

Supplemental Material to Pure Dephasing of Light-Matter Systems in the Ultrastrong and Deep-Strong Coupling Regimes

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COMPARISON OF THE BASIC FORMULAS IN THE COULOMB AND DIPOLE GAUGES FOR THE QUANTUM RABI AND HOPFIELD MODELS.

Hamiltonians & operators	Coulomb gauge	Dipole gauge
Rabi Hamiltonian	$\hat{\mathcal{H}}_C$ in Eq. (M-4)	$\hat{\mathcal{H}}_D = \hat{\mathcal{T}}\hat{\mathcal{H}}_C\hat{\mathcal{T}}^\dagger$ above Eq. (M-4) with $\hat{\mathcal{T}} = \exp[-i\hat{\mathcal{A}}\hat{\sigma}_x]$
Rabi annihilation operators	$\hat{a}_C = \hat{a}$	$\hat{a}_D = \hat{\mathcal{T}}\hat{a}\hat{\mathcal{T}}^\dagger = \hat{a} + i\eta\hat{\sigma}_x$ below Eq. (M-6)
Rabi perturbation Hamiltonian for pure dephasing	$\hat{\mathcal{V}}_\phi^C$ in Eq. (M-5)	$\hat{\mathcal{V}}_\phi^D$ below Eq. (M-4)
Hopfield Hamiltonian	\hat{H}_C in Eq. (M-10)	$\hat{H}_D = \hat{\mathcal{T}}\hat{H}_C\hat{\mathcal{T}}^\dagger$ in Eq. (M-9) with $\hat{\mathcal{T}} = \exp[-i\lambda(\hat{a} + \hat{a}^\dagger)(\hat{b} + \hat{b}^\dagger)]$
Hopfield annihilation operators	$\hat{a}_C = \hat{a}$ $\hat{b}_C = \hat{\mathcal{T}}^\dagger\hat{a}\hat{\mathcal{T}} = \hat{b} - i\lambda(\hat{a} + \hat{a}^\dagger)$ below Eq. (M-14)	$\hat{a}_D = \hat{\mathcal{T}}\hat{a}\hat{\mathcal{T}}^\dagger = \hat{a} + i\lambda(\hat{b} + \hat{b}^\dagger)$ below Eq. (M-14) $\hat{b}_D = \hat{b}$
Hopfield perturbation Hamiltonian for pure dephasing	\hat{V}_{dep}^C in Eq. (M-14)	\hat{V}_{dep}^D in Eq. (M-13)

TABLE I. Comparison of the basic formulas in the Coulomb and dipole gauges for the quantum Rabi and Hopfield models. Note the label M- is introduced to refer to equations number in the main text.

PURE DEPHASING IN THE QUANTUM RABI MODEL

Here we analyze how to describe the correct and gauge invariant pure dephasing effects in the quantum Rabi model (QRM), following the procedure described in Ref. [S1] and considering both cavity and qubit decoherence. We start by considering the quantum Rabi Hamiltonian with an additional zero-mean stochastic modulation of the qubit resonance frequency $\hat{\mathcal{V}}_{\text{dep}}^q = f_q(t)\hat{\sigma}_z$. Expressing the Hamiltonian in the dressed basis and moving to the interaction picture with respect to $\hat{\mathcal{V}}_{\text{dep}}^q$, we obtain

$$\hat{\mathcal{V}}_{\text{dep}}^q(t) = f(t) \sum_{j,k} \langle j|\hat{\sigma}_z|k\rangle |j\rangle\langle k| e^{i\omega_{jk}t}, \quad (\text{S1})$$

where $|j\rangle$ are the eigenstates of the total Hamiltonian and ω_{jk} are the transition frequencies. Expressing $f(t)$ in terms of its Fourier decomposition, and assuming that the main contribution to dephasing results from a small frequency interval around ω_{jk} [S1], we obtain

$$\hat{\mathcal{V}}_{\text{dep}}^q(t) = \sum_{j,k} \sigma_z^{jk} |j\rangle\langle k| f_{-\omega_{jk}}(t) , \quad (\text{S2})$$

where

$$f_{\omega_{jk}}(t) = \sqrt{S_f(\omega_{jk})} \xi_{\omega_{jk}}(t) , \quad (\text{S3})$$

$S_f(\omega)$ is the spectral density of $f(t)$, and $\xi(\omega)$ such that $\langle \xi(\omega) \rangle = 0$ and $\langle \xi(\omega) \xi(\omega') \rangle = \delta(\omega - \omega')$ (i.e., corresponding to white noise). If the transition frequencies ω_{jk} are well-separated, we can treat each term of the above summation as an independent noise [S1].

We are now able to write down the dressed Lindbladian in case of qubit pure dephasing:

$$\mathcal{L}_{\text{dr}} \cdot = \mathcal{D} \left[\sum_j \Phi^j |j\rangle\langle j| \right] \cdot + \sum_{j,k \neq j} \Gamma_\phi^{jk} \mathcal{D} [|j\rangle\langle k|] \cdot , \quad (\text{S4})$$

where

$$\Phi^j = \sqrt{\frac{\gamma_\phi(0)}{2}} \sigma_z^{jj} , \quad (\text{S5})$$

and

$$\Gamma_\phi^{jk} = \frac{\gamma_\phi(\omega_{kj})}{2} |\sigma_z^{jk}|^2 . \quad (\text{S6})$$

The whole procedure described above can also be applied to the case of cavity pure dephasing, by considering the QRM Hamiltonian with an additional zero-mean stochastic modulation of the cavity resonance frequency $\hat{\mathcal{V}}_{\text{dep}}^c = f_c(t) \hat{a}^\dagger \hat{a}$. In this case, this stochastic perturbation, expressed in the dressed basis and in the interaction picture, becomes

$$\hat{\mathcal{V}}_{\text{dep}}^c(t) = \sum_{j,k} \langle j | \hat{a}^\dagger \hat{a} | k \rangle |j\rangle\langle k| f_{-\omega_{jk}}(t) , \quad (\text{S7})$$

while the Lindbladian remains in the same form of Eq. (S4), with the only difference of Φ^j and Γ_ϕ^{jk} , which become respectively,

$$\Phi^j = \sqrt{\frac{\gamma_\phi(0)}{2}} \langle j | \hat{a}^\dagger \hat{a} | j \rangle , \quad (\text{S8})$$

$$\Gamma_\phi^{jk} = \frac{\gamma_\phi(\omega_{kj})}{2} |\langle j | \hat{a}^\dagger \hat{a} | k \rangle|^2 . \quad (\text{S9})$$

However, we have seen in the main text that the approach described above does not reproduce the correct results. In particular, we have shown that, if one uses the Coulomb or dipole gauge, significantly different results can be obtained. For example, when using the Coulomb gauge, the bare $\hat{\sigma}_z$ operator becomes $\hat{\sigma}_z^C = \hat{\mathcal{T}}^\dagger \hat{\sigma}_z \hat{\mathcal{T}}$, since the minimal coupling is applied to the matter system, while the photonic operator $\hat{a}^\dagger \hat{a}$ becomes $\hat{a}_D^\dagger \hat{a}_D = \hat{\mathcal{T}} \hat{a}^\dagger \hat{a} \hat{\mathcal{T}}^\dagger$ in the dipole gauge. Thus, to correctly describe pure dephasing effects, we need to substitute in the Lindbladian given in Eq. (S4): $\hat{\sigma}_z \rightarrow \hat{\sigma}_z^C$ in the Coulomb gauge, and $\hat{a}^\dagger \hat{a} \rightarrow \hat{a}_D^\dagger \hat{a}_D$ in the dipole gauge.

Analytical derivation of the pure dephasing rates

By adopting the procedure described above, we are able to derive analytically the pure dephasing rates of both cavity and qubit. Starting from the Coulomb gauge and using Eq. (S4), we discard the off-diagonal terms Γ_ϕ^{jk} since

this contribution is significant only if the dephasing bath has a spectral weight at the potentially high frequency ω_{jk} , leading to the following equation:

$$\dot{\hat{\rho}} = -i[\hat{\mathcal{H}}_C, \hat{\rho}] + \frac{\gamma_\phi(0)}{2} \mathcal{D} \left[\sum_j \sigma_z^{C,jj} |j\rangle\langle j| \right] \hat{\rho}, \quad (\text{S10})$$

where $\sigma_z^{C,jj} = \langle j|\hat{\sigma}_z|j\rangle$. We now expand the Lindblad dissipator

$$\mathcal{D} \left[\sum_j \sigma_z^{C,jj} |j\rangle\langle j| \right] \hat{\rho} = \frac{1}{2} \left[2 \sum_j \sum_{j'} \sigma_z^{C,jj} \sigma_z^{C,j'j'} |j\rangle\langle j| \hat{\rho} |j'\rangle\langle j'| - \sum_j \sum_{j'} \sigma_z^{C,jj} \sigma_z^{C,j'j'} |j'\rangle\langle j'| |j\rangle\langle j| \hat{\rho} \right. \quad (\text{S11})$$

$$\left. - \sum_j \sum_{j'} \sigma_z^{C,jj} \sigma_z^{C,j'j'} \hat{\rho} |j'\rangle\langle j'| |j\rangle\langle j| \right], \quad (\text{S12})$$

and we focus on the matrix element of the density matrix relative to the transition $(\tilde{1}_-, \tilde{0})$, but the same procedure can be applied to all the other transitions. The corresponding equation (in the interaction picture) for that matrix element becomes

$$\begin{aligned} \frac{d}{dt} \hat{\rho}_{\tilde{1}_-, \tilde{0}}^{(I)} &= \frac{\gamma_\phi(0)}{4} \langle \tilde{1}_- | \left[2 \sum_j \sum_{j'} \sigma_z^{C,jj} \sigma_z^{C,j'j'} |j\rangle\langle j| \hat{\rho}^{(I)} |j'\rangle\langle j'| - \sum_j |\sigma_z^{C,jj}|^2 |j\rangle\langle j| \hat{\rho}^{(I)} - \sum_j |\sigma_z^{C,jj}|^2 \hat{\rho}^{(I)} |j\rangle\langle j| \right] | \tilde{0} \rangle \\ &= \frac{\gamma_\phi(0)}{4} \left[2 \sum_j \sum_{j'} \sigma_z^{C,jj} \sigma_z^{C,j'j'} \langle \tilde{1}_- | j \rangle \langle j | \hat{\rho}^{(I)} | j' \rangle \langle j' | \tilde{0} \rangle - \sum_j |\sigma_z^{C,jj}|^2 \langle \tilde{1}_- | j \rangle \langle j | \hat{\rho}^{(I)} | \tilde{0} \rangle \right. \\ &\quad \left. - \sum_j |\sigma_z^{C,jj}|^2 \langle \tilde{1}_- | \hat{\rho}^{(I)} | j \rangle \langle j | \tilde{0} \rangle \right] \\ &= \frac{\gamma_\phi(0)}{4} \left[2 \sigma_z^{C, \tilde{1}_- \tilde{1}_-} \sigma_z^{C, \tilde{0} \tilde{0}} \langle \tilde{1}_- | \hat{\rho}^{(I)} | \tilde{0} \rangle - |\sigma_z^{C, \tilde{1}_- \tilde{1}_-}|^2 \langle \tilde{1}_- | \hat{\rho}^{(I)} | \tilde{0} \rangle - |\sigma_z^{C, \tilde{0} \tilde{0}}|^2 \langle \tilde{1}_- | \hat{\rho}^{(I)} | \tilde{0} \rangle \right] \\ &= -\frac{\gamma_\phi(0)}{4} \left| \sigma_z^{C, \tilde{1}_- \tilde{1}_-} - \sigma_z^{C, \tilde{0} \tilde{0}} \right|^2 \hat{\rho}_{\tilde{1}_-, \tilde{0}}^{(I)}. \end{aligned} \quad (\text{S13})$$

By choosing the dipole gauge, one should replace $\sigma_z^{C,jj} \rightarrow \sigma_z^{jj}$. The same procedure is valid also for cavity pre dephasing, where we need to use $\hat{a}^\dagger \hat{a}$ in the Coulomb gauge and $\hat{a}_D^\dagger \hat{a}_D$ in the dipole gauge.

PURE DEPHASING IN BOSONIC SYSTEMS

We now consider pure dephasing effects in bosonic systems. First, we consider a simple non-interacting harmonic oscillator, then we analyze the Hopfield model.

Non-interacting harmonic oscillator

Here we consider a single-mode bosonic field described by the harmonic oscillator Hamiltonian $\hat{H}_0 = \omega_0 \hat{a}^\dagger \hat{a}$ affected by pure dephasing. Analogously to what we described in previous sections, in order to consider the dephasing effects, we introduce an additional zero-mean stochastic modulation of the resonance frequency $\hat{V}_{\text{dep}}^h = f_h(t) \hat{a}^\dagger \hat{a}$. Moving to the interaction picture, we notice that this component does not rotate, since it has a zero-frequency oscillation. Thus, transforming $f_h(t)$ in its Fourier components, and assuming that the main contribution to dephasing comes from a small frequency interval around $\omega = 0$ [S1], we obtain

$$\hat{V}_{\text{dep}}^h(t) = f_0(t) \hat{a}^\dagger \hat{a}, \quad (\text{S14})$$

where $f_0(t) = \sqrt{S_f(0)} \xi_0(t)$. This equation is quite similar to Eq. (S2) with the only difference that here we do not have the expansion in the dressed basis (since we are not considering a hybrid quantum system), and that we have only

the zero-frequency contribution (since $\hat{\mathcal{V}}_{\text{dep}}^h$ rotates at zero frequency in the interaction picture). These considerations allow us to write the Lindbladian describing this pure dephasing effect as

$$\mathcal{L}\cdot = \sqrt{\frac{\gamma_\phi(0)}{2}} \mathcal{D} [\hat{a}^\dagger \hat{a}] \cdot, \quad (\text{S15})$$

with $\gamma_\phi(0) = 2S_f(0)$.

Hopfield model

Here we analyze pure dephasing effects in the Hopfield model, following the procedure described in the previous sections and extending the results of Ref. [S1]. Moreover, we consider both light and matter decoherence. First, it is useful to diagonalize the Hopfield Hamiltonian using the polaritonic operators [S2], where the lower and upper polariton operators ($\mu = 1, 2$) can be defined as

$$\hat{P}^\mu = U_b^\mu \hat{b} + U_a^\mu \hat{a} + V_b^\mu \hat{b}^\dagger + V_a^\mu \hat{a}^\dagger. \quad (\text{S16})$$

Using the property

$$|U_b^\mu|^2 + |U_a^\mu|^2 - |V_b^\mu|^2 - |V_a^\mu|^2 = 1, \quad (\text{S17})$$

which guarantee the correct polariton commutation rules [S2], we can invert Eq. (S16) in order to obtain

$$\hat{a} = \sum_{\mu=1}^2 \left(U_a^\mu \hat{P}_\mu - V_a^\mu \hat{P}_\mu^\dagger \right), \quad (\text{S18a})$$

$$\hat{b} = \sum_{\mu=1}^2 \left(U_b^\mu \hat{P}_\mu - V_b^\mu \hat{P}_\mu^\dagger \right). \quad (\text{S18b})$$

To describe the matter pure dephasing, we consider an additional zero-mean stochastic modulation of the matter resonance frequency $\hat{V}_{\text{dep}}^x = f_x(t) \hat{b}^\dagger \hat{b}$. In terms of the polaritonic operators we have

$$\hat{b}^\dagger \hat{b} = A_1 \hat{P}_1^\dagger \hat{P}_1 + A_2 \hat{P}_2^\dagger \hat{P}_2 + B_{12} \hat{P}_1^\dagger \hat{P}_2 + B_{21} \hat{P}_2^\dagger \hat{P}_1, \quad (\text{S19})$$

with

$$A_\mu = |U_b^\mu|^2 + |V_b^\mu|^2 \quad (\text{S20})$$

$$B_{12} = B_{21}^* = U_b^1{}^* U_b^2 + V_b^1 V_b^2{}^*, \quad (\text{S21})$$

where we have included only the terms which do not oscillate in time, or oscillate at low frequency, corresponding to applying the rotating wave approximation (RWA), and we have eliminated the constants derived from commutation rules, which have no dynamical consequences. Moving to the interaction picture, this contribution becomes

$$\hat{V}_{\text{dep}}^x(t) = f_x(t) \left[A_1 \hat{P}_1^\dagger \hat{P}_1 + A_2 \hat{P}_2^\dagger \hat{P}_2 + e^{-i\omega_{21}t} B_{12} \hat{P}_1^\dagger \hat{P}_2 + e^{i\omega_{21}t} B_{21} \hat{P}_2^\dagger \hat{P}_1 \right], \quad (\text{S22})$$

where $\omega_{21} = \omega_2 - \omega_1$ with the polaritonic eigenfrequencies ω_i . Equation (S22) can be written in a more compact form as

$$\hat{V}_{\text{dep}}^x = f_x(t) \left[\hat{D}_{12} + e^{-i\omega_{21}t} \hat{M}_{12} + e^{i\omega_{21}t} \hat{M}_{12}^\dagger \right],$$

with

$$\hat{D}_{12} = A_1 \hat{P}_1^\dagger \hat{P}_1 + A_2 \hat{P}_2^\dagger \hat{P}_2, \quad (\text{S23})$$

$$\hat{M}_{12} = B_{12} \hat{P}_1^\dagger \hat{P}_2, \quad (\text{S24})$$

and using the results presented in the previous sections, we obtain

$$\hat{V}_{\text{dep}}^x(t) = f_0(t) \hat{D}_{12} + f_{\omega_{21}}(t) \hat{M}_{12} + f_{-\omega_{21}}(t) \hat{M}_{12}^\dagger, \quad (\text{S25})$$

with $f_\omega(t)$ expressed in Eq. (S3). Thus, the resulting Lindbladian in the case of matter pure dephasing is

$$\mathcal{L} \cdot = \frac{1}{2} \gamma_\phi(\omega_{21}) \mathcal{D}[\hat{M}_{12}] \cdot + \frac{1}{2} \gamma_\phi(-\omega_{21}) \mathcal{D}[\hat{M}_{12}^\dagger] \cdot + \frac{1}{2} \gamma_\phi(0) \mathcal{D}[\hat{D}_{12}] \cdot, \quad (\text{S26})$$

with $\gamma_\phi(\omega) = 2S_f(\omega)$.

The same procedure, as described above, can also be applied to the case of cavity pure dephasing, by considering an additional zero-mean stochastic modulation of the cavity resonance frequency $\hat{V}_{\text{dep}}^c = f_c(t) \hat{a}^\dagger \hat{a}$. The procedure remains the same for the matter dephasing case, except that now we consider

$$\hat{a}^\dagger \hat{a} = A_1 \hat{P}_1^\dagger \hat{P}_1 + A_2 \hat{P}_2^\dagger \hat{P}_2 + B_{12} \hat{P}_1^\dagger \hat{P}_2 + B_{21} \hat{P}_2^\dagger \hat{P}_1, \quad (\text{S27})$$

where

$$A_\mu = |U_a^\mu|^2 + |V_a^\mu|^2, \quad (\text{S28})$$

$$B_{12} = B_{21}^* = U_a^{1*} U_a^2 + V_a^1 V_a^{2*}. \quad (\text{S29})$$

This yields a Lindbladian of the same form of Eq. (S26) with the only difference for the polariton coefficients expressed in Eqs. (S28) and (S29).

However, we have seen in the main text that this approach can lead to wrong results, depending on the chosen gauge. Indeed, when using the Coulomb gauge, the matter operator \hat{b} becomes $\hat{b}_C = \hat{T}^\dagger \hat{b} \hat{T}$, since the minimal coupling is applied to the matter system. On the contrary, when using the dipole gauge, the minimal coupling is applied to the photonic system, and the *dressed* photonic operator becomes $\hat{a}_D = \hat{T} \hat{a} \hat{T}^\dagger$. This consideration leads us to note that the polariton diagonalization leads to different Hopfield coefficients if we choose the Coulomb or dipole gauge. In particular, in the dipole gauge, we have

$$\hat{b} = \sum_{\mu=1}^2 \left(U_b^{\mu'} \hat{P}'_\mu - V_b^{\mu'} \hat{P}'_\mu^\dagger \right), \quad (\text{S30})$$

where P'_μ are the polariton operators obtained by diagonalizing the Hopfield Hamiltonian in the dipole gauge. While in the Coulomb gauge we have

$$\begin{aligned} \hat{b}_C &= \hat{T}^\dagger \left[\sum_{\mu=1}^2 \left(U_b^{\mu'} \hat{P}'_\mu - V_b^{\mu'} \hat{P}'_\mu^\dagger \right) \right] \hat{T} \\ &= \sum_{\mu=1}^2 \left(U_b^{\mu'} \hat{T}^\dagger \hat{P}'_\mu \hat{T} - V_b^{\mu'} \hat{T}^\dagger \hat{P}'_\mu^\dagger \hat{T} \right) \\ &= \sum_{\mu=1}^2 \left(U_b^{\mu''} \hat{P}_\mu - V_b^{\mu''} \hat{P}_\mu^\dagger \right), \end{aligned} \quad (\text{S31})$$

which contains the polariton operators obtained by diagonalizing the Hamiltonian in the Coulomb gauge, but with the same coefficients of the dipole gauge. To obtain Eq. (S31), we have used the relation

$$\hat{P}_\mu = \hat{T}^\dagger \hat{P}'_\mu \hat{T}, \quad (\text{S32})$$

which, although intuitively obvious, can be rigorously demonstrated using the definition of polaritonic operators; in particular, those operators that, each in its specific gauge, enable the diagonalization of the gauge-correspondent Hamiltonian. For example, we have:

$$[\hat{P}_\mu, \hat{\mathcal{H}}_C] = \Omega_\mu \hat{P}_\mu, \quad (\text{S33a})$$

$$[\hat{P}'_\mu, \hat{\mathcal{H}}_D] = \Omega_\mu \hat{P}'_\mu. \quad (\text{S33b})$$

In order to demonstrate Eq. (S32), we can calculate how Eq. (S33a) transforms from the Coulomb to dipole gauge. Gauge invariance implies that the final result has to be equal to Eq. (S33b). We obtain:

$$\hat{T} [\hat{P}_\mu, \hat{\mathcal{H}}_C] \hat{T}^\dagger = \Omega_\mu \hat{T} \hat{P}_\mu \hat{T}^\dagger, \quad (\text{S34a})$$

$$\begin{aligned} \hat{T} [\hat{P}_\mu, \hat{\mathcal{H}}_C] \hat{T}^\dagger &= \hat{T} (\hat{P}_\mu \hat{\mathcal{H}}_C - \hat{\mathcal{H}}_C \hat{P}_\mu) \hat{T}^\dagger \\ &= \hat{T} \hat{P}_\mu \hat{\mathcal{H}}_C \hat{T}^\dagger - \hat{T} \hat{\mathcal{H}}_C \hat{P}_\mu \hat{T}^\dagger \\ &= \hat{T} \hat{P}_\mu \hat{T}^\dagger \hat{T} \hat{\mathcal{H}}_C \hat{T}^\dagger - \hat{T} \hat{\mathcal{H}}_C \hat{T}^\dagger \hat{T} \hat{P}_\mu \hat{T}^\dagger \\ &= \hat{T} \hat{P}_\mu \hat{T}^\dagger \hat{\mathcal{H}}_D - \hat{\mathcal{H}}_D \hat{T} \hat{P}_\mu \hat{T}^\dagger = [\hat{T} \hat{P}_\mu \hat{T}^\dagger, \hat{\mathcal{H}}_D]. \end{aligned} \quad (\text{S34b})$$

Combining the results of Eqs. (S34a) and (S34b), we obtain:

$$[\hat{T}\hat{P}_\mu\hat{T}^\dagger, \hat{H}_D] = \Omega_\mu\hat{T}\hat{P}_\mu\hat{T}^\dagger, \quad (\text{S35})$$

which is the definition of the polariton operators \hat{P}'_μ in the dipole-gauge (which are the operators that allow the diagonalization of $\hat{\mathcal{H}}_D$) given by Eq. (S33b). Hence, Eq. (S32) is the correct gauge transformation for the polaritonic operators.

The whole analysis described above can be summarized as follows: in the case of matter pure dephasing, the stochastic perturbation is: $\hat{V}_{\text{dep}}^x = f_x(t)\hat{b}^\dagger\hat{b}$ in the dipole gauge, and $\hat{V}_{\text{dep}}^x = f_x(t)\hat{b}_C^\dagger\hat{b}_C$ in the Coulomb gauge, where

$$\hat{b}^\dagger\hat{b} = A'_1\hat{P}'_1{}^\dagger\hat{P}'_1 + A'_2\hat{P}'_2{}^\dagger\hat{P}'_2 + B'_{12}\hat{P}'_1{}^\dagger\hat{P}'_2 + B'_{21}\hat{P}'_2{}^\dagger\hat{P}'_1 \quad (\text{S36})$$

and

$$\hat{b}_C^\dagger\hat{b}_C = A'_1\hat{P}'_1{}^\dagger\hat{P}'_1 + A'_2\hat{P}'_2{}^\dagger\hat{P}'_2 + B'_{12}\hat{P}'_1{}^\dagger\hat{P}'_2 + B'_{21}\hat{P}'_2{}^\dagger\hat{P}'_1, \quad (\text{S37})$$

with

$$A'_\mu = |U_b^{\mu\mu}|^2 + |V_b^{\mu\mu}|^2, \quad (\text{S38})$$

$$B'_{12} = B'_{21}{}^* = U_b^{1'2'}U_b^{2'1'} + V_b^{1'}V_b^{2' *} . \quad (\text{S39})$$

As a result, to correctly describe the matter pure dephasing, we need to use the dipole coefficients, given in Eqs. (S38) and (S39), in the Lindbladian expressed in Eq. (S26), even when using the Coulomb gauge. On the contrary, for the photonic pure dephasing, the stochastic perturbation is: $\hat{V}_{\text{dep}}^c = f_c(t)\hat{a}^\dagger\hat{a}$ in the Coulomb gauge, and $\hat{V}_{\text{dep}}^c = f_c(t)\hat{a}_D^\dagger\hat{a}_D$ in the dipole gauge. Thus, we need to use the Coulomb polariton coefficients in the Lindbladian even when using the dipole gauge.

ADDITIONAL FIGURES

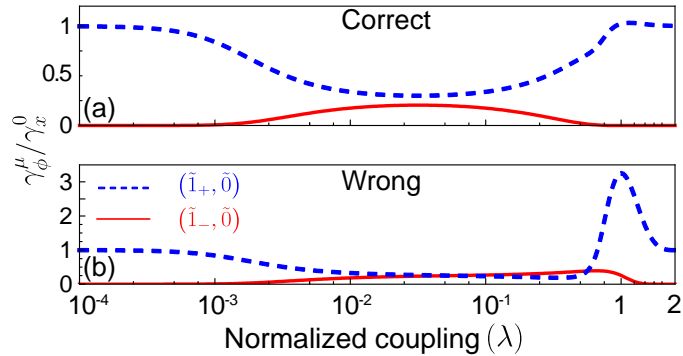


FIG. S1. Quantum Rabi model: Normalized pure dephasing rate for the two lowest energy transitions, for a small qubit-cavity detuning $\delta = 3 \times 10^{-3}$ assuming only the cavity pure dephasing. (a) Correct gauge-invariant results versus (b) wrong Coulomb gauge results.

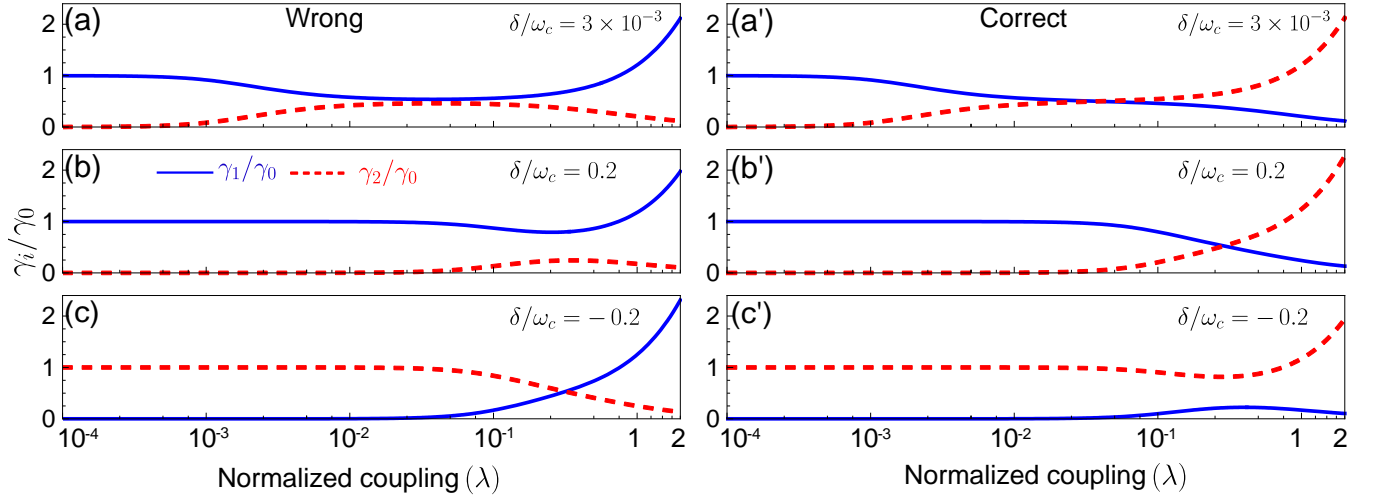


FIG. S2. Hopfield model: Pure dephasing rate of the lower and upper polaritons, originating from exciton dephasing, versus the normalized coupling strength, obtained for different exciton-cavity detunings, and considering only cavity pure dephasing.

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