_- - iJa_2 - \sqrt{\kappa_1}a_{\text{in},1}, \tag{3a}$$

$$\dot{a}_2 = (-i\Delta_2 - \frac{\kappa_2}{2})a_2 - iJa_1 - \sqrt{\kappa_2}a_{\text{in},2}, \tag{3b}$$

$$\dot{\sigma}_- = -\frac{\gamma}{2}\sigma_- + ig\sigma_z a_1 - \sqrt{\gamma}\sigma_{\text{in},-}, \tag{3c}$$

where κ_1, κ_2 and γ represent the decay rates of mode a_1, a_2 and the emitter; $a_{\text{in},1}$, $a_{\text{in},2}$ and $\sigma_{\text{in},-}$ are the noise operators associated with these dissipations. These equations can be formally integrated as

$$a_1(t) = a_1(0)\exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) + \exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) \times \int_0^t [-ig\sigma_-(\tau) - iJa_2(\tau) - \sqrt{\kappa_1}a_{\text{in},1}(\tau)]\exp(i\Delta_1\tau + \frac{\kappa_1}{2}\tau)d\tau, \tag{4a}$$

$$a_2(t) = a_2(0)\exp(-i\Delta_2 t - \frac{\kappa_2}{2}t) + \exp(-i\Delta_2 t - \frac{\kappa_2}{2}t)\int_0^t [-iJa_1(\tau) - \sqrt{\kappa_2}a_{\text{in},2}(\tau)]\exp(i\Delta_2\tau + \frac{\kappa_2}{2}\tau)d\tau, \tag{4b}$$

$$\sigma_-(t) = \sigma_-(0)\exp(-\frac{\gamma}{2}t) + \exp(-\frac{\gamma}{2}t)\int_0^t [ig\sigma_z(\tau)a_1(\tau) - \sqrt{\gamma}\sigma_{\text{in},-}(\tau)]\exp(\frac{\gamma}{2}\tau)d\tau, \tag{4c}$$

We consider the case of $(|\Delta_1|, \kappa_1) \gg (g, J)$, 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

$$a_2(t) \simeq a_2(0)\exp(-i\Delta_2 t - \frac{\kappa_2}{2}t) + A_{\text{in},2}(t), \tag{5a}$$

$$\sigma_-(t) \simeq \sigma_-(0)\exp(-\frac{\gamma}{2}t) + \Sigma_{\text{in},-}(t), \tag{5b}$$

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

$$\begin{aligned} a_1(t) &\simeq a_1(0)\exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) + \exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) \\ &\quad \times \int_0^t [-ig\sigma_-(0)\exp(-\frac{\gamma}{2}\tau) - iJa_2(0)\exp(-i\Delta_2\tau - \frac{\kappa_2}{2}\tau)]\exp(i\Delta_1\tau + \frac{\kappa_1}{2}\tau)d\tau + A_{\text{in},1}(t) \\ &= a_1(0)\exp(-i\Delta_1 t - \frac{\kappa_1}{2}t) + \frac{-ig\sigma_-(0)\exp(-\frac{\gamma}{2}t)}{i\Delta_1 + \frac{\kappa_1-\gamma}{2}} - \frac{iJa_2(0)\exp(-i\Delta_2 t - \frac{\kappa_2}{2}t)}{i(\Delta_1-\Delta_2) + \frac{\kappa_1-\kappa_2}{2}} + A_{\text{in},1}(t), \end{aligned} \tag{6}$$

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

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

where $A'_{\text{in},1}(t)$ describes the modified noise term. Since κ_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

$$\dot{a}_2 = [-i(\Delta_2 - \beta^2\Delta_1) - \frac{\kappa_2 + \beta^2\kappa_1}{2}]a_2 - i\beta e^{i\theta_\beta}g\sigma_- - A'_{\text{in},2}(t), \tag{8a}$$

$$\dot{\sigma}_- = (i\alpha^2\Delta_1 - \frac{\gamma + \alpha^2\kappa_1}{2})\sigma_- + i\beta e^{i\theta_\beta}g\sigma_z a_2 - \Sigma'_{\text{in},-}(t), \tag{8b}$$

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

$$\alpha = \frac{g}{\sqrt{\Delta_1^2 + \frac{\kappa_1^2}{4}}} \simeq \frac{g}{|\Delta_1|}, \quad \beta = \frac{J}{\sqrt{\Delta_1^2 + \frac{\kappa_1^2}{4}}} \simeq \frac{J}{|\Delta_1|}, \tag{9}$$

which holds for $|\Delta_1| \gg \kappa_1$. Here α and β 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 $\sigma_z\sigma_- = (|\mathrm{e}\rangle\langle\mathrm{e}| - |\mathrm{g}\rangle\langle\mathrm{g}|)\,|\mathrm{g}\rangle\langle\mathrm{e}| = -\sigma_-$. From Eqs. (8a) and (8b) we obtain the effective system Hamiltonian

$$H_{\mathrm{eff}} = \left(\Delta_2 - \beta^2\Delta_1\right) a_2^\dagger a_2 - \frac{1}{2}\alpha^2\Delta_1\sigma_z + \beta g(a_2^\dagger\sigma_- + a_2\sigma_+), \tag{10}$$

and we can define the following effective parameters

$$\begin{aligned}
g_{\mathrm{eff}} &= \beta g, && (11a)\\
\omega_{2,\mathrm{eff}} &= \omega_2 - \beta^2\Delta_1, \;\; \omega_{\mathrm{e,eff}} = \omega_{\mathrm{e}} - \alpha^2\Delta_1, && (11b)\\
\Delta_{2,\mathrm{eff}} &= \Delta_2 - \beta^2\Delta_1, \;\; \Delta_{\mathrm{e,eff}} = -\alpha^2\Delta_1, && (11c)\\
\Delta_{\mathrm{eff}} &= \omega_{2,\mathrm{eff}} - \omega_{\mathrm{e,eff}} = \Delta_2 + \left(\alpha^2 - \beta^2\right)\Delta_1, && (11d)\\
\kappa_{\mathrm{eff}} &= \kappa_2 + \beta^2\kappa_1, \;\; \gamma_{\mathrm{eff}} = \gamma + \alpha^2\kappa_1. && (11e)
\end{aligned}$$

II. EFFECTIVE LASER DRIVING

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

$$H_{\mathrm{d}} = \Omega\sigma_+ + E_1 a_1^\dagger + E_2 a_2^\dagger + H.c., \tag{12}$$

where Ω 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 $H + H_{\mathrm{d}}$, the quantum Langevin equations are given by

$$\begin{aligned}
\dot{a}_1 &= (-i\Delta_1 - \frac{\kappa_1}{2})a_1 - ig\sigma_- - iJa_2 - iE_1 - \sqrt{\kappa_1}a_{\mathrm{in},1}, && (13a)\\
\dot{a}_2 &= (-i\Delta_2 - \frac{\kappa_2}{2})a_2 - iJa_1 - iE_2 - \sqrt{\kappa_2}a_{\mathrm{in},2}, && (13b)\\
\dot{\sigma}_- &= -\frac{\gamma}{2}\sigma_- + ig\sigma_{\mathrm{z}}a_1 + i\Omega\sigma_{\mathrm{z}} - \sqrt{\gamma}\sigma_{\mathrm{in},-}, && (13c)
\end{aligned}$$

Perform the same procedure as the previous section and we obtain

$$\begin{aligned}
\dot{a}_2 &= [-i(\Delta_2 - \beta^2\Delta_1) - \frac{\kappa_2 + \beta^2\kappa_1}{2}]a_2 - i\beta e^{i\theta_\beta}g\sigma_- - i(E_2 + \beta e^{i\theta_\beta}E_1) - A'_{\mathrm{in},2}(t), && (14a)\\
\dot{\sigma}_- &= (i\alpha^2\Delta_1 - \frac{\gamma + \alpha^2\kappa_1}{2})\sigma_- + i\beta e^{i\theta_\beta}g\sigma_{\mathrm{z}}a_2 + i(\Omega + \alpha e^{i\theta_\alpha}E_1)\sigma_{\mathrm{z}} - \Sigma'_{\mathrm{in},-}(t). && (14b)
\end{aligned}$$

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

$$\Omega \ll \sqrt{\Delta_1^2 + \kappa_1^2/4} \simeq |\Delta_1|. \tag{15}$$

Under this condition, the effective driving Hamiltonian reads

$$H_{\mathrm{d,eff}} = \Omega_{\mathrm{eff}}\sigma_+ + E_{2,\mathrm{eff}}a_2^\dagger + H.c., \tag{16}$$

with the effective driving strength

$$\begin{aligned}
E_{2,\mathrm{eff}} &= E_2 + \beta e^{i\theta_\beta}E_1, && (17a)\\
\Omega_{\mathrm{eff}} &= \Omega + \alpha e^{i\theta_\alpha}E_1, && (17b)
\end{aligned}$$

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 $H_{\rm an}^{\rm osc} = \omega_{\rm e} b^\dagger b + \lambda(b+b^\dagger)^2$, where λ is the anharmonicity parameter. In the frame rotating at the frequency $\omega_{\rm e}$, the system Hamiltonian is given by

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

The quantum Langevin equations are given by

$$\dot{a}_1 = (-i\Delta_1 - \frac{\kappa_1}{2})a_1 - igb - iJa_2 - \sqrt{\kappa_1}a_{\rm in,1}, \tag{19a}$$

$$\dot{a}_2 = (-i\Delta_2 - \frac{\kappa_2}{2})a_2 - iJa_1 - \sqrt{\kappa_2}a_{\rm in,2}, \tag{19b}$$

$$\dot{b} = -2i\lambda(b+b^\dagger) - \frac{\gamma}{2}b - iga_1 - \sqrt{\gamma}b_{\rm in}. \tag{19c}$$

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

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

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

$$a_1(t) \simeq -\frac{ig}{i\Delta_1 + \frac{\kappa_1}{2}}b(t) - \frac{iJ}{i\Delta_1 + \frac{\kappa_1}{2}}a_2(t) + \frac{2\lambda g}{(i\Delta_1 + \frac{\kappa_1}{2})^2}[b(t) + b^\dagger(t)] + A'_{\rm in,1}(t). \tag{21}$$

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

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

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

$$\dot{a}_2 = [-i(\Delta_2 - \beta^2\Delta_1) - \frac{\kappa_2 + \beta^2\kappa_1}{2}]a_2 - i\beta e^{i\theta_\beta}gb - A'_{\rm in,2}(t), \tag{23a}$$

$$\dot{b} = -2i\lambda(b+b^\dagger) + (i\alpha^2\Delta_1 - \frac{\gamma + \alpha^2\kappa_1}{2})b - i\beta e^{i\theta_\beta}ga_2 - B'_{\rm in}(t). \tag{23b}$$

Then the effective Hamiltonian is given by

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

IV. PARAMETER RANGES

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

$$\beta g > \kappa_2 + \beta^2\kappa_1, \tag{25a}$$

$$\alpha J > \gamma + \alpha^2\kappa_1. \tag{25b}$$

Then we obtain

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

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

where

$$\eta = \frac{2\kappa_1}{\sqrt{\Delta_1^2 + \frac{\kappa_1^2}{4}}} \simeq \frac{2\kappa_1}{|\Delta_1|}. \tag{27}$$

Thus the necessary conditions for effective strong coupling read

$$g^2 > 4\kappa_1\kappa_2, \tag{28a}$$

$$J^2 > 4\kappa_1\gamma. \tag{28b}$$

From Eq. (26b) we further obtain

$$J > \frac{\eta^2 g^2 + 4\kappa_1\gamma}{2\eta g}. \tag{29}$$

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

$$\frac{\eta^2 g^2 + 4\kappa_1\gamma}{2\eta g} < \frac{g + \sqrt{g^2 - 4\kappa_1\kappa_2}}{\eta}, \tag{30}$$

which yields

$$\eta^2 < 2 - 4\frac{\kappa_1\gamma}{g^2} + 2\sqrt{1 - 4\frac{\kappa_1\kappa_2}{g^2}}. \tag{31}$$

Then the lower bound for $|\Delta_1|$ is approximately given by

$$|\Delta_1| > \kappa_1. \tag{32}$$

The upper bound for $|\Delta_1|$ can be obtained by considering $\beta g > \kappa_2$ and $\alpha J > \gamma$ from Eq. (25), which gives

$$|\Delta_1| < (\frac{Jg}{\kappa_2}, \frac{Jg}{\gamma}). \tag{33}$$

V. SYSTEM EIGENVALUES

To obtain the system eigenvalues, we consider the non-Hermitian Hamiltonian

$$H_{\mathrm{nonH}} = (\Delta_1 - i\frac{\kappa_1}{2})a_1^\dagger a_1 + (\Delta_2 - i\frac{\kappa_2}{2})a_2^\dagger a_2 - i\frac{\gamma}{2}|\mathrm{e}\rangle\langle\mathrm{e}| + g(a_1^\dagger\sigma_- + a_1\sigma_+) + J(a_1^\dagger a_2 + a_2^\dagger a_1). \tag{34}$$

For the first excited state, the uncoupled bases are $|\mathrm{g}\rangle|1\rangle_1|0\rangle_2$, $|\mathrm{g}\rangle|0\rangle_1|1\rangle_2$ and $|\mathrm{e}\rangle|0\rangle_1|0\rangle_2$. Under these bases, the matrix form of the Hamiltonian in the first-excited-state subspace is given by

$$H_{\mathrm{nonH}}^{(1)} = \begin{pmatrix} \Delta_1 - i\frac{\kappa_1}{2} & J & g \\ J & \Delta_2 - i\frac{\kappa_2}{2} & 0 \\ g & 0 & -i\frac{\gamma}{2} \end{pmatrix}. \tag{35}$$

Then the eigenvalues of $H_{\mathrm{nonH}}^{(1)}$ can be calculated straightforwardly, but the analytical expressions are tedious and not presented here. From the effective non-Hermitian Hamiltonian

$$H_{\mathrm{eff,nonH}} = (\Delta_{2,\mathrm{eff}} - i\frac{\kappa_{\mathrm{eff}}}{2})a_2^\dagger a_2 + (\Delta_{\mathrm{e,eff}} - i\frac{\gamma_{\mathrm{eff}}}{2})|\mathrm{e}\rangle\langle\mathrm{e}| + g_{\mathrm{eff}}(a_2^\dagger\sigma_- + a_2\sigma_+), \tag{36}$$

with the uncoupled bases $|\mathrm{g}\rangle|1\rangle_2$ and $|\mathrm{e}\rangle|0\rangle_2$, we obtain the matrix form of the effective Hamiltonian in the first-excited-state subspace as

$$H_{\mathrm{eff,nonH}}^{(1)} = \begin{pmatrix} \Delta_{2,\mathrm{eff}} - i\frac{\kappa_{\mathrm{eff}}}{2} & g_{\mathrm{eff}} \\ g_{\mathrm{eff}} & \Delta_{\mathrm{e,eff}} - i\frac{\gamma_{\mathrm{eff}}}{2} \end{pmatrix}. \tag{37}$$

The eigenvalues of $H_{\mathrm{eff,nonH}}^{(1)}$ are given by

$$\begin{aligned} E_{\pm}^{(1)} &= \Delta_{\mathrm{e,eff}} + \frac{1}{2}(\Delta_{\mathrm{eff}} - i\frac{\kappa_{\mathrm{eff}} + \gamma_{\mathrm{eff}}}{2}) \pm \frac{1}{2}\sqrt{(\Delta_{\mathrm{eff}} - i\frac{\kappa_{\mathrm{eff}} - \gamma_{\mathrm{eff}}}{2})^2 + 4g_{\mathrm{eff}}^2} \\ &= -\alpha^2\Delta_1 + \frac{1}{2}[\Delta_2 + \left(\alpha^2 - \beta^2\right)\Delta_1 - i\frac{\kappa_2 + \gamma + (\beta^2 + \alpha^2)\kappa_1}{2}] \\ &\pm \frac{1}{2}\sqrt{[\Delta_2 + (\alpha^2 - \beta^2)\,\Delta_1 - i\frac{\kappa_2 - \gamma + (\beta^2 - \alpha^2)\kappa_1}{2}]^2 + 4\beta^2 g^2}. \end{aligned} \tag{38}$$

For the second excited state, the uncoupled bases are $|\mathrm{g}\rangle|2\rangle_1|0\rangle_2$, $|\mathrm{g}\rangle|1\rangle_1|1\rangle_2$, $|\mathrm{e}\rangle|1\rangle_1|0\rangle_2$, $|\mathrm{g}\rangle|0\rangle_1|2\rangle_2$ and $|\mathrm{e}\rangle|0\rangle_1|1\rangle_2$, and the Hamiltonian in this subspace is given by

$$H_{\mathrm{nonH}}^{(2)}=\begin{pmatrix} 2\Delta_1-i\kappa_1 & \sqrt{2}J & \sqrt{2}g & 0 & 0 \\ \sqrt{2}J & \Delta_1+\Delta_2-i\frac{\kappa_1+\kappa_2}{2} & 0 & \sqrt{2}J & g \\ \sqrt{2}g & 0 & \Delta_1-i\frac{\kappa_1+\gamma}{2} & 0 & J \\ 0 & \sqrt{2}J & 0 & 2\Delta_2-i\kappa_2 & 0 \\ 0 & g & J & 0 & \Delta_2-i\frac{\kappa_2+\gamma}{2} \end{pmatrix}. \tag{39}$$

The corresponding effective Hamiltonian is given by

$$H_{\mathrm{eff,nonH}}^{(2)}=\begin{pmatrix} 2\Delta_{2,\mathrm{eff}}-i\kappa_{\mathrm{eff}} & \sqrt{2}g_{\mathrm{eff}} \\ \sqrt{2}g_{\mathrm{eff}} & \Delta_{2,\mathrm{eff}}+\Delta_{\mathrm{e,eff}}-i\frac{\kappa_{\mathrm{eff}}+\gamma_{\mathrm{eff}}}{2} \end{pmatrix}, \tag{40}$$

with the uncoupled bases $|\mathrm{g}\rangle|2\rangle_2$ and $|\mathrm{e}\rangle|1\rangle_2$. The eigenvalues of $H_{\mathrm{eff,nonH}}^{(2)}$ are given by

$$\begin{aligned} E_{\pm}^{(2)} &= 2\Delta_{\mathrm{e,eff}}+\frac{1}{2}(3\Delta_{\mathrm{eff}}-i\frac{3\kappa_{\mathrm{eff}}+\gamma_{\mathrm{eff}}}{2})\pm\frac{1}{2}\sqrt{(\Delta_{\mathrm{eff}}-i\frac{\kappa_{\mathrm{eff}}-\gamma_{\mathrm{eff}}}{2})^2+8g_{\mathrm{eff}}^2} \\ &= -2\alpha^2\Delta_1+\frac{1}{2}[3\Delta_2+3\left(\alpha^2-\beta^2\right)\Delta_1-i\frac{3\kappa_2+\gamma+(3\beta^2+\alpha^2)\kappa_1}{2}] \\ &\pm\frac{1}{2}\sqrt{[\Delta_2+(\alpha^2-\beta^2)\,\Delta_1-i\frac{\kappa_2-\gamma+(\beta^2-\alpha^2)\kappa_1}{2}]^2+8\beta^2g^2}. \end{aligned} \tag{41}$$

VI. WEAK EXCITATION SPECTRUM

Under weak excitation, the emitter is predominantly in the ground state. Thus σ_{z} 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

$$-i\omega\tilde{a}_1(\omega)=(-i\Delta_1-\frac{\kappa_1}{2})\tilde{a}_1(\omega)-ig\tilde{\sigma}_-(\omega)-iJ\tilde{a}_2(\omega)-\sqrt{\kappa_1}\tilde{a}_{\mathrm{in},1}(\omega), \tag{42a}$$

$$-i\omega\tilde{a}_2(\omega)=(-i\Delta_2-\frac{\kappa_2}{2})\tilde{a}_2(\omega)-iJ\tilde{a}_1(\omega)-\sqrt{\kappa_2}\tilde{a}_{\mathrm{in},2}(\omega), \tag{42b}$$

$$-i\omega\tilde{\sigma}_-(\omega)=-\frac{\gamma}{2}\tilde{\sigma}_-(\omega)-ig\tilde{a}_1(\omega)-\sqrt{\gamma}\tilde{\sigma}_{\mathrm{in},-}(\omega). \tag{42c}$$

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

$$-i\omega\tilde{a}_2(\omega)=(-i\Delta_{\mathrm{eff}}-\frac{\kappa_{\mathrm{eff}}}{2})\tilde{a}_2(\omega)-ig_{\mathrm{eff}}\tilde{\sigma}_-(\omega)-\sqrt{\kappa_{\mathrm{eff}}}\tilde{a}'_{\mathrm{in},2}(\omega), \tag{43a}$$

$$-i\omega\tilde{\sigma}_-(\omega)=-\frac{\gamma_{\mathrm{eff}}}{2}\tilde{\sigma}_-(\omega)-ig_{\mathrm{eff}}\tilde{a}_2(\omega)-\sqrt{\gamma_{\mathrm{eff}}}\tilde{\sigma}'_{\mathrm{in},-}(\omega), \tag{43b}$$

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

$$H_{\mathrm{cav}}=\omega_1a_1^\dagger a_1+\omega_2a_2^\dagger a_2+J(a_1^\dagger a_2+a_2^\dagger a_1). \tag{44}$$

The quantum Langevin equations are given by

$$\dot{a}_1=(-i\omega_1-\frac{\kappa_1}{2})a_1-iJa_2-\sqrt{\kappa_1}a_{\mathrm{in},1}, \tag{45a}$$

$$\dot{a}_2=(-i\omega_2-\frac{\kappa_2}{2})a_2-iJa_1-\sqrt{\kappa_2}a_{\mathrm{in},2}. \tag{45b}$$

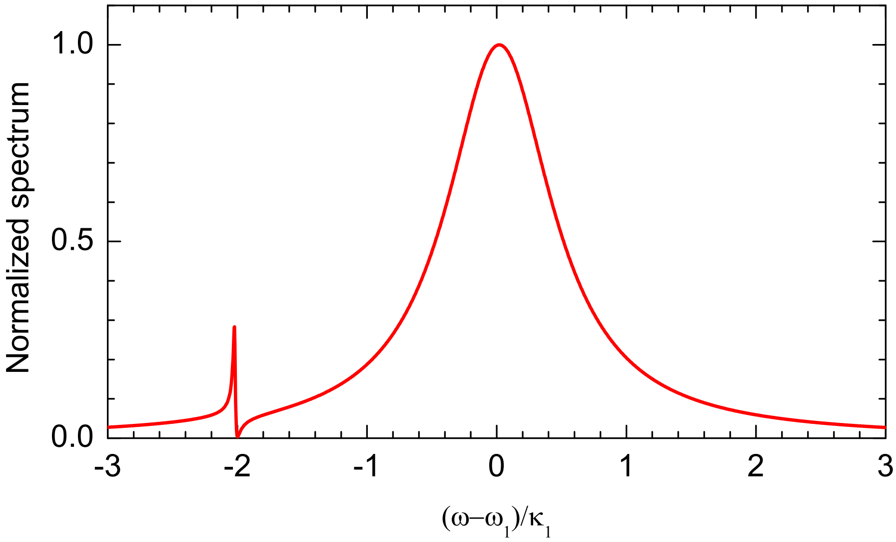

FIG. S1: Normalized spectrum of the coupled cavity system for $\kappa_2/\kappa_1 = 10^{-2}$, $J/\kappa_1 = 0.2$ and $\Delta_{12}/\kappa_1 = 2$.

In the frequency domain, the equations are given by

$$-i\omega\tilde{a}_1(\omega) = (-i\omega_1 - \frac{\kappa_1}{2})\tilde{a}_1(\omega) - iJ\tilde{a}_2(\omega) - \sqrt{\kappa_1}\tilde{a}_{\mathrm{in},1}(\omega), \tag{46a}$$

$$-i\omega\tilde{a}_2(\omega) = (-i\omega_2 - \frac{\kappa_2}{2})\tilde{a}_2(\omega) - iJ\tilde{a}_1(\omega) - \sqrt{\kappa_2}\tilde{a}_{\mathrm{in},2}(\omega). \tag{46b}$$

Then we obtain

$$\tilde{a}_1(\omega) = \frac{(i\omega - i\omega_2 - \frac{\kappa_2}{2})\sqrt{\kappa_1}\tilde{a}_{\mathrm{in},1}(\omega) + iJ\sqrt{\kappa_2}\tilde{a}_{\mathrm{in},2}(\omega)}{(i\omega - i\omega_1 - \frac{\kappa_1}{2})(i\omega - i\omega_2 - \frac{\kappa_2}{2}) + J^2}. \tag{47}$$

The spectrum of the coupled-cavity supermode is given by $S_{\mathrm{cav}}(\omega) = \int \tilde{a}_1^\dagger(\omega)\tilde{a}_1(\omega')d\omega'$. 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 show that the spectrum has Fano-type lineshape. Near the Fano lineshape region the mode density is the combined effect of the two cavity modes.