

Supplementary Material for “Model Order Reduction for Open Quantum Systems Based on Measurement-adapted Time-coarse Graining”

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(Dated: March 27, 2026)

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Appendix A: Input-output theory and the choice of interaction picture in TCG

Analogous to the canonical formulation of quantum input-output theory in [1], we can investigate the extraction of quantum information with finite time resolution. For instance, suppose that we probe a circuit QED system by sending in a microwave pulse to a quantum resonator through a waveguide and reading out the reflected signal with some finite time resolution τ . Then a typical Hamiltonian description of such a system can be written as

$$\hat{H} = \hat{H}_s + \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega \omega B_\omega^\dagger B_\omega - (a - a^\dagger) \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega (B_\omega - B_\omega^\dagger) \quad (\text{A1})$$

where $\hat{H}_s$ is the Schrodinger-picture Hamiltonian of the quantum system of interest, B_ω is the annihilation operator of a waveguide mode through which the system dissipates and gets probed at the same time, while $-i(a - a^\dagger)$ is the quadrature variable of

the resonator mode to which the waveguide modes couple. In the Heisenberg picture, we have the following operator equation:

$$\begin{aligned} & \partial_t B_\omega^H(t) \\ &= i \left[\int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega \omega B_\omega^{H\dagger}(t) B_\omega^H(t) - (a^H(t) - a^{H\dagger}(t)) \int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega g_\omega (B_\omega^H(t) - B_\omega^{H\dagger}(t)), B_\omega^H(t) \right] \\ &= -i\omega B_\omega^H(t) - ig_\omega (a^H(t) - a^{H\dagger}(t)), \end{aligned} \quad (\text{A2})$$

$$\begin{aligned} \partial_t a^H(t) &= i [\hat{H}_s, a^H(t)] - i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega (B_\omega^H(t) - B_\omega^{H\dagger}(t)) \\ &= i [\hat{H}_s, a^H(t)] - i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \left(e^{-i\omega(t-t_i)} B_\omega^H(t_i) - e^{i\omega(t-t_i)} B_\omega^{H\dagger}(t_i) \right) + \mathcal{O}(g^2), \end{aligned} \quad (\text{A3})$$

where the first one can be formally solved to obtain

$$\begin{aligned} B_\omega^H(t) &= e^{-i\omega(t-t_i)} B_\omega^H(t_i) - ig_\omega \int_0^{t-t_i} dt' \cdot e^{-i\omega t'} (a^H(t-t') - a^{H\dagger}(t-t')) \\ &= e^{i\omega(t_f-t)} B_\omega^H(t_f) + ig_\omega \int_0^{t_f-t} dt' \cdot e^{i\omega t'} (a^H(t+t') - a^{H\dagger}(t+t')), \end{aligned} \quad (\text{A4})$$

and the second one can be formally integrated to give us

$$a^H(t-t') \approx e^{i\omega_0 t'} a^H(t) + \int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega g_\omega \int_0^{t'} dt'' e^{i\omega_0(t'-t'')} \left(i e^{-i\omega(t-t'')} B_\omega^H + h.c. \right). \quad (\text{A5})$$

Here ω_0 is the system frequency of the Heisenberg-picture operator $a_H(t)$ during the measurement process, while the generation, interaction, and detection times t_i, t, t_f are related as $t_f = t_i + \frac{L}{c} = t + \frac{L}{c}$ with L (or l) the optical distance between the signal generator (or the artificial atom) and the detector. Therefore, if we define the input and output waveguide mode operators as

$$B_{\text{in}}^H(t) := \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega e^{-i\omega(t-t_i)} B_\omega^H(t_i) \quad (\text{A6})$$

and

$$B_{\text{out}}^H(t) := \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega e^{i\omega(t_f-t)} B_\omega^H(t_f) \quad (\text{A7})$$

respectively, then we can relate $B_{\text{in}}^H(t)$ to $B_{\text{out}}^H(t)$ by taking the difference between the following two expressions of $\int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega B_\omega^H(t)$

$$\begin{aligned} \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega B_\omega^H(t) &= \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega e^{-i\omega(t-t_i)} B_\omega^H(t_i) + i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_0^{t-t_i} dt' \\ &\quad \cdot \left(e^{-i(\omega-\omega_0)t'} e^{-i\omega_0 t'} a^H(t-t') - e^{-i(\omega+\omega_0)t'} e^{i\omega_0 t'} a^{H\dagger}(t-t') \right) \\ &= \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega e^{i\omega(t_f-t)} B_\omega^H(t_f) + i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_0^{t_f-t} dt' \\ &\quad \cdot \left(e^{i(\omega-\omega_0)t'} e^{i\omega_0 t'} a^H(t+t') - e^{i(\omega+\omega_0)t'} e^{-i\omega_0 t'} a^{H\dagger}(t+t') \right) \end{aligned} \quad (\text{A8})$$

which gives us the equation

$$B_{\text{out}}^H(t) = B_{\text{in}}^H(t) - i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_{-\frac{L-l}{c}}^{\frac{L}{c}} dt' \cdot \left(e^{i\omega t'} a^H(t+t') - e^{i\omega t'} a^{H\dagger}(t+t') \right) \quad (\text{A9})$$

In the usual formulation of input-output theory, one assumes the validity of the approximation $a^H(t+t') \approx e^{-i\omega_0 t'} a^H(t)$, for some $\omega_0 > 0$, which then allows us to write

$$\begin{aligned} B_{\text{out}}^H(t) &= B_{\text{in}}^H(t) - i \int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega g_\omega \int_{-\frac{L-l}{c}}^{\frac{L}{c}} dt' \left(e^{i\omega t'} a^H(t+t') - e^{i\omega t'} a^{H\dagger}(t+t') \right) \\ &\approx B_{\text{in}}^H(t) - i \int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega g_\omega \int_{-\infty}^\infty dt' \left(e^{i(\omega-\omega_0)t'} a^H(t) - e^{i(\omega+\omega_0)t'} a^{H\dagger}(t) \right) \\ &= B_{\text{in}}^H(t) - i D_{\omega_0} g_{\omega_0} a^H(t). \end{aligned} \quad (\text{A10})$$

Usually, Eq.(A10) is regarded as the final formula for the observed output signal in terms of the input signal $B_{\text{in}}^H(t)$ and the system observable $-iD_{\omega_0}g_{\omega_0}a^H(t)$. In light of the epistemology of measurement-informed coarse graining, however, we would like to delve deeper into the measurement process and assume finite time resolution for the physically observed signal. To this goal, we first evaluate the expectation value of the output signal as

$$\begin{aligned}
\langle B_{\text{out}}^H(t) \rangle &= \text{Tr}(\rho(t_0)B_{\text{out}}^H(t)) \\
&= -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_{-\frac{L-l}{c}}^{\frac{l}{c}} dt' \cdot \left(e^{i\omega t'} \text{Tr}[\rho(t_0)a^H(t+t')] - e^{i\omega t'} \text{Tr}[\rho(t_0)a^{H\dagger}(t+t')] \right) \\
&\quad + \text{Tr}[\rho(t_0)B_{\text{in}}^H(t)] \\
&= -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_{-\frac{L-l}{c}}^{\frac{l}{c}} dt' \cdot e^{i\omega t'} \left(e^{-i\omega'(t+t'-t_0)} \text{Tr}[\rho(t_0) \right. \\
&\quad \cdot U^\dagger(t+t', t_0)U_{\omega'}(t+t'-t_0)aU_{\omega'}^\dagger(t+t'-t_0)U(t+t', t_0)] - h.c.) \\
&\quad + \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega e^{-i\omega(t-t_i)} \text{Tr}[\rho(t_0)B_\omega^H(t_i)] \\
&= -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_{-\frac{L-l}{c}}^{\frac{l}{c}} dt' \cdot e^{i\omega t'} \left(e^{-i\omega'(t+t'-t_0)} \text{Tr}[\rho_{\omega'}(t+t')a] - h.c.) \right. \\
&\quad \left. + \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega e^{-i\omega(t-t_i)} \text{Tr}[\rho(t_0)B_\omega^H(t_i)] \right)
\end{aligned} \tag{A11}$$

for any ω' , where

$$U_{\omega'}(t+t'-t_0) \equiv e^{-i(\omega'a^\dagger a + \tilde{H}_0)(t+t'-t_0)} \tag{A12}$$

is the free time-evolution operator with arbitrary hermitian $\tilde{H}_0$ so that $[\tilde{H}_0, a] = 0$, and

$$\rho_{\omega'}(t+t') \equiv U_{\omega'}^\dagger(t+t'-t_0)U(t+t', t_0)\rho(t_0)U^\dagger(t+t', t_0)U_{\omega'}(t+t'-t_0) \tag{A13}$$

is the corresponding interaction-picture density matrix.

During heterodyne detection, the output signal $B_{\text{out}}^H(t)$ from the system is then combined with a local oscillator mode at frequency $\omega_{\text{LO}} \sim \omega_0$ as it passes through an IQ-mixer, and the mixed signals at the two output ports of the IQ-mixer are what get measured (with finite time resolution) and recorded eventually, as explained in [2, 3]. Therefore, the experimentally observed signals are proportional to the real and imaginary parts of

$$\begin{aligned}
&\langle \overline{e^{i(\omega_{\text{LO}}t-\varphi)}B_{\text{out}}^H(t)} \rangle \\
&= -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_{-\frac{L-l}{c}}^{\frac{l}{c}} dt' \cdot e^{i\omega t' - i\varphi} \left(e^{-i\omega'(t'-t_0)} \text{Tr}[\overline{e^{i(\omega_{\text{LO}}-\omega')t} \rho_{\omega'}(t+t')a}] - h.c. \right) \\
&\quad + \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega e^{-i\omega(t-t_i)} \text{Tr}[\rho(t_0)B_\omega^H(t_i)]
\end{aligned} \tag{A14}$$

for some real-valued phase φ . The first line of the equation above will be a functional of $\bar{\rho}(t+t')$ iff we set $\omega' = \omega_{\text{LO}}$, in which case we have

$$\begin{aligned}
\langle \overline{e^{i(\omega_{\text{LO}}t-\varphi)}B_{\text{out}}^H(t)} \rangle &\approx -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_{-\frac{L-l}{c}}^{\frac{l}{c}} dt' \cdot e^{i\omega t' - i\varphi} e^{-i\omega_{\text{LO}}(t'-t_0)} \text{Tr}[\overline{\rho_{\omega_{\text{LO}}}(t+t')a}] \\
&\quad + \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega e^{-i\omega(t-t_i)} \text{Tr}[\rho(t_0)B_\omega^H(t_i)] \\
&\approx -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \int_{-\frac{L-l}{c}}^{\frac{l}{c}} dt' \cdot e^{i\omega t' - i\varphi} e^{-i\omega_{\text{LO}}(t'-t_0)} \text{Tr}[\overline{\rho_{\omega_{\text{LO}}}(t+t')a}] \\
&\quad + e^{-i\varphi} \frac{\mathcal{D}_{\omega_{\text{LO}}}}{\sqrt{2\pi\tau}} e^{-\frac{(t-t_i)^2}{2\tau^2}} \text{Tr}[\rho(t_0)B_{\omega_{\text{LO}}}^H(t_i)]
\end{aligned} \tag{A15}$$

where we have assumed that $\mathcal{D}_\omega$ and $\text{Tr}[\rho(t_0)B_\omega^H(t_i)]$ are roughly flat for

$$\omega \in (\omega_{\text{LO}} - \tau^{-1}, \omega_{\text{LO}} + \tau^{-1}). \quad (\text{A16})$$

If we further make the Markov approximation

$$\text{Tr}[\overline{\rho_{\omega_{\text{LO}}}}(t+t')a] \approx e^{i(\omega_{\text{LO}}-\omega_0)t'} \text{Tr}[\overline{\rho_{\omega_{\text{LO}}}}(t)a] \quad (\text{A17})$$

then we can also make the usual secular approximation and obtain that

$$\begin{aligned} \langle \overline{e^{i(\omega_{\text{LO}}t-\varphi)} B_{\text{out}}^H(t)} \rangle &\approx -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega e^{-i\varphi+i\omega_{\text{LO}}t_0} \int_{-\frac{L-l}{c}}^{\frac{l}{c}} dt' \cdot e^{i(\omega-\omega_0)t'} \text{Tr}[\overline{\rho_{\omega_{\text{LO}}}}(t)a] \\ &\quad + e^{-i\varphi} \frac{\mathcal{D}_{\omega_{\text{LO}}}}{\sqrt{2\pi\tau}} e^{-\frac{(t-t_i)^2}{2\tau^2}} \text{Tr}[\rho(t_0)B_{\omega_{\text{LO}}}^H(t_i)] \\ &\approx -i\mathcal{D}_{\omega_0} g_{\omega_0} e^{-i\varphi+i\omega_{\text{LO}}t_0} \text{Tr}[\overline{\rho_{\omega_{\text{LO}}}}(t)a] \\ &\quad + e^{-i\varphi} \frac{\mathcal{D}_{\omega_{\text{LO}}}}{\sqrt{2\pi\tau}} e^{-\frac{(t-t_i)^2}{2\tau^2}} \text{Tr}[\rho(t_0)B_{\omega_{\text{LO}}}^H(t_i)] \end{aligned} \quad (\text{A18})$$

where the second term in the last equation is a recurring background signal independent of the system dynamics and can be ignored in the limit where $t - t_i \gg \tau$. Consequently, according to the analysis in [2, 4, 5], ideal homodyne measurements will correspond to the measurement of a particular resonator quadrature variable with respect to the TCG density matrix $\overline{\rho_{\omega_{\text{LO}}}}(t)$, whereas ideal heterodyne measurements will be sampling from the Husimi Q-distribution

$$Q(\phi, n) = \frac{1}{2\pi} \langle \alpha = \frac{\phi + ni}{\sqrt{2}} | \overline{\rho_{\omega_{\text{LO}}}} | \alpha = \frac{\phi + ni}{\sqrt{2}} \rangle. \quad (\text{A19})$$

The analysis above partially explains our particular choice of the interaction picture: the frame choice depends on the local oscillator frequency ω_{LO} of the IQ mixer. More explicitly, the free Hamiltonian can be written as

$$H_0 = \omega_{\text{LO}} a^\dagger a + \tilde{H}_0, \quad (\text{A20})$$

for some arbitrary hermitian $\tilde{H}_0$ so that $[\tilde{H}_0, a] = 0$. Although $\tilde{H}_0$ is quite arbitrary in principle, the remaining freedom in $\tilde{H}_0$ is usually fixed by minimizing coupling strengths in the interaction Hamiltonian in practice. Such choices will make the TCG perturbative expansion more likely to converge up to high orders. More precisely, if the interaction-picture Hamiltonian takes the form $H_I = \sum_k g_k e^{i\omega_k t} h_k$ for some coupling constants g_k , frequencies ω_k , and operators h_k , then the perturbative expansion of the TCG Liouvillian is only valid when either of the following two conditions are true:

$$\left[\frac{g_k}{\omega_k - \omega_{k'}} \ll 1 \quad \forall k' \right] \quad \text{or} \quad \left[g_k \tau \ll 1 \right] \quad \forall k. \quad (\text{A21})$$

With the frame choice understood, we will drop the subscript in $\overline{\rho_{\omega_{\text{LO}}}}(t)$ and simply denote the TCG density matrix by $\overline{\rho}(t)$ in the rest of this work.

Appendix B: Closed-form formula and diagrammatic representation of the TCG Liouvillian $\mathcal{L}_k$

We suppose that the interaction picture is chosen in such a way that H_I is controlled by some small parameters and may therefore be treated perturbatively to obtain an expansion of the Liouvillian superoperator $\mathcal{L}(t)$ in powers of H_I . In this section, we develop a relatively efficient method for explicitly calculating $\mathcal{L}(t)$ to arbitrary orders in this expansion.

To do this, we first expand the unitary time evolution operator $U(t) \equiv \mathcal{T} e^{-i \int_0^t dt' \cdot H_I(t')}$ as

$$\begin{aligned} U(t_0) &= 1 + \sum_{k=1}^{\infty} (-i)^k \int_0^{t_0} \cdots \int_0^{t_{k-1}} dt_k \cdot H_I(t_1) \cdots H_I(t_k) \\ &\equiv \sum_{k=0}^{\infty} U_k(t) \end{aligned} \quad (\text{B1})$$

where U_k is the k -th order time-evolution operator. And in terms of these operators, we can formally expand the time-coarse grained density matrix as

$$\bar{\rho}(t) = \sum_{k=0}^{\infty} \sum_{k_1=0}^k \overline{U_{k_1} \rho_0 U_{k-k_1}^\dagger}(t) \equiv \sum_{k=0}^{\infty} \mathcal{E}_k(t) \rho_0 \quad (\text{B2})$$

where $\mathcal{E}_k(t) \rho_0 = \sum_{k_1=0}^k \overline{U_{k_1} \rho_0 U_{k-k_1}^\dagger}(t)$ is considered a k -th order term. We can also formally expand the Liouvillian superoperator $\mathcal{L}(t)$ as $\mathcal{L}(t) = \sum_{k=1}^{\infty} \mathcal{L}_k(t)$ in powers of the interaction Hamiltonian. In order to obtain a functional equation for $\mathcal{L}_k$, we plug Eq.(B2) into Eq.(3) of the Main Text:

$$\begin{aligned} \partial_t \bar{\rho}(t) &= -i \sum_{k=1}^{\infty} \sum_{k_1=1}^k \left(\overline{H_I U_{k_1-1} \rho_0 U_{k-k_1}^\dagger}(t) - \overline{U_{k_1-1} \rho_0 U_{k-k_1}^\dagger} H_I(t) \right) \\ &= \sum_{j=1}^{\infty} \mathcal{L}_j(t) \bar{\rho}(t) = \sum_{j=1}^{\infty} \mathcal{L}_j(t) \sum_{k=0}^{\infty} \mathcal{E}_k(t) \rho_0 = \sum_{j=1}^{\infty} \mathcal{L}_j(t) \sum_{k=j=0}^{\infty} \mathcal{E}_{k-j}(t) \rho_0 \\ &= \sum_{k=1}^{\infty} \sum_{j=1}^k \mathcal{L}_j(t) \mathcal{E}_{k-j}(t) \rho_0 \end{aligned} \quad (\text{B3})$$

Matching terms that are at order H_I^k , expanding $\mathcal{E}_{k-j}(t)$ according to Eq.(B2), and replacing ρ_0 by a generic density matrix ρ , we find the following recurrence relation for any $k \geq 1$

$$\begin{aligned} i \mathcal{L}_k(t) \rho &= \sum_{k_1=1}^k \left(\overline{H_I U_{k_1-1} \rho U_{k-k_1}^\dagger}(t) - \overline{U_{k_1-1} \rho U_{k-k_1}^\dagger} H_I(t) \right) \\ &\quad - i \sum_{j=0}^{k-1} \mathcal{L}_j(t) \sum_{k_1=0}^{k-j} \overline{U_{k_1} \rho U_{k-j-k_1}^\dagger}(t) \end{aligned} \quad (\text{B4})$$

where we define $\mathcal{L}_0 \equiv 0$. Using this recurrence relation, one can calculate $\mathcal{L}_k(t)$ to all orders in H_I . More explicitly, one has

$$\overline{H_I U_k} = \overline{H_I U_k} - \sum_{j=0}^k \overline{H_I U_{j-1} U_{k+1-j}} \quad (\text{B5})$$

and

$$\overline{H_I U_{k_1-1} \rho U_{k+1-k_1}^\dagger}(t) = \overline{H_I U_{k_1-1} \rho U_{k+1-k_1}^\dagger}(t) - \sum_{j=1}^k \sum_{k_2=\max(0, k_1-j)}^{\min(k_1-1, k+1-j)} \overline{H_I U_{k_1-k_2-1} U_{k_2} \rho U_{k+1-j-k_2}^\dagger U_{j+k_2-k_1}^\dagger}(t) \quad (\text{B6})$$

where ρ is an arbitrary density matrix and does not participate in any of the time averaging or contraction operations.

If the interaction-picture Hamiltonian takes the form $H_I(t) = \sum_j e^{i\omega_j t} h_j$, then one can derive a number of explicit analytical formulas which will be useful for developing the closed-form expressions for the superoperator coefficients discussed in Section II of the Main Text. For example, the n -th order time evolution operator $U_n(t)$ can be explicitly written as

$$U_n(t) = -i \int_0^t dt' \cdot H_I(t') U_{n-1}(t') = \sum_{k_1, \dots, k_n} \sum_{j=0}^n \frac{(-1)^j e^{i(\omega_{k_1} + \omega_{k_2} + \dots + \omega_{k_j})t}}{(\omega_{k_j}, \omega_{k_{j-1}}, \dots, \omega_{k_1})! \cdot (\omega_{k_{j+1}}, \omega_{k_{j+2}}, \dots, \omega_{k_n})!} h_{k_1} h_{k_2} \dots h_{k_n} \quad (\text{B7})$$

where we define the factorial of a vector $\vec{\omega} = (\omega_1, \omega_2, \dots, \omega_n)$ as

$$\vec{\omega}! \equiv (\omega_1 + \omega_2 + \dots + \omega_n)(\omega_1 + \omega_2 + \dots + \omega_{n-1}) \dots \omega_1 \quad (\text{B8})$$

with the factorial of a zero-length vector defined to be unity, i.e., $\vec{0}! \equiv 1$.

On the other hand, when calculating the TCG master equation according to the recursive relations in Eq.(B4), one needs to calculate a large number of time averages of the form $\overline{H_I U_{k_1-1} \rho U_{k-k_1}^\dagger}(t)$ which does not consist of operators with single-frequency time dependence when $k \geq 2$. For example, one has

$$\overline{H_I U_1(t)} = \sum_{l_1, l_2} \left(- \frac{\tilde{f}(\omega_{l_1} + \omega_{l_2}) e^{i(\omega_{l_1} + \omega_{l_2})t} - \tilde{f}(\omega_{l_1}) e^{i\omega_{l_1}t}}{\omega_{l_2}} \right) h_{l_1} h_{l_2}. \quad (\text{B9})$$

for any operators $e^{i\omega_1 t} h_1$ and $e^{i\omega_2 t} h_2$ in the Hamiltonian. The complicated structure of $\overline{H_I U_{k_1-1} \rho U_{k-k_1}^\dagger}(t)$ for $k > 2$ makes it difficult to work with. However, one can group together all the terms in $\mathcal{L}_k(t)\rho$ which take the form of a c-number-valued function of time multiplied to a certain operator product of the form $h_{l_1} h_{l_2} \cdots h_{l_{k_1}} \rho h_{l_{k_1+1}} \cdots h_{l_k}$, and define their sum as the contraction

$$\overline{H_I U_{k_1-1} \rho U_{k-k_1}^\dagger}(t). \quad (\text{B10})$$

It then turns out that for any operator product $h_{l_1} h_{l_2} \cdots h_{l_{k_1}} \rho h_{l_{k_1+1}} \cdots h_{l_k}$ that appears in such a contraction, its time dependence is always a single-frequency evolution at the sum of the constituent operator frequencies $\omega_{l_1} + \cdots + \omega_{l_k}$. For example, one has

$$\overline{H_I U_1}(t) = \sum_{l_1, l_2} \left(-\frac{\tilde{f}(\omega_{l_1} + \omega_{l_2}) - \tilde{f}(\omega_{l_1}) \tilde{f}(\omega_{l_2})}{\omega_{l_2}} \right) e^{i(\omega_{l_1} + \omega_{l_2})t} h_{l_1} h_{l_2} \quad (\text{B11})$$

in contrast with Eq.(B9). More generally, for any $k_1 \geq 1$, the contractions will take the form

$$\overline{H_I U_{k_1-1} \rho U_{k-k_1}^\dagger}(t) = \sum_{l_1 \cdots l_k} C(\omega_{l_1}, \cdots, \omega_{l_k}; k_1) e^{i(\omega_{l_1} + \cdots + \omega_{l_k})t} h_{l_1} \cdots h_{l_{k_1}} \rho h_{l_{k_1+1}} \cdots h_{l_k} \quad (\text{B12})$$

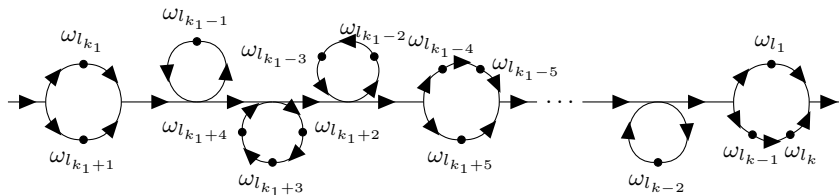
for some c-number coefficient $C(\omega_{l_1}, \cdots, \omega_{l_k}; k_1)$ whose explicit expression can be written as

$$C(\omega_{l_1}, \cdots, \omega_{l_k}; k_1) = \sum_{\sigma \in \Pi(k_1, k-k_1)} (-1)^{k_1 + |\sigma|} \text{sum}[\sigma_\uparrow^{\sigma}(\omega_{l_1}, \omega_{l_2}, \cdots, \omega_{l_k})] \prod_{j=1}^{|\sigma|} \frac{\tilde{f}(\sum_{\omega \in \sigma_\uparrow^j}(\omega_{l_1}, \omega_{l_2}, \cdots, \omega_{l_k}) \omega)}{\sigma_\uparrow^j(\omega_{l_1}, \omega_{l_2}, \cdots, \omega_{l_k})! \cdot \sigma_\downarrow^j(\omega_{l_1}, \omega_{l_2}, \cdots, \omega_{l_k})!} \quad (\text{B13})$$

where $\Pi(k_1, k-k_1)$ is the set of all ordered partitions of the 2-tuple $\begin{pmatrix} k_1 \\ k-k_1 \end{pmatrix}$ with the upper component of the last part being nonzero, $\sigma_\uparrow^j$ and $\sigma_\downarrow^j$ represent the upper and lower elements of the j -th part in the partition respectively¹, and for any $\sigma \in \Pi(k_1, k-k_1)$ we define

$$\begin{aligned} \sigma_\uparrow^j(\omega_{l_1}, \omega_{l_2}, \cdots, \omega_{l_k}) &\equiv (\omega_{l_{k_1 - \sigma_\uparrow^j - 1} \cdots \sigma_\uparrow^j - 1}, \omega_{l_{k_1 - \sigma_\uparrow^j - 1} \cdots \sigma_\uparrow^j - 1}, \cdots, \omega_{l_{k_1 - \sigma_\uparrow^j - 1} \cdots \sigma_\uparrow^j + 1}) \\ \sigma_\downarrow^j(\omega_{l_1}, \omega_{l_2}, \cdots, \omega_{l_k}) &\equiv (\omega_{l_{k_1 + 1 + \sigma_\downarrow^j + 1} \cdots \sigma_\downarrow^j - 1}, \omega_{l_{k_1 + 1 + \sigma_\downarrow^j + 1} \cdots \sigma_\downarrow^j - 1}, \cdots, \omega_{l_{k_1 + 1 + \sigma_\downarrow^j + 1} \cdots \sigma_\downarrow^j - 1}). \end{aligned} \quad (\text{B14})$$

Noticeably, every operator product in the contraction $\overline{H_I U_{k_1-1} \rho U_{k-k_1}^\dagger}(t)$ has single-frequency time dependence, unlike the time average $\overline{H_I U_{k_1-1} \rho U_{k-k_1}^\dagger}(t)$ which is much more cumbersome to work with. More visually, the coefficient $C(\omega_{l_1}, \cdots, \omega_{l_k}; k_1)$ can be associated with and computed according to the sum of diagrams of the form:



which corresponds to the following ordered partition of $\begin{pmatrix} k_1 \\ k-k_1 \end{pmatrix}$:

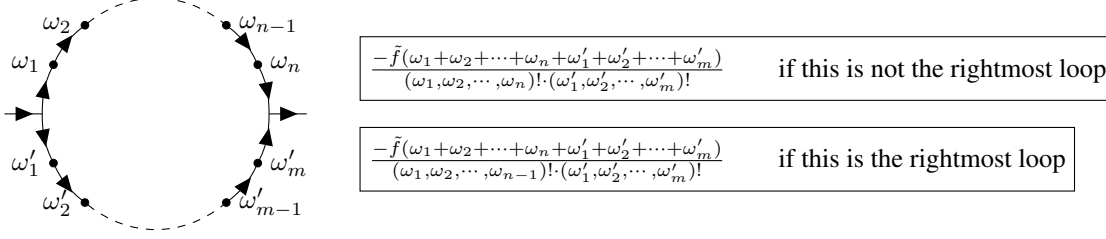
$$\left[\begin{pmatrix} 1 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ 3 \end{pmatrix}, \begin{pmatrix} 2 \\ 1 \end{pmatrix}, \cdots, \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 2 \end{pmatrix} \right].$$

¹ For example,

$$\begin{aligned} \Pi(2, 2) = \{ & \left[\begin{pmatrix} 2 \\ 2 \end{pmatrix} \right], \left[\begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 1 \\ 2 \end{pmatrix} \right], \left[\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 2 \\ 1 \end{pmatrix} \right], \left[\begin{pmatrix} 1 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 1 \end{pmatrix} \right], \left[\begin{pmatrix} 0 \\ 2 \end{pmatrix}, \begin{pmatrix} 2 \\ 0 \end{pmatrix} \right], \left[\begin{pmatrix} 1 \\ 2 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right], \left[\begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 1 \end{pmatrix} \right], \\ & \left[\begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ 2 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right], \left[\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 1 \\ 1 \end{pmatrix} \right], \left[\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right], \left[\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 2 \\ 0 \end{pmatrix} \right], \left[\begin{pmatrix} 1 \\ 1 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right], \\ & \left[\begin{pmatrix} 0 \\ 2 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right], \left[\begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right], \left[\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right], \left[\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \begin{pmatrix} 1 \\ 0 \end{pmatrix} \right] \} \end{aligned}$$

whereas $\sigma_\uparrow^2 = 2$ and $\sigma_\downarrow^2 = 1$ for $\sigma = \left[\begin{pmatrix} 0 \\ 1 \end{pmatrix}, \begin{pmatrix} 2 \\ 1 \end{pmatrix} \right]$.

Then, in addition to the overall sign $(-1)^{k_1}$, every loop in such a diagram will contribute a multiplicative factor according to the following correspondence:



To prove Eq.(B12) and Eq.(B13), notice first that these two equations are trivially true for $k = 1$; if Eq.(B12,B13) hold true up to some k , then the recurrence relation in Eq.(B4) implies that

$$\begin{aligned}
 & \overline{H_I U_k}(t) \\
 &= \sum_{l_1, \dots, l_{k+1}} \frac{(-1)^{k+1+1} \tilde{f}(\omega_{l_1} + \dots + \omega_{l_{k+1}})(\omega_{l_1} + \dots + \omega_{l_{k+1}})}{(\omega_{l_{k+1}}, \omega_{l_{k+1}-1}, \dots, \omega_{l_1})!} e^{i(\omega_{l_1} + \dots + \omega_{l_{k+1}})t} h_{l_1} \dots h_{l_{k+1}} \\
 &+ \sum_{l_1, \dots, l_{k+1}} \sum_{\sigma \in \Pi(k+1,0)}^{| \sigma | \geq 2} (-1)^{k+1+| \sigma |} \text{sum}[\sigma^{| \sigma |}(\omega_{l_1}, \dots, \omega_{l_{k+1}})] \prod_{j=1}^{| \sigma |} \frac{\tilde{f}(\sum_{\omega \in \sigma_{\uparrow\downarrow}^j}(\omega_{l_1}, \dots, \omega_{l_{k+1}}) \omega)}{\sigma_{\uparrow}^j(\omega_{l_1}, \dots, \omega_{l_{k+1}})!} \\
 &\quad \cdot e^{i(\omega_{l_1} + \dots + \omega_{l_{k+1}})t} h_{l_1} \dots h_{l_{k+1}} \\
 &= \sum_{l_1, \dots, l_{k+1}} \sum_{\sigma \in \Pi(k+1,0)} (-1)^{k+1+| \sigma |} \text{sum}[\sigma^{| \sigma |}(\omega_{l_1}, \dots, \omega_{l_{k+1}})] \prod_{j=1}^{| \sigma |} \frac{\tilde{f}(\sum_{\omega \in \sigma_{\uparrow\downarrow}^j}(\omega_{l_1}, \dots, \omega_{l_{k+1}}) \omega)}{\sigma_{\uparrow}^j(\omega_{l_1}, \dots, \omega_{l_{k+1}})!} \\
 &\quad \cdot e^{i(\omega_{l_1} + \dots + \omega_{l_{k+1}})t} h_{l_1} \dots h_{l_{k+1}}
 \end{aligned} \tag{B15}$$

if $k_1 = k + 1$; whereas for $1 \leq k_1 \leq k$, we have

$$\begin{aligned}
 & \overline{H_I U_{k_1-1} \rho U_{k+1-k_1}^\dagger}(t) \\
 &= \sum_{l_1, \dots, l_{k+1}} \left[(-1)^{k_1+1} \frac{\tilde{f}(\omega_{l_1} + \dots + \omega_{l_{k_1}} + \omega_{l_{k_1+1}} + \dots + \omega_{l_{k+1}}) \cdot (\omega_{l_1} + \dots + \omega_{l_{k_1}})}{(\omega_{l_{k_1}}, \dots, \omega_{l_1})! \cdot (\omega_{l_{k_1+1}}, \dots, \omega_{l_{k+1}})!} \right. \\
 &\quad + \sum_{\sigma \in \Pi(k_1, k+1-k_1)}^{| \sigma | \geq 2} (-1)^{k_1+| \sigma |} \text{sum}[\sigma^{| \sigma |}(\omega_{l_1}, \dots, \omega_{l_{k+1}})] \prod_{j=1}^{| \sigma |} \frac{\tilde{f}(\sum_{\omega \in \sigma_{\uparrow\downarrow}^j}(\omega_{l_1}, \dots, \omega_{l_{k+1}}) \omega)}{\sigma_{\uparrow}^j(\omega_{l_1}, \dots, \omega_{l_{k+1}})! \cdot \sigma_{\downarrow}^j(\omega_{l_1}, \dots, \omega_{l_{k+1}})!} \left. \right] \\
 &\quad \cdot e^{i(\omega_{l_1} + \dots + \omega_{l_{k+1}})t} h_{l_1} \dots h_{l_{k_1}} \rho h_{l_{k_1+1}} \dots h_{l_{k+1}} \\
 &= \sum_{l_1, \dots, l_{k+1}} \sum_{\sigma \in \Pi(k_1, k+1-k_1)} (-1)^{k_1+| \sigma |} \text{sum}[\sigma^{| \sigma |}(\omega_{l_1}, \dots, \omega_{l_{k+1}})] \prod_{j=1}^{| \sigma |} \frac{\tilde{f}(\sum_{\omega \in \sigma_{\uparrow\downarrow}^j}(\omega_{l_1}, \dots, \omega_{l_{k+1}}) \omega)}{\sigma_{\uparrow}^j(\omega_{l_1}, \dots, \omega_{l_{k+1}}; k_1)! \cdot \sigma_{\downarrow}^j(\omega_{l_1}, \dots, \omega_{l_{k+1}})!} \\
 &\quad \cdot e^{i(\omega_{l_1} + \dots + \omega_{l_{k+1}})t} h_{l_1} \dots h_{l_{k_1}} \rho h_{l_{k_1+1}} \dots h_{l_{k+1}}.
 \end{aligned} \tag{B16}$$

Therefore we see that Eq.(B12) and Eq.(B13) are also true for $k + 1$, and by induction we have proven Eq.(B12,B13) for any positive integer k in general.

In addition, we can rearrange the terms to write the TCG master equation as the sum of Hamiltonian commutators and Lindblad-type dissipators with possibly negative rates, i.e.,

$$\partial_t \bar{\rho}(t) = \mathcal{L}(t) \bar{\rho}(t) = \sum_{k=1}^{\infty} \mathcal{L}_k(t) \bar{\rho}(t) \equiv -i \left[\sum_{k=1}^{\infty} H_{\text{eff}}^{(k)}(t), \bar{\rho}(t) \right] + \sum_{k=1}^{\infty} D_{\text{eff}}^{(k)}(t) \bar{\rho}(t) \tag{B17}$$

with

$$\begin{aligned}
i\mathcal{L}_k(t)\rho &= \sum_{k_1=1}^k \overbrace{H_I U_{k_1-1} \rho U_{k-k_1}^\dagger}^{\text{}}(t) - h.c. \\
&= \sum_{k_1=1}^k \sum_{l_1 \dots l_k} C(\omega_{l_1}, \dots, \omega_{l_k}; k_1) e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} h_{l_1} \dots h_{l_{k_1}} \rho h_{l_{k_1+1}} \dots h_{l_k} \\
&\quad - \sum_{k_1=1}^k \sum_{l_1 \dots l_k} C(\omega_{l_1}, \dots, \omega_{l_k}; k_1) e^{-i(\omega_{l_1} + \dots + \omega_{l_k})t} h_{l_k}^\dagger \dots h_{l_{k_1+1}}^\dagger \rho h_{l_{k_1}}^\dagger \dots h_{l_1}^\dagger \\
&= \sum_{l_1 \dots l_k} C(\omega_{l_1}, \dots, \omega_{l_k}; k) e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} h_{l_1} \dots h_{l_k} \rho - \sum_{l_1 \dots l_k} C(-\omega_{l_k}, \dots, -\omega_{l_1}; k) e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} \rho h_{l_1} \dots h_{l_k} \\
&\quad + \sum_{k_1=1}^{k-1} \sum_{l_1 \dots l_k} \left[C(\omega_{l_1}, \dots, \omega_{l_k}; k_1) - C(-\omega_{l_k}, \dots, -\omega_{l_1}; k - k_1) \right] e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} h_{l_1} \dots h_{l_{k_1}} \rho h_{l_{k_1+1}} \dots h_{l_k} \\
&= \sum_{l_1 \dots l_k} C(\omega_{l_1}, \dots, \omega_{l_k}; k) e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} h_{l_1} \dots h_{l_k} \rho - \sum_{l_1 \dots l_k} C(-\omega_{l_k}, \dots, -\omega_{l_1}; k) e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} \rho h_{l_1} \dots h_{l_k} \\
&\quad + \sum_{k_1=1}^{k-1} \sum_{l_1 \dots l_k} \left[C(\omega_{l_1}, \dots, \omega_{l_k}; k_1) - C(-\omega_{l_k}, \dots, -\omega_{l_1}; k - k_1) \right] e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} D_{h_{l_1} \dots h_{l_{k_1}}, h_{l_{k_1+1}} \dots h_{l_k}} \rho \\
&\quad + \sum_{k_1=1}^{k-1} \sum_{l_1 \dots l_k} \frac{C(\omega_{l_1}, \dots, \omega_{l_{k_1}}, \omega_{l_{k_1+1}}, \dots, \omega_{l_k}; k_1)}{2} e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} \left\{ h_{l_{k_1+1}} \dots h_{l_k} h_{l_1} \dots h_{l_{k_1}}, \rho \right\} \\
&\quad - \sum_{k_1=1}^{k-1} \sum_{l_1 \dots l_k} \frac{C(-\omega_{l_k}, \dots, -\omega_{l_{k-k_1+1}}, -\omega_{l_{k-k_1}}, \dots, -\omega_{l_1}; k_1)}{2} e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} \left\{ h_{l_{k-k_1+1}} \dots h_{l_k} h_{l_1} \dots h_{l_{k-k_1}}, \rho \right\} \\
&= \sum_{l_1 \dots l_k} C(\omega_{l_1}, \dots, \omega_{l_k}; k) e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} h_{l_1} \dots h_{l_k} \rho - \sum_{l_1 \dots l_k} C(-\omega_{l_k}, \dots, -\omega_{l_1}; k) e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} \rho h_{l_1} \dots h_{l_k} \\
&\quad + \sum_{k_1=1}^{k-1} \sum_{l_1 \dots l_k} \left[C(\omega_{l_1}, \dots, \omega_{l_k}; k_1) - C(-\omega_{l_k}, \dots, -\omega_{l_1}; k - k_1) \right] e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} D_{h_{l_1} \dots h_{l_{k_1}}, h_{l_{k_1+1}} \dots h_{l_k}} \rho \\
&\quad - \sum_{l_1 \dots l_k} \frac{C(\omega_{l_1}, \dots, \omega_{l_k}; k)}{2} e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} \left\{ h_{l_1} \dots h_{l_k}, \rho \right\} + \sum_{l_1 \dots l_k} \frac{C(-\omega_{l_k}, \dots, -\omega_{l_1}; k)}{2} e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} \left\{ h_{l_1} \dots h_{l_k}, \rho \right\} \\
&\quad + \sum_{k_1=1}^k \sum_{l_1 \dots l_k} \frac{C(\omega_{l_k}, \omega_{l_1}, \dots, \omega_{l_{k-1}}; k_1) - C(-\omega_{l_k}, \dots, -\omega_{l_1}; k - k_1 + 1)}{2} e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} \left\{ h_{l_{k_1}} \dots h_{l_k} h_{l_1} \dots h_{l_{k_1-1}}, \rho \right\} \\
&= \left[\sum_{l_1 \dots l_k} \frac{C(\omega_{l_1}, \dots, \omega_{l_k}; k) + C(-\omega_{l_k}, \dots, -\omega_{l_1}; k)}{2} e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} h_{l_1} \dots h_{l_k}, \rho \right] \\
&\quad + \sum_{k_1=1}^{k-1} \sum_{l_1 \dots l_k} \left[C(\omega_{l_1}, \dots, \omega_{l_k}; k_1) - C(-\omega_{l_k}, \dots, -\omega_{l_1}; k - k_1) \right] e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} D_{h_{l_1} \dots h_{l_{k_1}}, h_{l_{k_1+1}} \dots h_{l_k}} \rho \\
&\equiv \left[H_{\text{eff}}^{(k)}(t), \rho \right] + iD_{\text{eff}}^{(k)}(t)\rho
\end{aligned} \tag{B18}$$

where we have defined the (generalized) dissipator $D_{L,R}$ as

$$D_{L,R}\rho \equiv L\rho R - \frac{RL\rho + \rho RL}{2}$$

for any operators L and R . In the derivation of Eq.(B18) above, we have used the identity

$$C(\omega_{l_k}, \omega_{l_1}, \dots, \omega_{l_{k-1}}; k_1) = C(-\omega_{l_k}, \dots, -\omega_{l_1}; k - k_1 + 1) \tag{B19}$$

for any $k_1 \in \{1, 2, \dots, k\}$, which can be derived from the explicit expression of C as follows:

$$\begin{aligned}
& C(\omega_{l_k}, \omega_{l_1}, \dots, \omega_{l_{k-1}}; k_1) \\
&= \sum_{\sigma \in \Pi(k_1, k-k_1)} (-1)^{k_1+|\sigma|} \text{sum}[\sigma_{\uparrow}^{|\sigma|}(\omega_{l_k}, \omega_{l_1}, \dots, \omega_{l_{k-1}})] \prod_{j=1}^{|\sigma|} \frac{\tilde{f}(\sum_{\omega \in \sigma_{\uparrow}^j}(\omega_{l_k}, \omega_{l_1}, \dots, \omega_{l_{k-1}}) \omega)}{\sigma_{\uparrow}^j(\omega_{l_k}, \omega_{l_1}, \dots, \omega_{l_{k-1}})! \cdot \sigma_{\downarrow}^j(\omega_{l_k}, \omega_{l_1}, \dots, \omega_{l_{k-1}})!} \\
&= \sum_{\sigma \in \Pi(k-k_1+1, k_1-1)} (-1)^{k_1+|\sigma|} \text{sum}[\sigma_{\uparrow}^{|\sigma|}(\omega_{l_k}, \omega_{l_{k-1}}, \dots, \omega_{l_1})] \prod_{j=1}^{|\sigma|} \frac{\tilde{f}(\sum_{\omega \in \sigma_{\uparrow}^j}(\omega_{l_k}, \omega_{l_{k-1}}, \dots, \omega_{l_1}) \omega)}{\sigma_{\uparrow}^j(\omega_{l_k}, \omega_{l_{k-1}}, \dots, \omega_{l_1})! \cdot \sigma_{\downarrow}^j(\omega_{l_k}, \omega_{l_{k-1}}, \dots, \omega_{l_1})!} \\
&= \sum_{\sigma \in \Pi(k-k_1+1, k_1-1)} (-1)^{k-k_1+1+|\sigma|} \text{sum}[\sigma_{\uparrow}^{|\sigma|}(-\omega_{l_k}, \dots, -\omega_{l_1})] \prod_{j=1}^{|\sigma|} \frac{\tilde{f}(\sum_{\omega \in \sigma_{\uparrow}^j}(-\omega_{l_k}, \dots, -\omega_{l_1}) \omega)}{\sigma_{\uparrow}^j(-\omega_{l_k}, \dots, -\omega_{l_1})! \cdot \sigma_{\downarrow}^j(-\omega_{l_k}, \dots, -\omega_{l_1})!} \\
&= C(-\omega_{l_k}, \dots, -\omega_{l_1}; k-k_1+1).
\end{aligned} \tag{B20}$$

Notice that we have employed the following bijection between $\Pi(k_1, k-k_1)$ and $\Pi(k-k_1+1, k_1-1)$ in obtaining the second equality:

$$\begin{aligned}
& \left[\begin{pmatrix} \sigma_{\uparrow}^1 \\ \sigma_{\downarrow}^1 \end{pmatrix}, \begin{pmatrix} \sigma_{\uparrow}^2 \\ \sigma_{\downarrow}^2 \end{pmatrix}, \dots, \begin{pmatrix} \sigma_{\uparrow}^{j-1} \\ \sigma_{\downarrow}^{j-1} \end{pmatrix}, \begin{pmatrix} \sigma_{\uparrow}^j \\ \sigma_{\downarrow}^j \end{pmatrix} \right] \\
& \quad \updownarrow \\
& \left[\begin{pmatrix} \sigma_{\downarrow}^1 \\ \sigma_{\uparrow}^1 \end{pmatrix}, \begin{pmatrix} \sigma_{\downarrow}^2 \\ \sigma_{\uparrow}^2 \end{pmatrix}, \dots, \begin{pmatrix} \sigma_{\downarrow}^{j-1} \\ \sigma_{\uparrow}^{j-1} \end{pmatrix}, \begin{pmatrix} \sigma_{\downarrow}^j + 1 \\ \sigma_{\uparrow}^j - 1 \end{pmatrix} \right]
\end{aligned}$$

In conclusion, we have

$$H_{\text{eff}}^{(k)}(t) = \sum_{l_1 \dots l_k} \frac{C(\omega_{l_1}, \dots, \omega_{l_k}; k) + C(-\omega_{l_k}, \dots, -\omega_{l_1}; k)}{2} e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} h_{l_1} \dots h_{l_k} \tag{B21}$$

and

$$D_{\text{eff}}^{(k)}(t) = -i \sum_{k_1=1}^{k-1} \sum_{l_1 \dots l_k} \left[C(\omega_{l_1}, \dots, \omega_{l_k}; k_1) - C(-\omega_{l_k}, \dots, -\omega_{l_1}; k-k_1) \right] e^{i(\omega_{l_1} + \dots + \omega_{l_k})t} D_{h_{l_1} \dots h_{l_{k_1}}, h_{l_{k_1+1}} \dots h_{l_k}}. \tag{B22}$$

Finally, in situations where individual diagrams contain zero-valued frequency sums in their denominators, we can regularize the apparent singularities by shifting every operator frequency ω_i by an infinitesimal amount $d\omega$, summing over all the diagrams, and taking the $d\omega \rightarrow 0$ limit. In order to obtain an explicit formula for $C(\omega_1, \dots, \omega_{l+r}; l)$ which is also valid in the presence of singular diagrams, we can rewrite it as

$$C(\omega_1, \dots, \omega_{l+r}; l) = \sum_{d \in \text{diagrams}} (-1)^{l+||d||} \frac{\tilde{f}(\omega_1 + \sum \mu_1 + \sum \nu_1) \tilde{f}(\sum \mu_2 + \sum \nu_2) \dots \tilde{f}(\sum \mu_{||d||} + \sum \nu_{||d||})}{\mu_1! \nu_1! \mu_2! \nu_2! \dots \mu_{||d||}! \nu_{||d||}!} \tag{B23}$$

where $||d||$ is the number of loops in the diagram d , while $\mu_i \equiv (\mu_i^1, \dots, \mu_i^{l_i})$ and $\nu_i \equiv (\nu_i^1, \dots, \nu_i^{r_i})$ are the ordered sets of upper and lower frequencies in the i -th loop respectively (with ω_1 excluded and the rest of the frequencies ordered from left to right in the diagrams). Here the factorial of an ordered set is defined as

$$\mu_i! \equiv (\mu_i^1 + \dots + \mu_i^{l_i}) \dots (\mu_i^1 + \mu_i^2) \mu_i^1. \tag{B24}$$

When some of the factors in $\mu_i!$ are zero, the corresponding diagram will contain an apparent divergence which can be canceled by other diagrams. Therefore, we are only interested in the finite part of the diagram. To get the finite part, suppose that the following factors in $\mu_i!$ and $\nu_i!$ are zero:

$$\begin{aligned}
& (\mu_i^1 + \dots + \mu_i^{s_i^1}), & (\mu_i^1 + \dots + \mu_i^{s_i^2}), & \dots & , & (\mu_i^1 + \dots + \mu_i^{s_i^n}) \\
& (\nu_i^1 + \dots + \nu_i^{s_i'^1}), & (\nu_i^1 + \dots + \nu_i^{s_i'^2}), & \dots & , & (\nu_i^1 + \dots + \nu_i^{s_i'^n})
\end{aligned} \tag{B25}$$

where $s_i^1 > s_i^2 > \dots > s_i^n$ and $s_i'^1 > s_i'^2 > \dots > s_i'^n$ (also, we define $\mathbf{s}_i := (s_i^1, s_i^2, \dots, s_i^n)$ and $\mathbf{s}_i' := (s_i'^1, s_i'^2, \dots, s_i'^n)$); in addition, we denote the product of nonzero factors in the factorials by

$$\overline{\mu}_i! := \prod_{j=1, j \notin \mathbf{s}_i}^{l_i} (\mu_i^1 + \dots + \mu_i^j) \quad (\text{B26})$$

and

$$\overline{\nu}_i! := \prod_{j=1, j \notin \mathbf{s}_i'}^{r_i} (\nu_i^1 + \dots + \nu_i^j), \quad (\text{B27})$$

while defining the total frequency in each loop to be

$$\tilde{\mu}_i := \mu_i^{s_i^1+1} + \mu_i^{s_i^1+2} + \dots + \mu_i^{l_i} + \omega_1 \delta_{i,1}; \quad \tilde{\nu}_i := \nu_i^{s_i'^1+1} + \nu_i^{s_i'^1+2} + \dots + \nu_i^{r_i}. \quad (\text{B28})$$

Then the finite contribution of the diagram can be obtained from Taylor expansion (sending each frequency ω_i to $\omega_i + d\omega$ and expanding in powers of $d\omega$) to be:

$$(-1)^l \prod_{i=1}^{|d|} \frac{\tilde{f}(\tilde{\mu}_i + \tilde{\nu}_i)}{-\overline{\mu}_i! \overline{\nu}_i!} \cdot \sum_{n_i \geq 0, u_i \geq 0, l_i \geq 0}^{\sum_i (n_i + u_i + l_i) = \sum_i (|\mathbf{s}_i| + |\mathbf{s}_i'|) \lfloor \frac{n_i}{2} \rfloor} \sum_{k_i=0}^{\lfloor \frac{n_i}{2} \rfloor} \frac{c(n_i, k_i)}{(n_i)!} \tau^{2(n_i - k_i)} (\tilde{\mu}_i + \tilde{\nu}_i)^{n_i - 2k_i} (|\mu_i| + |\nu_i|)^{n_i} \quad (\text{B29})$$

$$\sum_{\sum m_{J_u} = u_i, \sum m'_{J_l} = l_i} \frac{\prod_{J_u \notin \mathbf{s}_i, J_l \notin \mathbf{s}_i'} \left(\frac{-J_u}{\mu_i^1 + \dots + \mu_i^{J_u}} \right)^{m_{J_u}} \left(\frac{-J_l}{\nu_i^1 + \dots + \nu_i^{J_l}} \right)^{m'_{J_l}}}{\prod_{s_i^{j_1} \in \mathbf{s}_i, s_i'^{j_2} \in \mathbf{s}_i'} (s_i^{j_1} s_i'^{j_2})},$$

where $\lfloor \bullet \rfloor$ is the floor function, and the function $c(n, k)$ can be defined recursively by the relations:

$$c(n+1, k) = -c(n, k) + (n-2k+2)c(n, k-1) \quad (\text{B30})$$

together with the conditions

$$c(0, 0) = 1, \quad c(n, -1) \equiv 0 \text{ for any } n, \quad c(n, k) \equiv 0 \text{ for } n < 2k. \quad (\text{B31})$$

Appendix C: Explicit formula for the TCG master equation in the presence of a bosonic thermal bath

As discussed in Section III of the Main Text, when the coarse-graining time scale τ is much greater than the bath correlation time, the leading-order (in the system-bath coupling) dynamical equation for the reduced TCG system density matrix $\bar{\rho}(t) \equiv \text{Tr}_B(\bar{\rho}_{\text{tot}}(t))$ takes the form of a Markovian master equation:

$$\partial_t \bar{\rho}(t) \approx \mathcal{L}^{(0)}(t) \bar{\rho}(t) + \text{Tr}_B \left[\mathcal{L}^{(2)}(t) [\bar{\rho}(t) \otimes \rho_B(0)] \right] + \int_0^\infty dt_1 \mathcal{G}^{(2)}(t, t-t_1) \bar{\rho}(t). \quad (\text{C1})$$

More explicitly, if we assume the type of system-bath couplings in Eq.(9) of the Main Text, then, according to Eq.(B21) and Eq.(B22), $i\mathcal{L}^{(1)}(t)$ can in general be written as

$$i\mathcal{L}^{(1)}(t)\rho = \int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega \left[\sum_k \tilde{g}_k^l(\omega) e^{i(\omega_k^l - \omega)t} D_{L_k^l B_\omega, R_k^l} \rho + \sum_k \tilde{g}_k^r(\omega) e^{i(\omega_k^r - \omega)t} D_{L_k^r B_\omega, R_k^r} - h.c. \right] \quad (\text{C2})$$

for some set of operators $L_k^r, L_k^l, R_k^r, R_k^l$ acting on the system Hilbert space together with their corresponding frequencies $\omega_k^{l(r)}$ and coefficients $g_k^{l(r)}$. With this expression and the identity

$$\int_0^\infty dt_1 e^{i(\omega - \omega_0)t} = \pi \delta(\omega - \omega_0) + i\mathcal{P} \frac{1}{\omega - \omega_0} \quad (\text{C3})$$

where $\mathcal{P}$ denotes the Cauchy principal value, we can write the leading-order time integral of $\mathcal{G}^{(2)}(t, t-t_1)$ in Eq.(C1) as

$$\begin{aligned} \int_0^\infty dt_1 \mathcal{G}^{(2)}(t, t-t_1) \bar{\rho}(t) &\approx -i [H_{\text{corr}}^{(2)}(t), \bar{\rho}(t)] + D_{\text{corr}}^{(2)}(t) \bar{\rho}(t) \\ &\equiv -i [H_{ll}(t) + H_{lr}(t) + H_{rl}(t) + H_{rr}(t), \bar{\rho}(t)] + (D_{ll}(t) + D_{lr}(t) + D_{rl}(t) + D_{rr}(t)) \bar{\rho}(t), \end{aligned} \quad (\text{C4})$$

and the explicit expressions for $H_{ll}, H_{lr}, H_{rl}, H_{rr}, D_{ll}, D_{lr}, D_{rl}, D_{rr}$ will be given by the formulas below:

$$H_{ll} = \sum_{k_1, k_2} e^{i(\omega_{k_1}^l - \omega_{k_2}^l)t} \left[i \frac{\mathcal{D}_{\omega_{k_1}^l} \tilde{g}_{k_1}^l(\omega_{k_1}^l) \tilde{g}_{k_2}^{l*}(\omega_{k_1}^l)}{2} \bar{n}(\omega_{k_1}^l) - \mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^l(\omega) \tilde{g}_{k_2}^{l*}(\omega)}{2\pi} \frac{\bar{n}(\omega)}{\omega - \omega_{k_1}^l} \right] \frac{[L_{k_2}^{l\dagger} R_{k_2}^{l\dagger}, R_{k_1}^l L_{k_1}^l]}{8} + h.c. \quad (C5)$$

$$H_{lr} = \sum_{k_1, k_2} e^{i(\omega_{k_1}^r - \omega_{k_2}^l)t} \left[i \frac{\mathcal{D}_{\omega_{k_1}^r} \tilde{g}_{k_1}^r(\omega_{k_1}^r) \tilde{g}_{k_2}^{l*}(\omega_{k_1}^r)}{2} \bar{n}(\omega_{k_1}^r) - \mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^r(\omega) \tilde{g}_{k_2}^{l*}(\omega)}{2\pi} \frac{\bar{n}(\omega)}{\omega - \omega_{k_1}^r} \right] \frac{[L_{k_2}^{l\dagger} R_{k_2}^{l\dagger}, R_{k_1}^r L_{k_1}^r]}{8} + h.c. \quad (C6)$$

$$H_{rl} = \sum_{k_1, k_2} e^{i(\omega_{k_1}^l - \omega_{k_2}^r)t} \left[i \frac{\mathcal{D}_{\omega_{k_1}^l} \tilde{g}_{k_1}^l(\omega_{k_1}^l) \tilde{g}_{k_2}^{r*}(\omega_{k_1}^l)}{2} \bar{n}(\omega_{k_1}^l) - \mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^l(\omega) \tilde{g}_{k_2}^{r*}(\omega)}{2\pi} \frac{\bar{n}(\omega)}{\omega - \omega_{k_1}^l} \right] \frac{[L_{k_2}^{r\dagger} R_{k_2}^{r\dagger}, R_{k_1}^l L_{k_1}^l]}{8} + h.c. \quad (C7)$$

$$H_{rr} = \sum_{k_1, k_2} e^{i(\omega_{k_1}^r - \omega_{k_2}^r)t} \left[i \frac{\mathcal{D}_{\omega_{k_1}^r} \tilde{g}_{k_1}^r(\omega_{k_1}^r) \tilde{g}_{k_2}^{r*}(\omega_{k_1}^r)}{2} \bar{n}(\omega_{k_1}^r) - \mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^r(\omega) \tilde{g}_{k_2}^{r*}(\omega)}{2\pi} \frac{\bar{n}(\omega)}{\omega - \omega_{k_1}^r} \right] \frac{[L_{k_2}^{r\dagger} R_{k_2}^{r\dagger}, R_{k_1}^r L_{k_1}^r]}{8} + h.c. \quad (C8)$$

$$\begin{aligned} & D_{ll} \bar{\rho}(t) \\ &= \sum_{k_1, k_2} e^{i(\omega_{k_1}^l - \omega_{k_2}^l)t} \left[\frac{\mathcal{D}_{\omega_{k_1}^l} \tilde{g}_{k_1}^l(\omega_{k_1}^l) \tilde{g}_{k_2}^{l*}(\omega_{k_1}^l)}{2} \bar{n}(\omega_{k_1}^l) + i\mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^l(\omega) \tilde{g}_{k_2}^{l*}(\omega)}{2\pi} \frac{\bar{n}(\omega)}{\omega - \omega_{k_1}^l} \right] \\ & \quad \cdot \left(D_{R_{k_2}^{l\dagger} L_{k_1}^l, R_{k_1}^l L_{k_2}^{l\dagger}} - \frac{1}{2} D_{R_{k_2}^{l\dagger} R_{k_1}^l L_{k_1}^l, L_{k_2}^{l\dagger}} - \frac{1}{2} D_{L_{k_2}^{l\dagger} R_{k_2}^{l\dagger} L_{k_1}^l, R_{k_1}^l} - \frac{1}{2} D_{R_{k_2}^{l\dagger}, R_{k_1}^l L_{k_1}^l L_{k_2}^{l\dagger}} \right. \\ & \quad \left. - \frac{1}{2} D_{L_{k_1}^l, R_{k_1}^l L_{k_2}^{l\dagger} R_{k_2}^{l\dagger}} + \frac{1}{4} D_{R_{k_1}^l L_{k_1}^l, L_{k_2}^{l\dagger} R_{k_2}^{l\dagger}} \right) \bar{\rho}(t) \\ & + \sum_{k_1, k_2} e^{i(\omega_{k_1}^l - \omega_{k_2}^l)t} \left[\frac{\mathcal{D}_{\omega_{k_1}^l} \tilde{g}_{k_1}^l(\omega_{k_1}^l) \tilde{g}_{k_2}^{l*}(\omega_{k_1}^l)}{2} (\bar{n}(\omega_{k_1}^l) + 1) + i\mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^l(\omega) \tilde{g}_{k_2}^{l*}(\omega)}{2\pi} \frac{\bar{n}(\omega) + 1}{\omega - \omega_{k_1}^l} \right] \\ & \quad \cdot \frac{1}{4} D_{L_{k_2}^{l\dagger} R_{k_2}^{l\dagger}, R_{k_1}^l L_{k_1}^l} \bar{\rho}(t) \\ & + h.c. \end{aligned} \quad (C9)$$

$$\begin{aligned} & D_{lr} \bar{\rho}(t) \\ &= \sum_{k_1, k_2} e^{i(\omega_{k_1}^r - \omega_{k_2}^l)t} \left[\frac{\mathcal{D}_{\omega_{k_1}^r} \tilde{g}_{k_1}^r(\omega_{k_1}^r) \tilde{g}_{k_2}^{l*}(\omega_{k_1}^r)}{2} \bar{n}(\omega_{k_1}^r) + i\mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^r(\omega) \tilde{g}_{k_2}^{l*}(\omega)}{2\pi} \frac{\bar{n}(\omega)}{\omega - \omega_{k_1}^r} \right] \\ & \quad \cdot \left(D_{R_{k_2}^{l\dagger} L_{k_1}^r, R_{k_1}^r L_{k_2}^{l\dagger}} - \frac{1}{2} D_{R_{k_2}^{l\dagger} R_{k_1}^r L_{k_1}^r, L_{k_2}^{l\dagger}} - \frac{1}{2} D_{R_{k_2}^{l\dagger}, R_{k_1}^r L_{k_1}^r L_{k_2}^{l\dagger}} - \frac{1}{2} D_{L_{k_1}^r, R_{k_1}^r L_{k_2}^{l\dagger} R_{k_2}^{l\dagger}} + \frac{1}{4} D_{R_{k_1}^r L_{k_1}^r, L_{k_2}^{l\dagger} R_{k_2}^{l\dagger}} \right) \bar{\rho}(t) \\ & + \sum_{k_1, k_2} e^{i(\omega_{k_1}^r - \omega_{k_2}^l)t} \left[\frac{\mathcal{D}_{\omega_{k_1}^r} \tilde{g}_{k_1}^r(\omega_{k_1}^r) \tilde{g}_{k_2}^{l*}(\omega_{k_1}^r)}{2} (\bar{n}(\omega_{k_1}^r) + 1) + i\mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^r(\omega) \tilde{g}_{k_2}^{l*}(\omega)}{2\pi} \frac{\bar{n}(\omega) + 1}{\omega - \omega_{k_1}^r} \right] \\ & \quad \cdot \left(-\frac{1}{2} D_{L_{k_2}^{l\dagger} R_{k_2}^{l\dagger} L_{k_1}^r, R_{k_1}^r} + \frac{1}{4} D_{L_{k_2}^{l\dagger} R_{k_2}^{l\dagger}, R_{k_1}^r L_{k_1}^r} \right) \bar{\rho}(t) \\ & + h.c. \end{aligned} \quad (C10)$$

$$\begin{aligned}
& D_{rl}\bar{\rho}(t) \\
&= \sum_{k_1, k_2} e^{i(\omega_{k_1}^l - \omega_{k_2}^r)t} \left[\frac{\mathcal{D}_{\omega_{k_1}^l} \tilde{g}_{k_1}^l(\omega_{k_1}^l) \tilde{g}_{k_2}^{r*}(\omega_{k_1}^l)}{2} \bar{n}(\omega_{k_1}^l) + i\mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^l(\omega) \tilde{g}_{k_2}^{r*}(\omega)}{2\pi} \frac{\bar{n}(\omega)}{\omega - \omega_{k_1}^l} \right] \\
&\quad \cdot \left(D_{R_{k_2}^{\dagger} L_{k_1}^l, R_{k_1}^l L_{k_2}^{\dagger}} - \frac{1}{2} D_{R_{k_2}^{\dagger} R_{k_1}^l L_{k_1}^l, L_{k_2}^{\dagger}} - \frac{1}{2} D_{L_{k_2}^{\dagger} R_{k_2}^{\dagger} L_{k_1}^l, R_{k_1}^l} - \frac{1}{2} D_{L_{k_1}^l, R_{k_1}^l L_{k_2}^{\dagger} R_{k_2}^{\dagger}} + \frac{1}{4} D_{R_{k_1}^l L_{k_1}^l, L_{k_2}^{\dagger} R_{k_2}^{\dagger}} \right) \bar{\rho}(t) \\
&+ \sum_{k_1, k_2} e^{i(\omega_{k_1}^l - \omega_{k_2}^r)t} \left[\frac{\mathcal{D}_{\omega_{k_1}^l} \tilde{g}_{k_1}^l(\omega_{k_1}^l) \tilde{g}_{k_2}^{r*}(\omega_{k_1}^l)}{2} (\bar{n}(\omega_{k_1}^l) + 1) + i\mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^l(\omega) \tilde{g}_{k_2}^{r*}(\omega)}{2\pi} \frac{\bar{n}(\omega) + 1}{\omega - \omega_{k_1}^l} \right] \\
&\quad \cdot \left(-\frac{1}{2} D_{R_{k_2}^{\dagger}, R_{k_1}^l L_{k_1}^l L_{k_2}^{\dagger}} + \frac{1}{4} D_{L_{k_2}^{\dagger} R_{k_2}^{\dagger}, R_{k_1}^l L_{k_1}^l} \right) \bar{\rho}(t) \\
&+ h.c.
\end{aligned} \tag{C11}$$

$$\begin{aligned}
& D_{rr}\bar{\rho}(t) \\
&= \sum_{k_1, k_2} e^{i(\omega_{k_1}^r - \omega_{k_2}^r)t} \left[\frac{\mathcal{D}_{\omega_{k_1}^r} \tilde{g}_{k_1}^r(\omega_{k_1}^r) \tilde{g}_{k_2}^{r*}(\omega_{k_1}^r)}{2} \bar{n}(\omega_{k_1}^r) + i\mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^r(\omega) \tilde{g}_{k_2}^{r*}(\omega)}{2\pi} \frac{\bar{n}(\omega)}{\omega - \omega_{k_1}^r} \right] \\
&\quad \cdot \left(-\frac{1}{2} D_{L_{k_1}^r, R_{k_1}^r L_{k_2}^{\dagger} R_{k_2}^{\dagger}} - \frac{1}{2} D_{R_{k_2}^{\dagger} R_{k_1}^r L_{k_1}^r, L_{k_2}^{\dagger}} + \frac{1}{4} D_{R_{k_1}^r L_{k_1}^r, L_{k_2}^{\dagger} R_{k_2}^{\dagger}} \right) \bar{\rho}(t) \\
&+ \sum_{k_1, k_2} e^{i(\omega_{k_1}^r - \omega_{k_2}^r)t} \left[\frac{\mathcal{D}_{\omega_{k_1}^r} \tilde{g}_{k_1}^r(\omega_{k_1}^r) \tilde{g}_{k_2}^{r*}(\omega_{k_1}^r)}{2} (\bar{n}(\omega_{k_1}^r) + 1) + i\mathcal{P} \frac{\mathcal{D}_{\omega} \tilde{g}_{k_1}^r(\omega) \tilde{g}_{k_2}^{r*}(\omega)}{2\pi} \frac{\bar{n}(\omega) + 1}{\omega - \omega_{k_1}^r} \right] \\
&\quad \cdot \left(D_{R_{k_2}^{\dagger} L_{k_1}^r, R_{k_1}^r L_{k_2}^{\dagger}} - \frac{1}{2} D_{L_{k_2}^{\dagger} R_{k_2}^{\dagger} L_{k_1}^r, R_{k_1}^r} - \frac{1}{2} D_{R_{k_2}^{\dagger}, R_{k_1}^r L_{k_1}^r L_{k_2}^{\dagger}} + \frac{1}{4} D_{L_{k_2}^{\dagger} R_{k_2}^{\dagger}, R_{k_1}^r L_{k_1}^r} \right) \bar{\rho}(t) \\
&+ h.c.
\end{aligned} \tag{C12}$$

With these explicit formulas, it is straightforward to derive the time-coarse grained effective master equation for the system density matrix. In fact, all open quantum systems discussed in this work are modeled in this approach. And despite the seemingly complicated form of the expressions above, our calculations suggest that cancellation is often prevalent among the terms in Eq.(C5-C12), leading to only a few dissipators with distinctive physical interpretations (e.g. the Purcell decay or the Lamb shift) at low orders.

Appendix D: Time-coarse graining of more general open quantum systems

Our presentation of the TCG approach to open quantum systems in Section III of the Main Text assumes a single thermal bath consisting of bosonic degrees of freedom alone. Although this premise is sufficient for modeling many sources of dissipation in circuit QED, it is straightforward to generalize the previously presented recipe to include non-bosonic and/or multiple thermal baths. We first demonstrate the treatment of non-bosonic baths with a simple toy model consisting of a spin coupled to non-interacting spin modes.

1. Spin coupled to a spin bath

To demonstrate TCG's capability of modeling non-bosonic baths, we consider a minimal toy model described by the following total Hamiltonian in the Schrödinger-picture:

$$\hat{H}_{\text{tot}} = \frac{\omega_s}{2} \sigma_z + \int_0^\infty d\omega \cdot \mathcal{D}_\omega \frac{\omega}{2} \sigma_z^\omega + \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \cdot \sigma_x^\omega \sigma_x \tag{D1}$$

which models a spin linearly coupled to a non-interacting bath of spin degrees of freedom. The interaction Hamiltonian can be written as

$$H_{\text{tot}} = \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \cdot \left(e^{i(\omega + \omega_s)t} \sigma_+^\omega \sigma_+ + e^{i(\omega - \omega_s)t} \sigma_+^\omega \sigma_- + h.c. \right) \tag{D2}$$

Adopting the TCG-BM prescription discussed in the previous subsection, we can obtain the standard Lindblad master equation as well as its extensions to higher orders in g_ω if we assume that the coarse-graining time scale τ is sufficiently large ($\tau \gg 1/\omega_s$).

At leading orders in g_ω , we have

$$\mathcal{L}^{(1)}(t) = -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \cdot e^{-\frac{1}{2}(\omega-\omega_s)^2 \tau^2} \left[e^{i(\omega-\omega_s)t} \sigma_+^\omega \sigma_- + e^{-i(\omega-\omega_s)t} \sigma_-^\omega \sigma_+, \bullet \right] \quad (\text{D3a})$$

$$\begin{aligned} \mathcal{L}^{(2)}(t) = & -i \int_0^\infty \frac{d\omega_1 d\omega_2}{(2\pi)^2} \cdot \mathcal{D}_{\omega_1} \mathcal{D}_{\omega_2} g_{\omega_1} g_{\omega_2} \left[-e^{i(\omega_2-\omega_1)t} \frac{e^{-\frac{1}{2}(\omega_2-\omega_1)^2 \tau^2} (\omega_1 + \omega_2 + 2\omega_s)}{2(\omega_1 + \omega_s)(\omega_2 + \omega_s)} \sigma_- \sigma_+ \sigma_-^{\omega_1} \sigma_+^{\omega_2} \right. \\ & + e^{-i(\omega_2-\omega_1)t} \frac{(e^{-\frac{1}{2}(\omega_2-\omega_1)^2 \tau^2} - e^{-\frac{1}{2}((\omega_1-\omega_s)^2 + (\omega_2-\omega_s)^2) \tau^2}) (\omega_1 + \omega_2 - 2\omega_s)}{2(\omega_1 - \omega_s)(\omega_2 - \omega_s)} \sigma_- \sigma_+ \sigma_+^{\omega_1} \sigma_-^{\omega_2} \\ & - e^{i(\omega_2-\omega_1)t} \frac{(e^{-\frac{1}{2}(\omega_2-\omega_1)^2 \tau^2} - e^{-\frac{1}{2}((\omega_1-\omega_s)^2 + (\omega_2-\omega_s)^2) \tau^2}) (\omega_1 + \omega_2 - 2\omega_s)}{2(\omega_1 - \omega_s)(\omega_2 - \omega_s)} \sigma_+ \sigma_- \sigma_-^{\omega_1} \sigma_+^{\omega_2} \\ & \left. + e^{-i(\omega_2-\omega_1)t} \frac{e^{-\frac{1}{2}(\omega_2-\omega_1)^2 \tau^2} (\omega_1 + \omega_2 + 2\omega_s)}{2(\omega_1 + \omega_s)(\omega_2 + \omega_s)} \sigma_+ \sigma_- \sigma_+^{\omega_1} \sigma_-^{\omega_2}, \bullet \right] \end{aligned} \quad (\text{D3b})$$

and therefore

$$\text{Tr}_B \left[\mathcal{L}^{(2)}(t) [\bar{\rho}(t) \otimes \rho_B(0)] \right] = -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega^2 \left(-\frac{1 - e^{-(\omega-\omega_s)^2 \tau^2}}{\omega - \omega_s} + \frac{1}{\omega + \omega_s} \right) \left[\frac{\sigma_z}{2}, \bar{\rho}(t) \right] \quad (\text{D4a})$$

$$\int_0^\infty dt_1 \mathcal{G}^{(2)}(t, t-t_1) \bar{\rho}(t) \equiv \int_0^\infty dt_1 \text{Tr}_B [\mathcal{L}^{(1)}(t) \mathcal{L}^{(1)}(t-t_1) \rho_B(0)] \bar{\rho}(t) \quad (\text{D4b})$$

which leads to the following TCG master equation at order g_ω^2 according to the general formula in Section III of the Main Text:

$$\partial_t \bar{\rho}(t) \approx -i [\delta H^{(2)}, \bar{\rho}(t)] + \mathcal{D}_{\omega_s} g_\omega^2 \left(n_{\omega_s} D_{\sigma_+, \sigma_-} \bar{\rho}(t) + (1 - n_{\omega_s}) D_{\sigma_-, \sigma_+} \bar{\rho}(t) \right) \quad (\text{D5})$$

where $n_\omega \equiv \text{Tr}_B [\sigma_+^\omega \sigma_-^\omega]$ is the thermal bath occupation number at frequency ω which follows Dirac-Fermi statistics, and $\delta H^{(2)}$ is the effective Hamiltonian correction taking contributions from both Eq.(D4a) and Eq.(D4b):

$$\begin{aligned} \delta H^{(2)} &= \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega^2 \left(-\frac{1 - e^{-(\omega-\omega_s)^2 \tau^2}}{2(\omega - \omega_s)} + \frac{1}{2(\omega + \omega_s)} - \mathcal{P} \frac{e^{-(\omega-\omega_s)^2 \tau^2}}{2(\omega - \omega_s)} \right) \sigma_z \\ &= \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega^2 \left(\mathcal{P} \frac{1}{\omega_s - \omega} + \frac{1}{\omega + \omega_s} \right) \frac{\sigma_z}{2} \equiv \Delta_{\text{Lamb}} \frac{\sigma_z}{2} \end{aligned} \quad (\text{D6})$$

which gives us exactly the familiar Lamb-shift together with contributions from counter-rotating terms:

$$\Delta_{\text{Lamb}} = \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega^2 \left(\mathcal{P} \frac{1}{\omega_s - \omega} + \frac{1}{\omega + \omega_s} \right) \quad (\text{D7})$$

When the spin is only part of the quantum system of interest, it is not guaranteed in general that the right-hand side of Eq.(D5) still faithfully represents the effects of the thermal bath, even though the common practice is to assume that this is true when modeling interacting open quantum systems. As argued in the Main Text, we can regard such a practice as good approximation when all the internal system dynamics in the measurement frame is slow compared to the bath relaxation time scale. More generally, as we will show in Subsection D 3, TCG can give rise to corrections to the bath-induced dynamics, mixing the bath effects with the internal system dynamics.

2. Spin baths and multiple thermal environments

In this section we demonstrate the construction of effective master equations for multiple bosonic or spin baths via TCG. More concretely, we use the system + baths approach and consider the following total Hamiltonian in the Schrödinger picture:

$$\hat{H}_{\text{tot}}(t) = \hat{H}_s(t) + \hat{H}_f + \hat{H}_b + \hat{H}_{sf} + \hat{H}_{sb} \quad (\text{D8})$$

where $\hat{H}_s(t)$ is the system Hamiltonian while

$$\begin{aligned}\hat{H}_f &= \sum_{j=1,2,\dots,\|F\|} \sum_{k \in F^j} \varepsilon_k^j c_k^{j\dagger} c_k^j, & \hat{H}_b &= \sum_{l=1,2,\dots,\|B\|} \sum_{k \in B^l} \epsilon_k^l b_k^{l\dagger} b_k^l \\ \hat{H}_{sf} &= \sum_{j=1,2,\dots,\|F\|} \int_0^\infty \frac{d\omega}{2\pi} \bar{\mathcal{D}}_\omega^j \bar{g}_\omega^j \sum_{k \in F^j} \left(S_k^j c_\omega + S_k^{j\dagger} c_\omega^\dagger \right) \\ \hat{H}_{sb} &= \sum_{l=1,2,\dots,\|B\|} \int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega^l g_\omega^l \sum_{k \in B^l} \left(S_k^l b_\omega + S_k^{l\dagger} b_\omega^\dagger \right)\end{aligned}\tag{D9}$$

with S_k^j, S_k^l operators on the system part of the Hilbert space. In the interaction picture, the total Hamiltonian takes the form

$$\begin{aligned}H &= H_s(t) + \sum_{j=1,2,\dots,\|F\|} \int_0^\infty \frac{d\omega}{2\pi} \bar{\mathcal{D}}_\omega^j \bar{g}_\omega^j \sum_{k \in F^j} \left(e^{-i(\omega - \omega_k^j)t} S_k^j c_\omega + h.c. \right) \\ &+ \sum_{l=1,2,\dots,\|B\|} \int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega^l g_\omega^l \sum_{k \in B^l} \left(e^{-i(\omega - \omega_k^l)t} S_k^l b_\omega + h.c. \right)\end{aligned}\tag{D10}$$

Following the same analysis as in the Main Text, we perturbatively calculate the TCG Liouvillian superoperator $\mathcal{L}$ before tracing out the bath degrees of freedom. Expanding in powers of the system-bath coupling strengths g_ω^j , we rewrite $\mathcal{L}$ as

$$\mathcal{L}(t) = \sum_{\mathbf{n}^f \in \mathbb{Z}_{\geq 0}^{\|F\|}} \sum_{\mathbf{n}^b \in \mathbb{Z}_{\geq 0}^{\|B\|}} \mathcal{L}^{\mathbf{n}^f, \mathbf{n}^b}(t)\tag{D11}$$

where $\mathbf{n}^f := (n_1^f, n_2^f, \dots, n_{\|F\|}^f)$ denotes a list of $\|F\|$ non-negative integers and $\mathbf{n}^b := (n_1^b, n_2^b, \dots, n_{\|B\|}^b)$ denotes a list of $\|B\|$ non-negative integers, while $\mathcal{L}^{\mathbf{n}^f, \mathbf{n}^b}(t)$ is the part of the TCG Liouvillian containing $(n_1^f, n_2^f, \dots, n_{\|F\|}^f)$ spin bath coupling terms and $(n_1^b, n_2^b, \dots, n_{\|B\|}^b)$ bosonic bath coupling terms for each of the thermal baths respectively. We can then rewrite the leading-order time derivative of the coarse-grained system density matrix $\bar{\rho}$ as

$$\begin{aligned}\partial_t \bar{\rho}(t) &\approx \mathcal{L}^{0,0}(t) \bar{\rho}(t) + \sum_{j_f=1,\dots,\|F\|} \text{Tr}_F \left[\mathcal{L}^{(n_j^f=2\delta_{j,j_f}),0}(t) [\bar{\rho}(t) \otimes \rho_F(0)] \right] + \sum_{j_b=1,\dots,\|B\|} \text{Tr}_B \left[\mathcal{L}^{0,(n_j^b=2\delta_{j,j_b})}(t) [\bar{\rho}(t) \otimes \rho_B(0)] \right] \\ &+ \sum_{j_f=1,\dots,\|F\|} \int_0^\infty dt_1 \text{Tr}_F \left[\mathcal{L}^{(n_j^f=\delta_{j,j_f}),0}(t) (\mathcal{T} e^{\int_{t-t_1}^t dt_2 \mathcal{L}^{0,0}(t_2)}) \mathcal{L}^{(n_j^f=\delta_{j,j_f}),0}(t-t_1) \rho_F(0) \right] \bar{\rho}(t) \\ &+ \sum_{j_b=1,\dots,\|B\|} \int_0^\infty dt_1 \text{Tr}_B \left[\mathcal{L}^{0,(n_j^b=\delta_{j,j_b})}(t) (\mathcal{T} e^{\int_{t-t_1}^t dt_2 \mathcal{L}^{0,0}(t_2)}) \mathcal{L}^{0,(n_j^b=\delta_{j,j_b})}(t-t_1) \rho_B(0) \right] \bar{\rho}(t)\end{aligned}\tag{D12}$$

as long as the time resolution τ of feasible measurements are always much longer than the bath relaxation time scale. The prescription presented here shows that one can use the MaTCG framework to model a very general class of dissipative environments, not limited to the bosonic thermal baths assumed in the Main Text.

3. TCG corrections to the dissipative Rabi model

In this subsection we enlarge the quantum system (i.e. the spin) in Subsection D 1 by coupling it to a resonant cavity mode so that we end up with a resonant dissipative Rabi model. Naive application of the results in Subsection D 1 then suggests the following master equation in the interaction picture after the rotating-wave approximation:

$$\partial_t \rho(t) = -i \left[\frac{\eta}{2} (a \sigma_+ + a^\dagger \sigma_-) + \Delta_{\text{Lamb}} \frac{\sigma_z}{2}, \bar{\rho}(t) \right] + J(\omega_s) \left(n_{\omega_s} D_{\sigma_+, \sigma_-} \bar{\rho}(t) + (1 - n_{\omega_s}) D_{\sigma_-, \sigma_+} \bar{\rho}(t) \right)\tag{D13}$$

with $J(\omega) \equiv \mathcal{D}_\omega g_\omega^2$, where the bath-induced superoperators only act on the spin degrees of freedom regardless of the state of the cavity mode. This can be a good approximation if we limit our interest to the part of Hilbert space where the system dynamics (e.g. Rabi oscillations) are much slower than the bath relaxation rate. More generally however, our analysis in Section III of the Main Text shows that at leading order one should use the following master equation when one is only allowed to make

approximations based on the fact that the measurement time resolution τ (and not necessarily all the closed-system time scales) is much greater than the bath relaxation time τ_B :

$$\begin{aligned} \partial_t \bar{\rho}(t) &= \text{Tr}_B [\mathcal{L}(t) [\bar{\rho}(t) \otimes \rho_B(0)]] + \text{Tr}_B [\mathcal{L}(t) \bar{\chi}_{\text{corr}}(t)] \\ &= \mathcal{L}^{(0)}(t) \bar{\rho}(t) + \text{Tr}_B [\mathcal{L}^{(2)}(t) [\bar{\rho}(t) \otimes \rho_B(0)]] + \int_0^\infty dt_1 \mathcal{G}^{(2)}(t, t-t_1) \bar{\rho}(t) + \mathcal{O}(g^3) \end{aligned} \quad (\text{D14})$$

where the main difference compared to the naive Lindblad master equation, apart from the closed-system TCG corrections in $\mathcal{L}^{(0)}$ and $\mathcal{L}^{(2)}$, is the expression

$$\mathcal{G}^{(2)}(t, t-t_1) = \text{Tr}_B \left(\mathcal{L}^{(1)}(t) (\mathcal{T} e^{\int_{t-t_1}^t dt_2 \mathcal{L}^{(0)}(t_2)}) \mathcal{L}^{(1)}(t-t_1) \rho_B(0) \right) \quad (\text{D15})$$

which contains the closed-system TCG propagator $\mathcal{T} e^{\int_{t-t_1}^t dt_2 \mathcal{L}^{(0)}(t_2)}$ in between the two system-bath couplers $\mathcal{L}^{(1)}(t)$ and $\mathcal{L}^{(1)}(t-t_1)$. At the zero-th order in the closed-system coupling constants, $\mathcal{G}^{(2)}(t, t-t_1)$ reduces to the expression

$$\mathcal{G}^{(2)}(t, t-t_1) \longrightarrow -\text{Tr}_B \left([H_{\text{sb}}(t), [H_{\text{sb}}(t-t_1), \rho_B(0)]] \right) \quad (\text{D16})$$

which gives rise to the conventional Lindblad dissipators as well as the Lamb shift(s). With finite closed-system coupling strengths in $H(t)$, we expect the closed-system TCG propagator to provide corrections to the naive Lindblad master equation even if we make all the Markovian and secular approximations.

To illustrate this point, we discuss the resonant dissipative Rabi model defined by the following interaction-picture Hamiltonian (non-resonant models can be analyzed from the same starting point, but would be more complicated):

$$\begin{aligned} H_{\text{tot}} &= \frac{\eta}{2} (a\sigma_+ + a^\dagger\sigma_-) + \frac{\eta}{2} (a^\dagger\sigma_+ e^{2i\omega_s t} + a\sigma_- e^{-2i\omega_s t}) \\ &\quad + \int_0^\infty d\omega \cdot \mathcal{D}_\omega g_\omega \cdot \left(e^{i(\omega+\omega_s)t} \sigma_+^\omega \sigma_+ + e^{i(\omega-\omega_s)t} \sigma_+^\omega \sigma_- + h.c. \right) \end{aligned} \quad (\text{D17})$$

Working in the parameter regime where $\frac{1}{\omega_s}, \tau_B \ll \tau \ll \frac{2}{g}$, the leading-order TCG superoperators can be written as

$$\mathcal{L}^{(0)} \approx -i \left[\frac{\eta^2}{8\omega_s} (a^\dagger a + \frac{1}{2}) \sigma_z + \frac{\eta}{2} (a\sigma_+ + a^\dagger\sigma_-), \bullet \right] \equiv -i [H_{\text{eff}}, \bullet] \quad (\text{D18a})$$

$$\begin{aligned} \mathcal{L}^{(1)} &\approx -i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega \cdot e^{-\frac{1}{2}(\omega-\omega_s)^2 \tau^2} \left[e^{i(\omega-\omega_s)t} \sigma_+^\omega \sigma_- + e^{-i(\omega-\omega_s)t} \sigma_-^\omega \sigma_+, \bullet \right] \\ &\quad - i \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega \tilde{g}_\omega \cdot e^{-\frac{1}{2}(\omega-\omega_s)^2 \tau^2} \left[e^{i(\omega-\omega_s)t} \sigma_z \sigma_+^\omega a + e^{-i(\omega-\omega_s)t} \sigma_z \sigma_-^\omega a^\dagger, \bullet \right] \end{aligned} \quad (\text{D18b})$$

$$\begin{aligned} \mathcal{L}^{(2)} &\approx -i \int_0^\infty \frac{d\omega_1 d\omega_2}{(2\pi)^2} \cdot \mathcal{D}_{\omega_1} \mathcal{D}_{\omega_2} g_{\omega_1} g_{\omega_2} \left[-e^{i(\omega_2-\omega_1)t} \frac{e^{-\frac{1}{2}(\omega_2-\omega_1)^2 \tau^2} (\omega_1 + \omega_2 + 2\omega_s)}{2(\omega_1 + \omega_s)(\omega_2 + \omega_s)} \sigma_- \sigma_+ \sigma_+^{\omega_1} \sigma_+^{\omega_2} \right. \\ &\quad + e^{-i(\omega_2-\omega_1)t} \frac{(e^{-\frac{1}{2}(\omega_2-\omega_1)^2 \tau^2} - e^{-\frac{1}{2}((\omega_1-\omega_s)^2 + (\omega_2-\omega_s)^2) \tau^2}) (\omega_1 + \omega_2 - 2\omega_s)}{2(\omega_1 - \omega_s)(\omega_2 - \omega_s)} \sigma_- \sigma_+ \sigma_+^{\omega_1} \sigma_-^{\omega_2} \\ &\quad - e^{i(\omega_2-\omega_1)t} \frac{(e^{-\frac{1}{2}(\omega_2-\omega_1)^2 \tau^2} - e^{-\frac{1}{2}((\omega_1-\omega_s)^2 + (\omega_2-\omega_s)^2) \tau^2}) (\omega_1 + \omega_2 - 2\omega_s)}{2(\omega_1 - \omega_s)(\omega_2 - \omega_s)} \sigma_+ \sigma_- \sigma_-^{\omega_1} \sigma_+^{\omega_2} \\ &\quad \left. + e^{-i(\omega_2-\omega_1)t} \frac{e^{-\frac{1}{2}(\omega_2-\omega_1)^2 \tau^2} (\omega_1 + \omega_2 + 2\omega_s)}{2(\omega_1 + \omega_s)(\omega_2 + \omega_s)} \sigma_+ \sigma_- \sigma_+^{\omega_1} \sigma_-^{\omega_2}, \bullet \right] \end{aligned} \quad (\text{D18c})$$

with

$$\tilde{g}_\omega := \frac{g\omega\eta}{4} \left(\frac{1}{\omega + \omega_s} + \frac{1}{2\omega_s} + \tau^2(\omega_s - \omega) \right). \quad (\text{D19})$$

It follows from Eq.(D14) that we have the following master equation

$$\partial_t \bar{\rho}(t) = -i [H^{(2)}, \bar{\rho}(t)] + \mathcal{L}_D \bar{\rho}(t) + \mathcal{L}_H \bar{\rho}(t) \quad (\text{D20})$$

where

$$H^{(2)} \equiv H_{\text{eff}} - \int_0^\infty \frac{d\omega}{2\pi} \mathcal{D}_\omega g_\omega^2 \left(\frac{1 - e^{-(\omega - \omega_s)^2 \tau^2}}{\omega - \omega_s} - \frac{1}{\omega + \omega_s} \right) \frac{\sigma_z}{2} \quad (\text{D21})$$

while $\mathcal{L}_D$ and $\mathcal{L}_H$ are contributions from $\int_0^\infty dt_1 \mathcal{G}^{(2)}(t, t - t_1)$ which reduce to standard Lindblad dissipators and additional Lamb shift corrections to $H^{(2)}$ respectively in the $\eta \rightarrow 0$ limit. The naive master equation **D13** corresponds to this limit. When η is finite, $\mathcal{L}_D$ and $\mathcal{L}_H$ are much more complicated and have to be represented by their actions on the matrix elements of $\bar{\rho}(t)$:

$$\bar{\rho}(t) \equiv \sum_{(n_1, n_2) \in \mathbb{Z}_{\geq 0}^2} \sum_{s_1, s_2 \in \{+, -\}} \rho_{n_1, s_1; n_2, s_2} |n_1, s_1\rangle \langle n_2, s_2| \quad (\text{D22})$$

Since $\mathcal{L}^{(0)} \equiv -i[H_{\text{eff}}, \bullet]$ where the eigenstates and corresponding eigenvalues of H_{eff} can be written as

$$\begin{aligned} |n, +\rangle &:= \cos\left(\frac{\theta_n}{2}\right) |n, e\rangle + \sin\left(\frac{\theta_n}{2}\right) |n+1, g\rangle & \omega_{n,+} &:= \frac{\eta^2}{8\omega_s} \left(\sqrt{(n+1)^2 + \frac{16\omega_s^2}{\eta^2}(n+1)} - \frac{1}{2} \right) \\ |n, -\rangle &:= \sin\left(\frac{\theta_n}{2}\right) |n, e\rangle - \cos\left(\frac{\theta_n}{2}\right) |n+1, g\rangle & \omega_{n,-} &:= -\frac{\eta^2}{8\omega_s} \left(\sqrt{(n+1)^2 + \frac{16\omega_s^2}{\eta^2}(n+1)} + \frac{1}{2} \right) \end{aligned} \quad (\text{D23})$$

with $\tan \theta_n \equiv \frac{4\omega_s}{\eta\sqrt{n+1}}$ and

$$\Omega(n) \equiv \frac{\eta^2}{8\omega_s} \sqrt{(n+1)^2 + \frac{16\omega_s^2}{\eta^2}(n+1)},$$

we know from Eq.(**D15**) that the dissipator $\mathcal{L}_D$ acts on $\bar{\rho}(t)$ as

$$\begin{aligned} \langle n_1, e | \mathcal{L}_D \bar{\rho}(t) | n_2, e \rangle &\approx \frac{1}{8} \sum_{\delta \in \Delta_8} e^{-\delta^2 \tau^2} J(\omega_s + \delta) \left(n_{\omega_s + \delta} \rho_{n_1, g; n_2, g} - (1 - n_{\omega_s + \delta}) \rho_{n_1, e; n_2, e} \right) \\ \langle n_1, g | \mathcal{L}_D \bar{\rho}(t) | n_2, g \rangle &\approx -\frac{1}{8} \sum_{\delta \in \Delta_8} e^{-\delta^2 \tau^2} J(\omega_s + \delta) \left(n_{\omega_s + \delta} \rho_{n_1, g; n_2, g} - (1 - n_{\omega_s + \delta}) \rho_{n_1, e; n_2, e} \right) \\ \langle n_1, e | \mathcal{L}_D \bar{\rho}(t) | n_2, g \rangle &\approx -\frac{1}{8} \left(\sum_{\delta \in \Delta_4} n_{\omega_s + \delta} + \sum_{\delta \in \Delta_4^-} (1 - n_{\omega_s + \delta}) \right) e^{-\delta^2 \tau^2} J(\omega_s + \delta) \rho_{n_1, e; n_2, g} \end{aligned} \quad (\text{D24})$$

at leading order in $\frac{\eta}{\omega_s}$, if we define

$$\begin{aligned} \Delta_8 &\equiv \{\pm\Omega(n_1) \pm \Omega(n_2 - 1), \pm\Omega(n_1 - 1) \pm \Omega(n_2)\} \\ \Delta_4 &\equiv \{\pm\Omega(n_1) \pm \Omega(n_2)\}, \quad \Delta_4^- \equiv \{\pm\Omega(n_1 - 1) \pm \Omega(n_2 - 1)\} \end{aligned} \quad (\text{D25})$$

What we learn from the form of the above component representation of the master equation is that (1) thermal dissipation can no longer be described by state-independent Lindblad dissipators in general, and deviations from standard Lindblad dissipators are greater for states that involve fast virtual exchange of energies between the dissipating component (the spin) and the rest of the system (the cavity mode); (2) the thermal bath is no longer only probed at the transition frequency of the dissipating component as the naive Lindblad master equation would conclude, and in general bath modes at the transition frequencies among many virtual states of the quantum system can be probed during the dissipative time evolution, which effectively broadens the linewidth of the quantum system when some of the internal (virtual) transitions involve time scales not much greater than the bath relaxation time scale. It is also straightforward to obtain higher-order TCG corrections to the thermal dissipators, although they are much more complicated in form and therefore we refrain from presenting the full expression here.

Finally, we should mention that the TCG corrections investigated here for the dissipative resonant Rabi model is only one special theoretical use case for the TCG framework. In more experimentally relevant scenarios, TCG is useful for identifying the most significant observables (e.g. the particular cavity quadrature variables in the spin-cavity model) that evolve in time scales widely separated from that of one's interest (e.g. the spin dynamics) and yet modulate the time evolution of the latter. This allows one to model the system dynamics in a way that is hierarchical in time scales with clear prescription for how dissipative effects can be induced from one level to the next (e.g. dissipation in the cavity mode is responsible for the time-dependent dissipation of the spin, as discussed in the Main Text). We argue in the next subsection that this feature of TCG has important implications for system identification approaches in which one is capable of physically modeling parts, but not all, of an interacting (open) quantum system.

4. TCG-based hybrid (graybox) method for quantum system identification

In most setups of experiments on quantum devices, one does not have access to most of the system parameters, while naive assumptions and approximations (e.g. the Markovian approximation) are not valid for some system components in general. In particular, non-Markovian effective dynamics prevents explicit analytical modeling of the system components that have unknown interactions with their environments. In fact, it is a significant challenge even to identify which degrees of freedom should be included in the “quantum system” and which in the environment for those components with conspicuous information backflow[6, 7]. To address this challenge, some have adopted the “blackbox” approach using a variety of machine learning (ML) methods where input signals (control sequences) are directly mapped to the output measurement results without revealing the underlying physics that leads to the observed dynamics[8–11]. While useful for tasks such as characterizing qubit parameters and performing certain quantum controls, the blackbox approach is not sufficiently informative for the design and optimization of quantum systems, and it can be more desirable to use a hybrid or “graybox” approach where an effective Hamiltonian or master equation can still be used to model key components of the quantum system, while some or all the model parameters are supplied by the output of some blackbox ML algorithm[12–14]. This is particularly suitable for interacting quantum systems with a wide separation of time scales where one only has access to specific output signals within some frequency band while the quantum system of interest (e.g. a qubit) is affected by dynamics that take place over much shorter (e.g. readout resonators or TLS in the superconductor) or much longer (e.g. adiabatic evolution of external drives) time scales. It is in such cases where measurement-adapted TCG can be conveniently incorporated into such graybox methods.

Take the spin readout problem discussed in Section IV of the Main Text for example, the fourth-order TCG gives rise to the following effective master equation for the spin:

$$\partial_t \bar{\rho}_S \approx -i \frac{\omega_S}{2} [\sigma_z, \bar{\rho}_S] + \frac{\gamma_{\varphi_{\text{eff}}}}{2} D_{\sigma_z, \sigma_z} \bar{\rho}_S + \gamma_{\downarrow} D_{\sigma_-, \sigma_+} \bar{\rho}_S + \gamma_{\uparrow} D_{\sigma_+, \sigma_-} \bar{\rho}_S \quad (\text{D26})$$

where

$$\begin{aligned} \omega_S = & \chi + 2[\chi + \zeta(1 + |\alpha_e|^2 + |\alpha_g|^2) + \frac{\lambda^4 \kappa_c}{4}] \text{Re}(\alpha_g \alpha_e^*) - \zeta(|\alpha_g|^4 + |\alpha_e|^4) + \lambda^2 \text{Re}(\epsilon_d(\alpha_e^* + \alpha_g^*)) \\ & + 2\mu + \text{Im}(\kappa_-^d + \kappa_+^d) \text{Re}(\alpha_e - \alpha_g) \end{aligned} \quad (\text{D27})$$

and

$$\gamma_{\varphi_{\text{eff}}} = 2[\chi + \zeta(1 + |\alpha_e|^2 + |\alpha_g|^2) - \frac{\lambda^4 \kappa_c}{4}] \text{Im}(\alpha_g \alpha_e^*) + \lambda^2 \text{Im}(\epsilon_d(\alpha_e^* - \alpha_g^*)) \quad (\text{D28})$$

are parameters for the spin time evolution that depend on the dynamics of the readout cavity mode.

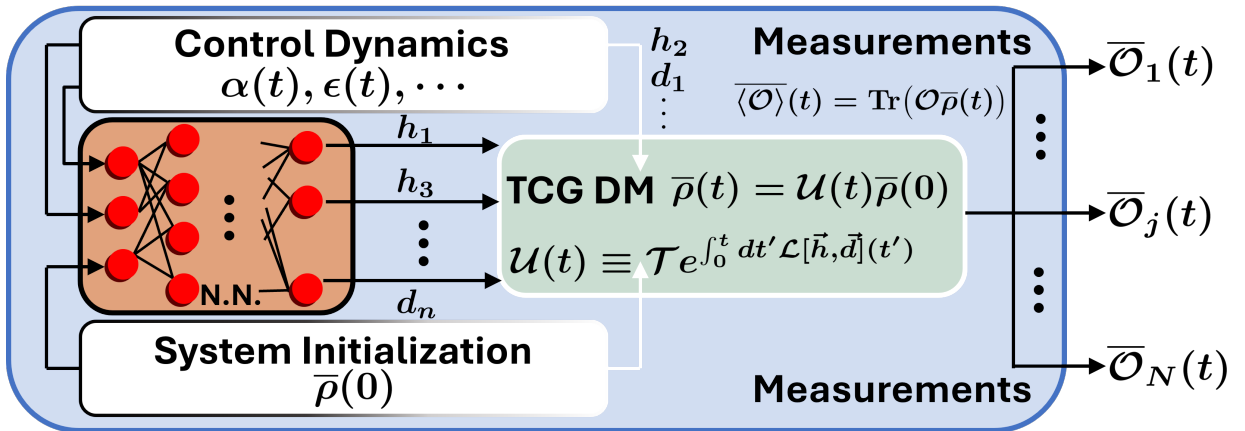


Figure 1: The schematics of a graybox architecture. In general, some parameters of the TCG master equation can be directly calculated from the predetermined or calibrated control dynamics and initial state information, which constitutes the “whitebox” part of the model. Other master equation parameters can be provided by the outputs of a neural network (N.N.) whose inputs come from the control dynamics and initial state information. These N.N. outputs account for the dynamics of coupled inaccessible degrees of freedom or uncalibrated model parameters, and thereby constitute the “blackbox” part of the model. Compared to the graybox architecture proposed in [14], the TCG master equation generalizes the Hamiltonian-based whitebox model to account for the effective non-unitary dynamics of the measured observables

In many experimentally relevant situations, one has relatively good control over the interactions of some more well-isolated parts of the quantum system (e.g. the spin), while the dynamics of other parts (e.g. the cavity mode) may be subject to

complicated mechanisms the details of which are not fully available to the experimentalists. In such situations, the effective TCG master equation offers a clear recipe for combining physical and machine learning models in a “graybox” framework – explicit physical models will be provided by the TCG effective master equation for the parts of the system whose interactions are well-understood; machine learning models, on the other hand, can be used to map the control sequences to a set of output (time-dependent) parameters that are then used to inform the TCG master equation with dynamical variables that cannot be directly measured or calibrated. See Fig. 1 for a schematic of the MaTCG-based graybox architecture.

Compared to graybox schemes that have been studied in the literature[12–14], the MaTCG method contends that, depending on the measurements to be performed on a quantum system, the most appropriate model for the “whitebox” part of the graybox structure is not a unitary Hamiltonian evolution in general, and is rather better described by a TCG master equation with parameters informed by the output of the ML blackbox. Since the graybox model has to be trained on measurement results with finite time resolution, our analysis in the Main Text has shown that it is necessary in many situations to include corrections from MaTCG beyond effective Hamiltonian methods to avoid the assumption of unphysical parameters.

5. The TCG stochastic master equation and quantum jumps

In reality, photons emitted from a quantum system are never detected at precise moments in time, even with perfect detector efficiency. This not only affects the effective master equation discussed previously corresponding to situations with no record of any information leaking out of the system through incoherent processes, but also has implications for the “quantum trajectory” picture where leaked photons are monitored during an extended period of time. One way to account for the finite time resolution of photocurrent detection is to start from the time-coarse grained master equation

$$\partial_t \bar{\rho}(t; \tau) = \mathcal{L}(t; \tau) \bar{\rho}(t; \tau) \equiv [\mathcal{J} + \mathcal{K}(t; \tau)] \bar{\rho}(t; \tau) \quad (\text{D29})$$

where the coarse-graining time scale τ reflects the finite resolution of the recorded photocurrent, and

$$\mathcal{J} \bar{\rho} \equiv \gamma c \bar{\rho} c^\dagger \quad (\text{D30})$$

is the quantum jump superoperator that can be related to photon detection events, while

$$\mathcal{K}(t; \tau) \equiv \mathcal{L}(t; \tau) - \mathcal{J} \quad (\text{D31})$$

generates smooth evolution for the unmonitored part of the system.

The master equation can be formally solved by the propagator

$$\begin{aligned} \mathcal{U}(t; \tau) &\equiv \mathcal{T} e^{\int_0^t dt_1 \mathcal{L}(t_1, \tau)} \\ &= \sum_{m=0}^{\infty} \int_0^t dt_m \int_0^{t_m} dt_{m-1} \cdots \int_0^{t_2} dt_1 \cdot \mathcal{S}(t, t_{m-1}; \tau) \mathcal{J} \mathcal{S}(t_m, t_{m-1}; \tau) \cdots \mathcal{J} \mathcal{S}(t_1, 0; \tau) \end{aligned} \quad (\text{D32})$$

with

$$\mathcal{S}(t_n, t_{n-1}; \tau) \equiv \mathcal{T} e^{\int_{t_{n-1}}^{t_n} dt' \cdot \mathcal{K}(t'; \tau)}. \quad (\text{D33})$$

To see the reason behind the formula above, notice that if we define

$$\mathcal{V}(t; \tau) \equiv \bar{\mathcal{T}} e^{-\int_0^t dt_1 \cdot \mathcal{K}(t_1; \tau)} \mathcal{U}(t; \tau) \quad (\text{D34})$$

then

$$\begin{aligned} \partial_t \mathcal{V}(t; \tau) &= - \left(\bar{\mathcal{T}} e^{-\int_0^t dt_1 \cdot \mathcal{K}(t_1; \tau)} \right) \mathcal{K}(t; \tau) \mathcal{U}(t; \tau) + \left(\bar{\mathcal{T}} e^{-\int_0^t dt_1 \cdot \mathcal{K}(t_1; \tau)} \right) (\mathcal{J} + \mathcal{K}(t; \tau)) \mathcal{U}(t; \tau) \\ &= \left(\bar{\mathcal{T}} e^{-\int_0^t dt_1 \cdot \mathcal{K}(t_1; \tau)} \right) \mathcal{J} \bar{\mathcal{T}} e^{\int_0^t dt_1 \cdot \mathcal{K}(t_1; \tau)} \equiv \mathcal{J}_I(t; \tau) \end{aligned} \quad (\text{D35})$$

which allows us to make the following Dyson expansion:

$$\begin{aligned} \mathcal{V}(t; \tau) &= \sum_{m=0}^{\infty} \int_0^t dt_m \int_0^{t_m} dt_{m-1} \cdots \int_0^{t_2} dt_1 \cdot \mathcal{J}_I(t_m; \tau) \mathcal{J}_I(t_{m-1}; \tau) \cdots \mathcal{J}_I(t_1; \tau) \\ &= \sum_{m=0}^{\infty} \int_0^t dt_m \int_0^{t_m} dt_{m-1} \cdots \int_0^{t_2} dt_1 \cdot \left(\bar{\mathcal{T}} e^{-\int_0^{t_m} dt'_m \cdot \mathcal{K}(t'_m; \tau)} \right) \mathcal{J} \mathcal{S}(t_m, t_{m-1}; \tau) \cdots \mathcal{J} \mathcal{S}(t_1, 0; \tau) \end{aligned} \quad (\text{D36})$$

if we define $\mathcal{S}(t_n, t_{n-1}; \tau) \equiv e^{\int_{t_{n-1}}^{t_n} dt' \cdot \mathcal{K}(t'; \tau)}$. Multiplying both sides by $\mathcal{S}(t, 0; \tau)$, we obtain that

$$\begin{aligned} \mathcal{U}(t; \tau) &= \mathcal{S}(t, 0; \tau) \mathcal{V}(t; \tau) \\ &= \sum_{m=0}^{\infty} \int_0^t dt_m \int_0^{t_m} dt_{m-1} \cdots \int_0^{t_2} dt_1 \cdot \mathcal{S}(t, t_{m-1}; \tau) \mathcal{J} \mathcal{S}(t_m, t_{m-1}; \tau) \cdots \mathcal{J} \mathcal{S}(t_1, 0; \tau) \end{aligned} \quad (\text{D37})$$

As demonstrated in [15], the probability density for a perfect detector with time resolution τ to record m photon detection events in the time interval $[0, t]$ is

$$p_m(t_1, \dots, t_m; [0, t], \tau) = \text{Tr} [\mathcal{S}(t, t_{m-1}; \tau) \mathcal{J} \mathcal{S}(t_m, t_{m-1}; \tau) \cdots \mathcal{J} \mathcal{S}(t_1, 0; \tau) \bar{\rho}(0; \tau)] \quad (\text{D38})$$

if the detection events center around time $t = t_1, t_2, \dots, t_m$ respectively. Consequently, if one records a particular series of photon detection events centered around those moments in time, the conditional density operator is

$$\rho_c^{t_1, \dots, t_m}(t; \tau) = \frac{\mathcal{S}(t, t_{m-1}; \tau) \mathcal{J} \mathcal{S}(t_m, t_{m-1}; \tau) \cdots \mathcal{J} \mathcal{S}(t_1, 0; \tau) \bar{\rho}(0; \tau)}{\text{Tr} [\mathcal{S}(t, t_{m-1}; \tau) \mathcal{J} \mathcal{S}(t_m, t_{m-1}; \tau) \cdots \mathcal{J} \mathcal{S}(t_1, 0; \tau) \bar{\rho}(0; \tau)]}. \quad (\text{D39})$$

In particular, following similar reasoning as that leading to the theory of Wiseman, Milburn, and Carmichael [16], for the third-order TCG master equation of the spin-cavity model we have the following stochastic master equation after adding the white noise term due to homodyne monitoring of the cavity amplitude quadrature variable $x = a + a^\dagger$ with efficiency $\eta = 1$ and time resolution τ :

$$\begin{aligned} \partial_t \bar{\rho}_c(t; \tau) &= -i \left[\epsilon_d \left(1 + \frac{\lambda^2 \sigma_z}{2} \right) (c + c^\dagger) - \chi \frac{\sigma_z}{2} \left(c^\dagger c + \frac{1}{2} \right), \bar{\rho}_c(t; \tau) \right] \\ &\quad + \kappa_c D_{(1+\lambda^2 \sigma_z/2)c, (1+\lambda^2 \sigma_z/2)c^\dagger} \bar{\rho}_c(t; \tau) + \kappa_s D_{\sigma_-, \sigma_+} \bar{\rho}_c(t; \tau) \\ &\quad + [i \kappa_d^- (D_{\sigma_-, \sigma_+ c} + D_{\sigma_+ c, \sigma_-}) \bar{\rho}_c(t; \tau) + i \kappa_d^+ (D_{\sigma_-, \sigma_+} + D_{\sigma_+, \sigma_-}) \bar{\rho}_c(t; \tau) + h.c.] \\ &\quad + \sqrt{\kappa_c} \xi(t; \tau) \left((1 + \lambda^2 \sigma_z/2) c \bar{\rho}_c(t; \tau) + \bar{\rho}_c(t; \tau) (1 + \lambda^2 \sigma_z/2) c^\dagger \right. \\ &\quad \left. - \text{Tr} [\bar{\rho}_c(t; \tau) ((1 + \lambda^2 \sigma_z/2) c + (1 + \lambda^2 \sigma_z/2) c^\dagger)] \bar{\rho}_c(t; \tau) \right) \end{aligned} \quad (\text{D40})$$

where

$$\kappa_d^\pm \equiv \frac{\epsilon_d g_{ac}^2}{(\omega_d \pm \omega_a)^2} \quad (\text{D41})$$

while $\xi(t; \tau)$ denotes a stationary colored Gaussian noise process determined by the coarse-graining window function $f(t; \tau)$:

$$\mathbb{E} [\xi(t; \tau)] = 0, \quad \text{Cov} [\xi(t; \tau), \xi(t + \Delta t; \tau)] = \int dt_1 dt_2 \cdot f'(t_1) f'(t_2) \min(t - t_1, t + \Delta t - t_2). \quad (\text{D42})$$

In particular, for a rectangular window of length τ we have

$$\text{Cov}_{\text{rec}} [\xi(t; \tau), \xi(t + \Delta t; \tau)] = \frac{\max(\tau - |\Delta t|, 0)}{\tau^2} \quad (\text{D43})$$

whereas for a gaussian window of width τ we have

$$\text{Cov}_{\text{gs}} [\xi(t; \tau), \xi(t + \Delta t; \tau)] = \frac{1}{2\sqrt{\pi}\tau} e^{-\frac{\Delta t^2}{4\tau^2}} \quad (\text{D44})$$

We see that the Wiener white noise is recovered in the $\tau \rightarrow 0$ limit as one would expect.

Since the diagonal elements of the spin density matrix decouple from the off-diagonal ones in Eq.(D40), it becomes naturally justified to use the same ansatz as in [17] and write

$$\bar{\rho}_c(t; \tau) = \sum_{n \in \{0,1\}} |n\rangle \langle n| \otimes \int \frac{dx dy}{\pi} \cdot P_n(t, x, y; \tau) |\bar{\alpha} + (x + iy)/2\rangle \langle \bar{\alpha} + (x + iy)/2| \quad (\text{D45})$$

where

$$\bar{\alpha} \approx -2\epsilon_d \kappa_c \frac{2\lambda^2 \chi \kappa_c + i(\kappa_c^2 + \chi^2)}{(\kappa_c^2 + \chi^2)^2}$$

is the mean value of the two (conditional) classical fixed points in the cavity variable a . According to this ansatz, the population of the spin state $|n\rangle$ can be calculated as

$$p_n(t; \tau) \equiv \int \frac{dx dy}{\pi} \cdot P_n(t, x, y; \tau) = \text{Tr}(\bar{\rho}(t; \tau) |n\rangle \langle n|) \quad (\text{D46})$$

It then follows from Eq.(D40) that

$$\begin{aligned} \partial_t P_0 &= 2\epsilon_d (1 - \lambda^2/2) \partial_y P_0 + \chi ((\text{Re}(\bar{\alpha}) + x/2) \partial_y - (\text{Im}(\bar{\alpha}) + y/2) \partial_x) P_0 \\ &\quad + \kappa_c (1 - \lambda^2) [\partial_x (\text{Re}(\bar{\alpha}) + x/2) + \partial_y (\text{Im}(\bar{\alpha}) + y/2)] P_0 \\ &\quad + \kappa_s P_1 - 2(\kappa_d^- + \kappa_d^+) (\text{Im}(\bar{\alpha}) + y/2) (P_1 - P_0) + 2\kappa_d^- \partial_y P_1 - (\kappa_d^- + \kappa_d^+) \partial_y P_0 \\ &\quad + \sqrt{\kappa_c} \xi(t; \tau) ((1 + \lambda^2 \sigma_z/2)x - (1 - \lambda^2/2)p_0 \langle x \rangle_0 - (1 + \lambda^2/2)p_1 \langle x \rangle_1) P_0 \\ \partial_t P_1 &= 2\epsilon_d (1 + \lambda^2/2) \partial_y P_1 - \chi ((\text{Re}(\bar{\alpha}) + x/2) \partial_y - (\text{Im}(\bar{\alpha}) + y/2) \partial_x) P_1 \\ &\quad + \kappa_c (1 + \lambda^2) [\partial_x (\text{Re}(\bar{\alpha}) + x/2) + \partial_y (\text{Im}(\bar{\alpha}) + y/2)] P_1 \\ &\quad - \kappa_s P_1 + 2(\kappa_d^- + \kappa_d^+) (\text{Im}(\bar{\alpha}) + y/2) (P_1 - P_0) + 2\kappa_d^+ \partial_y P_0 - (\kappa_d^- + \kappa_d^+) \partial_y P_1 \\ &\quad + \sqrt{\kappa_c} \xi(t; \tau) ((1 + \lambda^2 \sigma_z/2)x - (1 - \lambda^2/2)p_0 \langle x \rangle_0 - (1 + \lambda^2/2)p_1 \langle x \rangle_1) P_1 \end{aligned} \quad (\text{D47})$$

These equations of motion in terms of the Glauber–Sudarshan P representation gives rise to the following dynamical equation for $p_1(t; \tau) \equiv 1 - p_0(t; \tau)$:

$$\begin{aligned} \partial_t p_1(t; \tau) &= -\kappa_s p_1(t; \tau) + 2(\kappa_d^- + \kappa_d^+) \left(2\text{Im}(\bar{\alpha}) (p_1(t; \tau) - 1/2) - \frac{1}{2} \langle y \rangle_0 + \frac{1}{2} p_1(t; \tau) (\langle y \rangle_1 + \langle y \rangle_0) \right) \\ &\quad + \sqrt{\kappa_c} \xi(t; \tau) p_1(t; \tau) (1 - p_1(t; \tau)) \left(\langle x \rangle_1 - \langle x \rangle_0 + \frac{\lambda^2}{2} (\langle x \rangle_1 + \langle x \rangle_0) \right) \end{aligned} \quad (\text{D48})$$

where the statistical behavior of the predicted dynamics depends on the coarse-graining time scale τ not only explicitly through the noise term $\xi(t; \tau)$, but also implicitly through the effective master equation (which has negligible dependence on τ here only in the “dispersive limit” discussed previously).

Appendix E: Positivity of the TCG master equation

As noted in Subsection IV A of the Main Text, the TCG dissipators (and therefore the TCG master equation) are not positive-definite in general if $\bar{\rho}(t)$ is allowed to represent an arbitrary mixed state. For the spin-cavity model discussed in the Main Text, it is straightforward to show that we can write the drive-induced dissipator $D_{\epsilon_d g_{ac}^2}$ as

$$D_{\epsilon_d g_{ac}^2} = \sum_{k=1}^4 \kappa_k D_{X_k, X_k^\dagger} \quad (\text{E1})$$

where

$$\kappa_1 = \kappa_2 = -\kappa_3 = -\kappa_4 = \epsilon_d g_{ac}^2 \frac{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}}{(\omega_d - \omega_a)^2 (\omega_d + \omega_a)^2} \quad (\text{E2})$$

and

$$X_1 = \frac{\sigma_+}{\sqrt{2}} \left(1 + i \frac{(\omega_d + \omega_a)^2}{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}} c - i \frac{(\omega_d - \omega_a)^2}{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}} c^\dagger \right) \quad (\text{E3})$$

$$X_2 = \frac{\sigma_-}{\sqrt{2}} \left(1 + i \frac{(\omega_d - \omega_a)^2}{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}} c - i \frac{(\omega_d + \omega_a)^2}{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}} c^\dagger \right) \quad (\text{E4})$$

$$X_3 = \frac{\sigma_+}{\sqrt{2}} \left(1 - i \frac{(\omega_d + \omega_a)^2}{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}} c + i \frac{(\omega_d - \omega_a)^2}{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}} c^\dagger \right) \quad (\text{E5})$$

$$X_4 = \frac{\sigma_-}{\sqrt{2}} \left(1 - i \frac{(\omega_d - \omega_a)^2}{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}} c + i \frac{(\omega_d + \omega_a)^2}{\sqrt{(\omega_d - \omega_a)^4 + (\omega_d + \omega_a)^4}} c^\dagger \right). \quad (\text{E6})$$

These dissipators cannot appear in a master equation with infinite time-resolution, since two of the four κ 's are negative. However, they are legitimate in a TCG master equation with sufficiently large coarse-graining time scale τ since the domain of time-averaged density matrices $\bar{\rho}(t)$ are smaller than that of the exact $\rho(t)$ in the $\tau = 0$ limit.

For example, with the readout model analyzed here, one cannot have $\text{Tr}(|1\rangle\langle 1|\bar{\rho}(t)) = 0$ since $\text{Tr}(|1\rangle\langle 1|\rho(t')) \geq 0$ for any t' , and the pure ground of the spin is not stable under the interaction-picture Hamiltonian $H_I(t)$. If one forces the imaginary part of $\text{Tr}(c|l\rangle\langle l|\rho(0))$ to be positive at the initial time $t = 0$ for any spin level l , then the drive and cavity dissipation will evolve $\text{Tr}(c|l\rangle\langle l|\bar{\rho}(t))$ to have negative imaginary part before the population $\bar{n}_l^z \equiv \text{Tr}(|l\rangle\langle l|\bar{\rho})$ reaches zero.

To test one of the most extreme cases, consider starting with the pure initial state $|1\rangle \otimes |\alpha\rangle$ for some cavity coherent state $|\alpha\rangle$ with $\text{Im}(\alpha) > 0$. As a leading-order semi-classical approximation, we can approximate the almost coherently occupied cavity by a classical drive whose frequency is detuned from that of the spin by $\delta_{ac} \equiv \omega_d - \omega_a = 2\pi \cdot 2\text{GHz}$. The amplitude of the effective classical drive is then $2g_{ac}|\alpha|$, and therefore we expect the spin to undergo approximate Rabi oscillations with frequency $\sqrt{(2g_{ac}|\alpha|)^2 + \delta_{ac}^2} \gg 1\text{GHz}$, and therefore the initial coarse-grained ground state population is on the order of

$$\bar{n}_0^z(0) \approx \frac{1}{2} \cdot \frac{(2g_{ac}|\alpha|)^2}{(2g_{ac}|\alpha|)^2 + \delta_{ac}^2}. \quad (\text{E7})$$

Assuming that $|\alpha| \ll \frac{2|\omega_d - \omega_a|}{2g_{ac}}$, we focus on the cases where $\bar{n}_0^z(0) \ll 1$ and $\bar{n}_1^z(0) \approx 1$ in order to find out whether the initially negative spin decay rate K in the TCG master equation will lead to unphysical states with $\bar{n}_0^z < 0$. At the leading order, $\langle c(t) \rangle_0$ rotates in the clockwise direction at the rate $\Delta \approx \delta\omega_c^{(2)}(0)$ around its steady-state value

$$\langle c(\infty) \rangle_0 \approx \frac{\epsilon_d}{-\Delta + \frac{\kappa_c}{2}i}. \quad (\text{E8})$$

Therefore, before $\text{Im}(\langle \alpha \rangle_0)$ becomes negative, the ground state population will reduce by less than

$$\frac{\theta(\epsilon_d; \alpha) \cdot \epsilon_d}{\Delta} \cdot \frac{2g_{ac}^2}{(\omega_d - \omega_a)^2} \text{Im}(\alpha) \approx \frac{\theta(\epsilon_d; \alpha) \epsilon_d}{\Delta} \frac{\text{Im}(\alpha)}{|\alpha|^2} \bar{n}_1^z(0) \quad (\text{E9})$$

where

$$\theta(\epsilon_d; \alpha) := \arcsin \frac{\text{Im}(\alpha) - \frac{-\epsilon_d \frac{\kappa_c}{2}}{\Delta^2 + \frac{\kappa_c^2}{4}}}{\sqrt{(\text{Im}(\alpha) - \frac{-\epsilon_d \frac{\kappa_c}{2}}{\Delta^2 + \frac{\kappa_c^2}{4}})^2 + (\text{Re}(\alpha) - \frac{-\Delta}{\Delta^2 + \frac{\kappa_c^2}{4}})^2}} - \arcsin \frac{-\frac{\epsilon_d \frac{\kappa_c}{2}}{\Delta^2 + \frac{\kappa_c^2}{4}}}{\sqrt{(-\frac{\epsilon_d \frac{\kappa_c}{2}}{\Delta^2 + \frac{\kappa_c^2}{4}})^2 + (-\frac{-\Delta}{\Delta^2 + \frac{\kappa_c^2}{4}})^2}}. \quad (\text{E10})$$

It is straightforward to verify that

$$\frac{\theta(\epsilon_d; \alpha) \epsilon_d}{\Delta} \frac{\text{Im}(\alpha)}{|\alpha|^2} \leq \frac{\theta(\epsilon_d; \text{Im}(\alpha)i) \epsilon_d}{\Delta} \frac{1}{\text{Im}(\alpha)} < \lim_{\epsilon_d \rightarrow \infty} \frac{\theta(\epsilon_d; \text{Im}(\alpha)i) \epsilon_d}{\Delta} \frac{1}{\text{Im}(\alpha)} = 1, \quad (\text{E11})$$

which, according to Eq.(E9), implies that the coarse-grained ground state population will not drop below zero before the drive-induced spin transition rates become positive. In fact, we expect $D_{\epsilon_d g_{ac}^2}$ to be a well-behaving dissipator for $\bar{\rho}(t)$ as long as the conditions stated in the beginning of this subsection are satisfied, and that the initial $\bar{\rho}(0)$ is allowed by the dynamics of $\rho(t)$. For example, with $\alpha = \sqrt{2}i$, the time evolution of the coarse-grained excited state population $\bar{n}_1^z(t)$ is given in Fig.2:

Remarkably, $\bar{n}_1^z(t)$ increases until about $t = 90\text{ns}$, despite the fact that $\bar{n}_1^z(t)$ is already close to 1 while $\bar{n}_0^z(t)$ is close to 0. And similar phenomenon can also be observed for $\bar{n}_0^z(t)$ if the initial state is chosen to be $|0\rangle \otimes |\alpha\rangle$ instead. This behavior of the spin population cannot be phenomenologically explained by any positive and spin state-independent effective transition rates between the ground and excited levels. However, as shown in Fig.2, the dynamics of $\bar{n}_0^z(t)$ and $\bar{n}_1^z(t)$ can be quite accurately predicted by state-independent spin transition rates if we allow them to be negative for certain states of the cavity mode. Therefore, the possibly negative effective transition rates predicted by the TCG master equation are not due to any failure of the perturbation theory, but rather a faithful representation of physical phenomena that cannot be explained by Lindblad master equations with positive rates of quantum jumps.

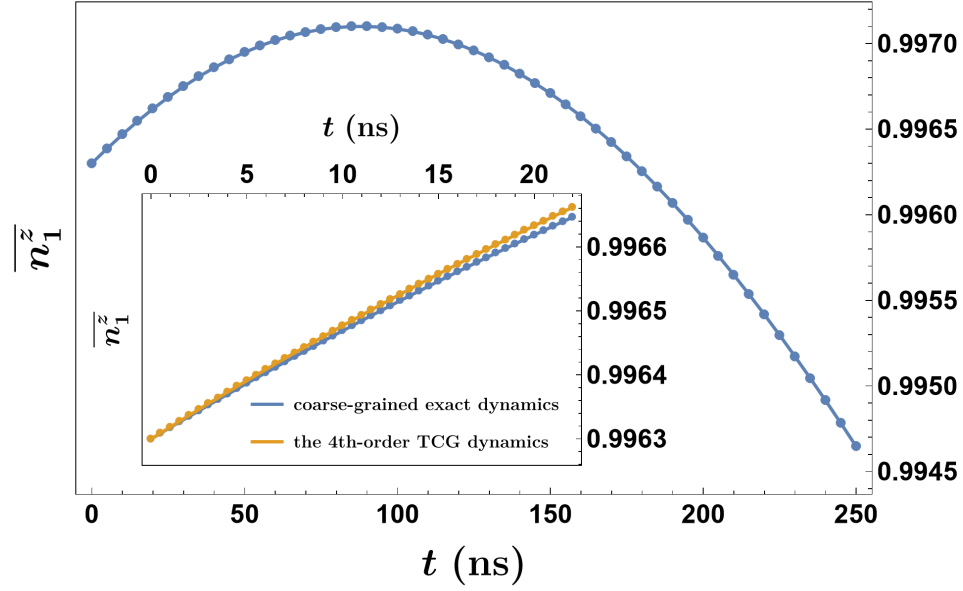


Figure 2: The simulated time evolution of the coarse-grained excited spin state population $\overline{n}_1^z(t)$. The inset compares the result obtained from the fourth-order TCG master equation with that obtained by directly coarse-graining the exact dynamics. In particular, both results confirm that the excited state population immediately increases after $t = 0$, even though $\overline{n}_1^z(t)$ is close to 1 and $\overline{n}_0^z(t)$ is close to 0. This phenomena can be explained by a (temporarily) negative decay rate from $|1\rangle$ to $|0\rangle$.

Appendix F: The effective master equation for the reduced spin density matrix

For the spin-cavity system discussed in Section IV of the Main Text, we can trace out the cavity degrees of freedom and obtain an effective master equation for the reduced spin density matrix $\overline{\rho}_S(t) = \text{Tr}_C \overline{\rho}(t)$. In particular, we are interested in the situation where the cavity is approximately in coherent states with amplitudes α_g and α_e when the spin is in the ground and excited states respectively. In [18], this is done by applying a polaron-type transformation to the rotating-frame system. In this work, we accept the same set of basic assumptions as in [18], which allows us to take a shortcut by making the following equivalent approximations:

$$\text{Tr}_C [C \langle 0 | \overline{\rho} | 0 \rangle] \approx C|_{c \rightarrow \alpha_g, c^\dagger \rightarrow \alpha_g^*} \quad \text{and} \quad \text{Tr}_C [C \langle 1 | \overline{\rho} | 1 \rangle] \approx C|_{c \rightarrow \alpha_e, c^\dagger \rightarrow \alpha_e^*} \quad (\text{F1})$$

for C an arbitrary product of c and $c^\dagger$, whereas

$$\begin{aligned} \text{Tr}_C [c \langle 0 | \overline{\rho} | 1 \rangle] &\approx \alpha_g \text{Tr}_C [\langle 0 | \overline{\rho} | 1 \rangle]; & \text{Tr}_C [c \langle 1 | \overline{\rho} | 0 \rangle] &\approx \alpha_e \text{Tr}_C [\langle 1 | \overline{\rho} | 0 \rangle]; \\ \text{Tr}_C [c^\dagger \langle 0 | \overline{\rho} | 1 \rangle] &\approx \alpha_e^* \text{Tr}_C [\langle 0 | \overline{\rho} | 1 \rangle]; & \text{Tr}_C [c^\dagger \langle 1 | \overline{\rho} | 0 \rangle] &\approx \alpha_g^* \text{Tr}_C [\langle 1 | \overline{\rho} | 0 \rangle]; \\ \text{Tr}_C [c^\dagger c \langle 0 | \overline{\rho} | 1 \rangle] &\approx \alpha_g \alpha_e^* \text{Tr}_C [\langle 0 | \overline{\rho} | 1 \rangle]; & \text{Tr}_C [c^\dagger c \langle 1 | \overline{\rho} | 0 \rangle] &\approx \alpha_e \alpha_g^* \text{Tr}_C [\langle 1 | \overline{\rho} | 0 \rangle]; \\ \text{Tr}_C [(c^\dagger c)^2 \langle 0 | \overline{\rho} | 1 \rangle] &\approx ((1 + |\alpha_e|^2 + |\alpha_g|^2) \alpha_g \alpha_e^* - \frac{1}{2} |\alpha_g|^4 - \frac{1}{2} |\alpha_e|^4) \text{Tr}_C [\langle 0 | \overline{\rho} | 1 \rangle]; \\ \text{Tr}_C [(c^\dagger c)^2 \langle 1 | \overline{\rho} | 0 \rangle] &\approx ((1 + |\alpha_e|^2 + |\alpha_g|^2) \alpha_e \alpha_g^* - \frac{1}{2} |\alpha_g|^4 - \frac{1}{2} |\alpha_e|^4) \text{Tr}_C [\langle 1 | \overline{\rho} | 0 \rangle]. \end{aligned} \quad (\text{F2})$$

With these approximations, it is straightforward to derive the following approximate expressions:

$$\begin{aligned} &-i \text{Tr}_C [(c + c^\dagger) \sigma_z, \overline{\rho}] \approx -i \text{Tr}_C \left[-2(\alpha_g + \alpha_e^*) \langle 0 | \overline{\rho} | 1 \rangle \cdot |0\rangle \langle 1| + 2(\alpha_e + \alpha_g^*) \langle 1 | \overline{\rho} | 0 \rangle \cdot |1\rangle \langle 0| \right] \\ &= -i \left[\frac{\alpha_e + \alpha_e^* + \alpha_g + \alpha_g^*}{2} \sigma_z, \overline{\rho}_S \right] + \frac{i}{2} (\alpha_e - \alpha_e^* - \alpha_g + \alpha_g^*) D_{\sigma_z, \sigma_z} \overline{\rho}_S \\ &= -i \left[\text{Re}(\alpha_e + \alpha_g) \sigma_z, \overline{\rho}_S \right] + \text{Im}(\alpha_e^* - \alpha_g^*) D_{\sigma_z, \sigma_z} \overline{\rho}_S \end{aligned} \quad (\text{F3})$$

$$\begin{aligned}
& -i\text{TrC}\left[(c^\dagger c\sigma_z, \bar{\rho}]\right] \approx -i\text{TrC}\left[-2\alpha_g\alpha_e^*\langle 0|\bar{\rho}|1\rangle \cdot |0\rangle\langle 1| + 2\alpha_e\alpha_g^*\langle 1|\bar{\rho}|0\rangle \cdot |1\rangle\langle 0|\right] \\
& = -i\left[\frac{\alpha_e\alpha_g^* + \alpha_g\alpha_e^*}{2}\sigma_z, \bar{\rho}_S\right] + \frac{i}{2}(\alpha_e\alpha_g^* - \alpha_g\alpha_e^*)D_{\sigma_z, \sigma_z}\bar{\rho}_S \\
& = -i\left[\text{Re}(\alpha_g\alpha_e^*)\sigma_z, \bar{\rho}_S\right] + \text{Im}(\alpha_g\alpha_e^*)D_{\sigma_z, \sigma_z}\bar{\rho}_S
\end{aligned} \tag{F4}$$

$$\begin{aligned}
& -i\text{TrC}\left[(c^\dagger c)^2\sigma_z, \bar{\rho}]\right] \approx -i\text{TrC}\left[-2(|\alpha_g|^2 + |\alpha_e|^2)\alpha_g\alpha_e^* - \frac{1}{2}|\alpha_g|^4 - \frac{1}{2}|\alpha_e|^4\right]\langle 0|\bar{\rho}|1\rangle \cdot |0\rangle\langle 1| \\
& \quad + 2(|\alpha_g|^2 + |\alpha_e|^2)\alpha_e\alpha_g^* - \frac{1}{2}|\alpha_g|^4 - \frac{1}{2}|\alpha_e|^4\langle 1|\bar{\rho}|0\rangle \cdot |1\rangle\langle 0| \\
& = -i\left[\left((1 + |\alpha_e|^2 + |\alpha_g|^2)\frac{\alpha_e\alpha_g^* + \alpha_g\alpha_e^*}{2} - \frac{|\alpha_e|^4 + |\alpha_g|^4}{2}\right)\sigma_z, \bar{\rho}_S\right] + \frac{i}{2}(1 + |\alpha_e|^2 + |\alpha_g|^2)(\alpha_e\alpha_g^* - \alpha_g\alpha_e^*)D_{\sigma_z, \sigma_z}\bar{\rho}_S \\
& = -i\left[\left((1 + |\alpha_e|^2 + |\alpha_g|^2)\text{Re}(\alpha_g\alpha_e^*) - \frac{|\alpha_e|^4 + |\alpha_g|^4}{2}\right)\sigma_z, \bar{\rho}_S\right] + (1 + |\alpha_e|^2 + |\alpha_g|^2)\text{Im}(\alpha_g\alpha_e^*)D_{\sigma_z, \sigma_z}\bar{\rho}_S
\end{aligned} \tag{F5}$$

$$\begin{aligned}
& \text{TrC}\left[D_{c\sigma_z, c^\dagger\sigma_z}\bar{\rho}\right] \approx -i\text{TrC}\left[-2i\alpha_g\alpha_e^*\langle 0|\bar{\rho}|1\rangle \cdot |0\rangle\langle 1| - 2i\alpha_e\alpha_g^*\langle 1|\bar{\rho}|0\rangle \cdot |1\rangle\langle 0|\right] \\
& = -i\left[\frac{-i\alpha_e\alpha_g^* + i\alpha_g\alpha_e^*}{2}\sigma_z, \bar{\rho}_S\right] + \frac{1}{2}(\alpha_e\alpha_g^* + \alpha_g\alpha_e^*)D_{\sigma_z, \sigma_z}\bar{\rho}_S \\
& = -i\left[-\text{Im}(\alpha_g\alpha_e^*)\sigma_z, \bar{\rho}_S\right] + \text{Re}(\alpha_g\alpha_e^*)D_{\sigma_z, \sigma_z}\bar{\rho}_S
\end{aligned} \tag{F6}$$

$$\begin{aligned}
& \text{TrC}\left[\kappa_-^d D_{\sigma_-, c\sigma_+}\bar{\rho} + h.c.\right] \\
& \approx -i\text{TrC}\left[i(\kappa_-^d\alpha_e + \kappa_-^{d*}\alpha_e^*)D_{\sigma_-, \sigma_+}\bar{\rho} + i\kappa_-^d\frac{\alpha_e - \alpha_g}{2}\langle 0|\bar{\rho}|1\rangle \cdot |0\rangle\langle 1| + i\kappa_-^{d*}\frac{\alpha_e^* - \alpha_g^*}{2}\langle 1|\bar{\rho}|0\rangle \cdot |1\rangle\langle 0|\right] \\
& = -i\left[\frac{-i(\kappa_-^d(\alpha_e - \alpha_g) - \kappa_-^{d*}(\alpha_e^* - \alpha_g^*))}{8}\sigma_z, \bar{\rho}_S\right] + 2\text{Re}(\kappa_-^d\alpha_e)D_{\sigma_-, \sigma_+}\bar{\rho}_S - \frac{1}{8}(\kappa_-^d(\alpha_e - \alpha_g) + \kappa_-^{d*}(\alpha_e^* - \alpha_g^*))D_{\sigma_z, \sigma_z}\bar{\rho}_S \\
& = -i\left[\frac{\text{Im}(\kappa_-^d)\text{Re}(\alpha_e - \alpha_g)}{4}\sigma_z, \bar{\rho}_S\right] - 2\text{Im}(\kappa_-^d)\text{Im}(\alpha_e)D_{\sigma_-, \sigma_+}\bar{\rho}_S + \frac{1}{4}\text{Im}(\kappa_-^d)\text{Im}(\alpha_e - \alpha_g)D_{\sigma_z, \sigma_z}\bar{\rho}_S
\end{aligned} \tag{F7}$$

$$\begin{aligned}
& \text{TrC}\left[\kappa_-^d D_{c\sigma_+, \sigma_-}\bar{\rho} + h.c.\right] \\
& \approx -i\text{TrC}\left[i(\kappa_-^d\alpha_g + \kappa_-^{d*}\alpha_g^*)D_{\sigma_+, \sigma_-}\bar{\rho} - i\kappa_-^{d*}\frac{\alpha_e^* - \alpha_g^*}{2}\langle 0|\bar{\rho}|1\rangle \cdot |0\rangle\langle 1| - i\kappa_-^d\frac{\alpha_e - \alpha_g}{2}\langle 1|\bar{\rho}|0\rangle \cdot |1\rangle\langle 0|\right] \\
& = -i\left[\frac{-i(\kappa_-^d(\alpha_e - \alpha_g) - \kappa_-^{d*}(\alpha_e^* - \alpha_g^*))}{8}\sigma_z, \bar{\rho}_S\right] + 2\text{Re}(\kappa_-^d\alpha_g)D_{\sigma_+, \sigma_-}\bar{\rho}_S + \frac{1}{8}(\kappa_-^d(\alpha_e - \alpha_g) + \kappa_-^{d*}(\alpha_e^* - \alpha_g^*))D_{\sigma_z, \sigma_z}\bar{\rho}_S \\
& = -i\left[\frac{\text{Im}(\kappa_-^d)\text{Re}((\alpha_e - \alpha_g))}{4}\sigma_z, \bar{\rho}_S\right] - 2\text{Im}(\kappa_-^d)\text{Im}(\alpha_g)D_{\sigma_+, \sigma_-}\bar{\rho}_S - \frac{1}{4}\text{Im}(\kappa_-^d)\text{Im}((\alpha_e - \alpha_g))D_{\sigma_z, \sigma_z}\bar{\rho}_S
\end{aligned} \tag{F8}$$

$$\begin{aligned}
& \text{TrC}\left[\tilde{\gamma}D_{\sigma_-, c^\dagger c, \sigma_+}\bar{\rho} + h.c.\right] \\
& \approx -i\text{TrC}\left[i\tilde{\gamma}|\alpha_e|^2 D_{\sigma_-, \sigma_+}\bar{\rho} + i\tilde{\gamma}\frac{|\alpha_e|^2 - \alpha_g\alpha_e^*}{2}\langle 0|\bar{\rho}|1\rangle \cdot |0\rangle\langle 1| + i\tilde{\gamma}\frac{|\alpha_e|^2 - \alpha_e\alpha_g^*}{2}\langle 1|\bar{\rho}|0\rangle \cdot |1\rangle\langle 0|\right] + h.c. = 0
\end{aligned} \tag{F9}$$

$$\begin{aligned}
& \text{TrC}\left[\tilde{\gamma}D_{\sigma_+, c^\dagger c, \sigma_-}\bar{\rho} + h.c.\right] \\
& \approx -i\text{TrC}\left[i\tilde{\gamma}|\alpha_g|^2 D_{\sigma_+, \sigma_-}\bar{\rho} + i\tilde{\gamma}\frac{|\alpha_g|^2 - \alpha_e\alpha_g^*}{2}\langle 1|\bar{\rho}|0\rangle \cdot |1\rangle\langle 0| + i\tilde{\gamma}\frac{|\alpha_g|^2 - \alpha_g\alpha_e^*}{2}\langle 0|\bar{\rho}|1\rangle \cdot |0\rangle\langle 1|\right] + h.c. = 0
\end{aligned} \tag{F10}$$

$$\text{TrC}\left[\tilde{\kappa}_\pm D_{\sigma_-, (c^\dagger c)^2, \sigma_+}\bar{\rho} + h.c.\right] \approx 0; \quad \text{TrC}\left[\tilde{\kappa}_\pm D_{\sigma_+, (c^\dagger c)^2, \sigma_-}\bar{\rho} + h.c.\right] \approx 0 \tag{F11}$$

$$\begin{aligned}
& \text{TrC}\left[D_{c\sigma_z, c^\dagger}\bar{\rho} + h.c.\right] \approx -i\text{TrC}\left[-i\alpha_g\alpha_e^*\langle 0|\bar{\rho}|1\rangle \cdot |0\rangle\langle 1| + i\alpha_e\alpha_g^*\langle 1|\bar{\rho}|0\rangle \cdot |1\rangle\langle 0|\right] + h.c. \\
& = -i\left[\frac{i\text{Re}(\alpha_g\alpha_e^*)}{2}\sigma_z, \bar{\rho}_S\right] + \frac{i}{2}\text{Im}(\alpha_g\alpha_e^*)D_{\sigma_z, \sigma_z}\bar{\rho}_S + h.c. = 0
\end{aligned} \tag{F12}$$

$$\begin{aligned} \text{Tr}_C [D_{c^\dagger \sigma_z, c} \bar{\rho} + h.c.] &\approx -i \text{Tr}_C [-i \alpha_e \alpha_g^* \langle 1 | \bar{\rho} | 0 \rangle \cdot |1\rangle \langle 0| + i \alpha_g \alpha_e^* \langle 0 | \bar{\rho} | 1 \rangle \cdot |0\rangle \langle 1|] + h.c. \\ &= -i \left[\frac{i \text{Re}(\alpha_e \alpha_g^*)}{2} \sigma_z, \bar{\rho}_S \right] + \frac{i}{2} \text{Im}(\alpha_e \alpha_g^*) D_{\sigma_z, \sigma_z} \bar{\rho}_S + h.c. = 0 \end{aligned} \quad (\text{F13})$$

At the fourth order, the TCG master equation can be written as

$$\begin{aligned} \partial_t \bar{\rho} = \mathcal{L} \bar{\rho} &= -i \left[\chi (c^\dagger c + \frac{1}{2}) \sigma_z + \epsilon_d (1 + \frac{\lambda^2}{2} \sigma_z) (c + c^\dagger) + (\zeta' - \xi) c^\dagger c + \mu \sigma_z + \zeta (c^\dagger c)^2 \sigma_z, \bar{\rho} \right] \\ &+ \kappa_c D_{c(1+\lambda^2 \sigma_z/2), c^\dagger(1+\lambda^2 \sigma_z/2)} \bar{\rho} + \kappa_s D_{\sigma_-, \sigma_+} \bar{\rho} \\ &+ \left[\kappa_-^d (D_{\sigma_-, \sigma_+ c} + D_{\sigma_+ c, \sigma_-}) \bar{\rho} + \kappa_+^d (D_{\sigma_-, c, \sigma_+} + D_{\sigma_+, \sigma_-, c}) \bar{\rho} + \tilde{\kappa}_+ (D_{\sigma_- c^\dagger c^2, \sigma_+ c^\dagger} - D_{\sigma_+ c^\dagger c^2, \sigma_- c}) \bar{\rho} \right. \\ &\left. + \tilde{\kappa}_- (D_{\sigma_- c^\dagger c^2, \sigma_+ c} - D_{\sigma_+ c^\dagger c^2, \sigma_- c^\dagger}) \bar{\rho} + \tilde{\gamma} (D_{\sigma_- c^\dagger c, \sigma_+} + D_{\sigma_+ c^\dagger c, \sigma_-}) \bar{\rho} + \tilde{\gamma}_z (D_{\sigma_z c, c^\dagger} - D_{\sigma_z c^\dagger, c}) \bar{\rho} + h.c. \right] \end{aligned} \quad (\text{F14})$$

where

$$\chi = -g_{ac}^2 \left(\frac{1}{\omega_d - \omega_a} - \frac{1}{\omega_d + \omega_a} \right) + \zeta \quad \text{and} \quad \kappa_\pm^d = i \frac{\epsilon_d g_{ac}^2}{(\omega_d \pm \omega_a)^2} \quad (\text{F15})$$

while the other parameters are defined in Section IV of the Main Text. It then follows that we can trace out the cavity degrees of freedom and obtain

$$\partial_t \bar{\rho}_S = \text{Tr}_C [\mathcal{L} \bar{\rho}] \approx -i \frac{\omega_S}{2} [\sigma_z, \bar{\rho}_S] + \frac{\gamma_{\varphi_{\text{eff}}}}{2} D_{\sigma_z, \sigma_z} \bar{\rho}_S + \gamma_\downarrow D_{\sigma_-, \sigma_+} \bar{\rho}_S + \gamma_\uparrow D_{\sigma_+, \sigma_-} \bar{\rho}_S \quad (\text{F16})$$

where

$$\begin{aligned} \omega_S = \chi + 2\mu + 2 \left[\chi + \zeta (1 + |\alpha_e|^2 + |\alpha_g|^2) + \frac{\lambda^4 \kappa_c}{4} \right] \text{Re}(\alpha_g \alpha_e^*) - \zeta (|\alpha_g|^4 + |\alpha_e|^4) \\ + \lambda^2 \text{Re}(\epsilon_d (\alpha_e^* + \alpha_g^*)) + \text{Im}(\kappa_-^d + \kappa_+^d) \text{Re}(\alpha_e - \alpha_g) \end{aligned} \quad (\text{F17})$$

$$\gamma_{\varphi_{\text{eff}}} = 2 \left[\chi + (1 + |\alpha_e|^2 + |\alpha_g|^2) - \frac{\lambda^4 \kappa_c}{4} \right] \text{Im}(\alpha_g \alpha_e^*) + \lambda^2 \text{Im}(\epsilon_d (\alpha_e^* - \alpha_g^*)) \quad (\text{F18})$$

$$\gamma_\downarrow = \kappa_s - 2 \text{Im}(\kappa_-^d + \kappa_+^d) \text{Im}(\alpha_e) = \kappa_s + K(t) \quad (\text{F19})$$

$$\gamma_\uparrow = -2 \text{Im}(\kappa_-^d + \kappa_+^d) \text{Im}(\alpha_g) = \Gamma(t). \quad (\text{F20})$$

Approximation:

$$\begin{aligned} \partial_t \bar{\rho} = \mathcal{L} \bar{\rho} &= -i \left[\chi (c^\dagger c + \frac{1}{2}) \sigma_z + \epsilon_d (1 + \frac{\lambda^2}{2} \sigma_z) (c + c^\dagger), \bar{\rho} \right] \\ &+ \kappa_c D_{c(1+\lambda^2 \sigma_z/2), c^\dagger(1+\lambda^2 \sigma_z/2)} \bar{\rho} + \kappa_s D_{\sigma_-, \sigma_+} \bar{\rho} \\ &+ \left[\kappa_-^d (D_{\sigma_-, \sigma_+ c} + D_{\sigma_+ c, \sigma_-}) \bar{\rho} + \kappa_+^d (D_{\sigma_-, c, \sigma_+} + D_{\sigma_+, \sigma_-, c}) \bar{\rho} + h.c. \right] \end{aligned} \quad (\text{F21})$$

Appendix G: Representation of the spin-cavity state by conditional Husimi functions

To solve the fourth-order TCG master equation (F14) for the spin-cavity system, one may adopt the following Wigner representation of the time-coarse grained density matrix $\bar{\rho}$:

$$\bar{W}_\mu^{ab}(\phi, n) := \frac{1}{\pi} \int_{-\infty}^{\infty} d\phi' \cdot \langle a; \phi - \phi' |_\mu \bar{\rho} | b; \phi + \phi' \rangle_\mu e^{2in\phi'} \quad (\text{G1})$$

for $\mu \in x, y, z$ and $a, b \in 0, 1$, where the state $|a; \phi\rangle_\mu$ is the simultaneous eigenstate of σ_μ and $\Phi \equiv \frac{c+\phi}{\sqrt{2}}$ with eigenvalues $2a - 1$ and ϕ respectively (when the subscript μ is omitted, it is assumed to be z which refers to the energy basis). The TCG effective master equation for $\bar{\rho}(t)$ can then be translated into a PDE for the Wigner function $\bar{W}_z^{ab}(\phi, n)$ by replacing operator products with Moyal products of the corresponding functions of the phase space variables ϕ and n for different spin states.

The Wigner function can also be related to physical observables in a heterodyne measurement of the cavity quadrature variables. In fact, the joint probability density of obtaining the heterodyne measurement result (ϕ, n) when the spin is in state $|a\rangle_\mu$ can be written as the following conditional Husimi functions [5, 19, 20]:

$$\bar{Q}_\mu^{aa}(\phi, n) = \int d\phi' dn' \cdot \frac{e^{-(\phi'^2 + n'^2)}}{\pi} \bar{W}_\mu^{aa}(\phi + \phi', n + n'). \quad (\text{G2})$$

More generally, we can define

$$\bar{Q}_\mu^{ab}(\phi, n) := \int d\phi' dn' \cdot \frac{e^{-(\phi'^2 + n'^2)}}{\pi} \bar{W}_\mu^{ab}(\phi + \phi', n + n') \equiv \frac{1}{2\pi} \langle a; \alpha = \frac{\phi + ni}{\sqrt{2}} |_\mu \bar{\rho} | \alpha = \frac{\phi + ni}{\sqrt{2}}; b \rangle_\mu \quad (\text{G3})$$

where $|a; \alpha = \zeta\rangle_\mu$ is the simultaneous eigenstate of σ_μ and c with eigenvalues $2a - 1$ and ζ respectively, and the PDE for the conditional Wigner functions $\bar{W}_z^{ab}(\phi, n)$ can be straightforwardly translated into a set of equations of motion for $\bar{Q}_\mu^{ab}(\phi, n)$ if we make the following replacements:

$$\begin{aligned} \partial_\phi^k \partial_n^j \bar{W}_z^{ab} &\longrightarrow \partial_\phi^k \partial_n^j \bar{Q}_z^{ab} \\ \phi \partial_\phi^k \partial_n^j \bar{W}_z^{ab} &\longrightarrow \phi \partial_\phi^k \partial_n^j \bar{Q}_z^{ab} + \frac{1}{2} \partial_\phi^{k+1} \partial_n^j \bar{Q}_z^{ab} \\ n \partial_\phi^k \partial_n^j \bar{W}_z^{ab} &\longrightarrow n \partial_\phi^k \partial_n^j \bar{Q}_z^{ab} + \frac{1}{2} \partial_\phi^k \partial_n^{j+1} \bar{Q}_z^{ab} \\ \phi^2 \partial_\phi^k \partial_n^j \bar{W}_z^{ab} &\longrightarrow (\phi^2 + \frac{1}{2}) \partial_\phi^k \partial_n^j \bar{Q}_z^{ab} + \phi \partial_\phi^{k+1} \partial_n^j \bar{Q}_z^{ab} + \frac{1}{4} \partial_\phi^{k+2} \partial_n^j \bar{Q}_z^{ab} \\ n^2 \partial_\phi^k \partial_n^j \bar{W}_z^{ab} &\longrightarrow (n^2 + \frac{1}{2}) \partial_\phi^k \partial_n^j \bar{Q}_z^{ab} + n \partial_\phi^k \partial_n^{j+1} \bar{Q}_z^{ab} + \frac{1}{4} \partial_\phi^k \partial_n^{j+2} \bar{Q}_z^{ab} \\ &\dots \end{aligned}$$

which can be derived from the definition of $\bar{Q}_z^{ab}$ in Eq.(G2). In terms of the Husimi Q functions in the z -basis of the spin, the third-order TCG master equation at zero temperature can be written as

$$\begin{aligned} \partial_t Q_z^{00}(\phi, n) &= \left(\kappa_c + \frac{\sqrt{2} g_{ac}^2 \epsilon_d}{(\omega_d - \omega_a)^2} n \right) Q_z^{00}(\phi, n) + g_{ac}^2 \left(\kappa_c \frac{\omega_a}{1 + (\frac{\omega_a}{\Lambda})^2} \frac{4\omega_d}{(\omega_d^2 - \omega_a^2)^2} - \frac{\sqrt{2} \epsilon_d}{(\omega_d - \omega_a)^2} n \right) Q_z^{11}(\phi, n) \\ &+ \left(\frac{\kappa_c}{2} \phi - \frac{2g_{ac}^2 \omega_a}{\omega_d^2 - \omega_a^2} n \right) \partial_\phi Q_z^{00}(\phi, n) + \left(\sqrt{2} \epsilon_d - \frac{\sqrt{2} g_{ac}^2 \epsilon_d}{\omega_d^2 - \omega_a^2} + \frac{2g_{ac}^2 \omega_a}{\omega_d^2 - \omega_a^2} \phi + \frac{\kappa_c}{2} n \right) \partial_n Q_z^{00}(\phi, n) \\ &+ \frac{\kappa_c}{2} \partial_\phi^2 Q_z^{00}(\phi, n) + \frac{\kappa_c}{2} \partial_n^2 Q_z^{00}(\phi, n) \end{aligned} \quad (\text{G4})$$

$$\begin{aligned} \partial_t Q_z^{11}(\phi, n) &= - \frac{\sqrt{2} g_{ac}^2 \epsilon_d}{(\omega_d - \omega_a)^2} n Q_z^{00}(\phi, n) + \left(\kappa_c \left[1 - \frac{\omega_a}{1 + (\frac{\omega_a}{\Lambda})^2} \frac{4g_{ac}^2 \omega_d}{(\omega_d^2 - \omega_a^2)^2} \right] + \frac{\sqrt{2} g_{ac}^2 \epsilon_d}{(\omega_d - \omega_a)^2} n \right) Q_z^{11}(\phi, n) \\ &+ \left(\sqrt{2} \epsilon_d + \frac{\sqrt{2} \epsilon_d g_{ac}^2 (2\omega_a \omega_d + \omega_d^2 - \omega_a^2)}{\omega_d (\omega_d + \omega_a) (\omega_d - \omega_a)^2} - \frac{2g_{ac}^2 \omega_a}{\omega_d^2 - \omega_a^2} \phi + \frac{\kappa_c}{2} n \right) \partial_n Q_z^{11}(\phi, n) \\ &- \frac{\sqrt{2} g_{ac}^2 \epsilon_d}{(\omega_d - \omega_a)^2} \partial_n Q_z^{00}(\phi, n) + \left(\frac{\kappa_c}{2} \phi + \frac{2g_{ac}^2 \omega_a}{\omega_d^2 - \omega_a^2} n \right) \partial_\phi Q_z^{11}(\phi, n) \\ &+ \frac{\kappa_c}{2} \partial_\phi^2 Q_z^{11}(\phi, n) + \frac{\kappa_c}{2} \partial_n^2 Q_z^{11}(\phi, n) \end{aligned} \quad (\text{G5})$$

$$\begin{aligned}
\partial_t \text{Re} Q_z^{01}(\phi, n) = & g_{ac}^2 \left(\frac{2\omega_a}{\omega_d^2 - \omega_a^2} (2 + \partial_n^2) - \frac{\sqrt{2}\epsilon_d(2\omega_d^2 + \omega_a\omega_d - \omega_a^2)}{\omega_d(\omega_d + \omega_a)(\omega_d - \omega_a)^2} \partial_\phi + \frac{2\omega_a}{\omega_d^2 - \omega_a^2} \partial_\phi^2 \right) \text{Im} Q_z^{01}(\phi, n) \\
& + \left(\kappa_c + \frac{\sqrt{2}g_{ac}^2\epsilon_d}{(\omega_d - \omega_a)^2} \partial_n \right) \text{Re} Q_z^{01}(\phi, n) + \frac{2g_{ac}^2\omega_a}{\omega_d^2 - \omega_a^2} n \partial_n \text{Im} Q_z^{01}(\phi, n) \\
& + \left(\sqrt{2}\epsilon_d + \frac{g_{ac}^2\epsilon_d(3\omega_d - \omega_a)\omega_a}{\sqrt{2}\omega_d(\omega_d + \omega_a)(\omega_d - \omega_a)^2} + \frac{\kappa_c}{2} n \right) \partial_n \text{Re} Q_z^{01}(\phi, n) \\
& - \left(\frac{g_{ac}^2\epsilon_d(2\omega_d - \omega_a)}{\sqrt{2}\omega_d(\omega_d - \omega_a)^2} - \frac{2g_{ac}^2\omega_a}{\omega_d^2 - \omega_a^2} \phi \right) \partial_\phi \text{Im} Q_z^{01}(\phi, n) \\
& + \frac{\kappa_c}{2} \phi \partial_\phi \text{Re} Q_z^{01}(\phi, n) + \frac{\kappa_c}{2} \partial_\phi^2 \text{Re} Q_z^{01}(\phi, n) + \frac{\kappa_c}{2} \partial_n^2 \text{Re} Q_z^{01}(\phi, n)
\end{aligned} \tag{G6}$$

$$\begin{aligned}
\partial_t \text{Im} Q_z^{01}(\phi, n) = & -g_{ac}^2 \left(\frac{2\omega_a}{\omega_d^2 - \omega_a^2} \phi^2 + \frac{2\omega_a}{\omega_d^2 - \omega_a^2} (2 + n^2) - \frac{\sqrt{2}\epsilon_d(2\omega_d - \omega_a)}{\omega_d(\omega_d - \omega_a)^2} \phi \right) \text{Re} Q_z^{01}(\phi, n) \\
& + \left(\kappa_c + \frac{\sqrt{2}g_{ac}^2\epsilon_d}{(\omega_d - \omega_a)^2} n \right) \text{Im} Q_z^{01}(\phi, n) - \frac{2g_{ac}^2\omega_a}{\omega_d^2 - \omega_a^2} n \partial_n \text{Re} Q_z^{01}(\phi, n) \\
& + \left(\sqrt{2}\epsilon_d + \frac{g_{ac}^2\epsilon_d\omega_a(3\omega_d - \omega_a)}{\sqrt{2}\omega_d(\omega_d - \omega_a)^2(\omega_d + \omega_a)} + \frac{\kappa_c}{2} n \right) \partial_n \text{Im} Q_z^{01}(\phi, n) \\
& + \left(\frac{g_{ac}^2\epsilon_d(2\omega_d - \omega_a)}{\sqrt{2}(\omega_d - \omega_a)^2\omega_d} - \frac{2g_{ac}^2\omega_a}{\omega_d^2 - \omega_a^2} \phi \right) \partial_\phi \text{Re} Q_z^{01}(\phi, n) \\
& + \frac{\kappa_c}{2} \phi \partial_\phi \text{Im} Q_z^{01}(\phi, n) + \frac{\kappa_c}{2} \partial_\phi^2 \text{Im} Q_z^{01}(\phi, n) + \frac{\kappa_c}{2} \partial_n^2 \text{Im} Q_z^{01}(\phi, n)
\end{aligned} \tag{G7}$$

The equations of motion for $\bar{Q}_z^{ab}$ can then be solved numerically to obtain the time evolution of the joint probability densities $\bar{Q}_z^{00}(\phi, n)$ and $\bar{Q}_z^{11}(\phi, n)$ for z -component spin values 0 and 1 respectively, whereas $\bar{Q}_x^{aa}(\phi, n)$ and $\bar{Q}_y^{aa}(\phi, n)$ are related to $\bar{Q}_z^{ab}(\phi, n)$ by the relations

$$\begin{aligned}
\bar{Q}_x^{00} &= \frac{\bar{Q}_z^{00} + \bar{Q}_z^{11}}{2} - \text{Re} \bar{Q}_z^{01}; \quad \bar{Q}_x^{11} = \frac{\bar{Q}_z^{00} + \bar{Q}_z^{11}}{2} + \text{Re} \bar{Q}_z^{01} \\
\bar{Q}_y^{00} &= \frac{\bar{Q}_z^{00} - \bar{Q}_z^{11}}{2} - \text{Im} \bar{Q}_z^{01}; \quad \bar{Q}_y^{11} = \frac{\bar{Q}_z^{00} - \bar{Q}_z^{11}}{2} + \text{Im} \bar{Q}_z^{01}.
\end{aligned} \tag{G8}$$

The conditional Husimi functions $\bar{Q}_\mu^{aa}$ with $a \in \{0, 1\}$ and $\mu \in \{x, y, z\}$ can be interpreted as the probability distribution of single-shot heterodyne measurement results with a particular spin state a [5, 19]. And for the transient spin-cavity dynamics, solving the differential equations for $\bar{Q}_\mu^{aa}$ provide the most detailed information about the physically observable system dynamics. Our dynamical simulation of the spin-cavity model in Subsection IV C is based on numerical solutions of those conditional Husimi functions.

Appendix H: The transmon readout model and the corresponding TCG master equation up to the fourth order

Following [21, 22], we model the system of a transmon coupled to a driven dissipative linear resonator by the following Schrödinger picture Hamiltonian:

$$\hat{H} = \hat{H}_s + \hat{H}_d(t) + \hat{H}_{sb} + \hat{H}_b \tag{H1}$$

where

$$\hat{H}_s = \omega_c c^\dagger c - g_{ac}(c - c^\dagger)(a - a^\dagger) + \omega_a a^\dagger a - \frac{\epsilon\omega_a}{48}(a + a^\dagger)^4 \tag{H2}$$

is the system Hamiltonian describing the transmon, the linear resonator, as well as their mutual coupling;

$$\hat{H}_d(t) = -(c - c^\dagger)(\epsilon_d e^{-i\omega_d t} - \epsilon_d^* e^{i\omega_d t}) \tag{H3}$$

is the drive Hamiltonian representing the readout drive on the linear resonator;

$$\hat{H}_b = \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega \omega B_\omega^\dagger B_\omega \quad (\text{H4})$$

is the bath Hamiltonian modeling the bosonic bath modes supported by a transmission line through which we drive and monitor the system; finally,

$$\hat{H}_{sb} = -(c - c^\dagger) \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega g_\omega (B_\omega - B_\omega^\dagger) \quad (\text{H5})$$

is the system-bath coupling Hamiltonian modeling the coupling between the resonator and the bath modes. Instead of the coupling strengths g_ω , a more experimentally relevant characterization of the resonator-bath coupling is the spectral density $J(\omega) := \mathcal{D}_\omega g_\omega^2$ which incorporates in it the density of states $\mathcal{D}_\omega$.

We assume for the rest of this paper that the transmon-resonator coupling is in the dispersive limit, i.e., the TCG perturbative expansion has a small parameter $\frac{g_{ac}}{|\omega_c - \omega'_a(n_a)|} \ll 1$. As motivated in the Appendix(A), we suppose that the physically measured signal is the output signal from the system, down-converted by some local oscillator (LO) in a heterodyne detector at frequency ω_{LO} , and then time-averaged over some window with finite time resolution. Therefore, we first need to go to the LO-frequency rotating frame by transforming to the interaction picture with the free Hamiltonian being

$$\hat{H}_0 := \omega_{LO} c^\dagger c + \omega_a a^\dagger a - \frac{\epsilon \omega_a}{48} (a + a^\dagger)^4 + \int_0^\infty \frac{d\omega}{2\pi} \cdot \mathcal{D}_\omega \omega B_\omega^\dagger B_\omega. \quad (\text{H6})$$

For the sake of simplicity, we further assume that the local oscillator has the same frequency as the readout drive, so that $\omega_{LO} = \omega_d$, and we have the following interaction-picture Hamiltonian to linear order in ϵ :

$$H_I(t) = H_{AC}(t) + H_{A^3C}(t) + H_D(t) + H_\delta + H_{CB}(t) \quad (\text{H7})$$

with

$$\begin{aligned} H_{AC}(t) &= g_{ac} e^{i(\omega'_a(n_a) - \omega_d)t} \left(1 - \frac{\epsilon n_a}{8}\right) c a^\dagger - g_{ac} c a \left(1 - \frac{\epsilon n_a}{8}\right) e^{-i(\omega'_a(n_a) + \omega_d)t} + h.c.; \\ H_{A^3C}(t) &= -\frac{\epsilon g_{ac}}{16} e^{i(3\omega'_a(n_a-1) - \omega_d)t} c a^{\dagger 3} + \frac{\epsilon g_{ac}}{16} c a^3 e^{-i(3\omega'_a(n_a-1) + \omega_d)t} + h.c.; \\ H_D(t) &= (\epsilon_d^* c - \epsilon_d e^{-2i\omega_d t} c) + h.c.; \quad H_\delta = -(\omega_d - \omega_c) c^\dagger c; \\ H_{CB}(t) &= \int \frac{d\omega}{2\pi} \mathcal{D}_\omega g_\omega (e^{i(\omega - \omega_d)t} c B_\omega^\dagger - e^{-i(\omega + \omega_d)t} c B_\omega) + h.c. \end{aligned} \quad (\text{H8})$$

where the operator-valued function

$$\omega'_a(n_a) := \omega_a \left(1 - \frac{\epsilon}{4} n_a\right) \quad (\text{H9})$$

gives us the phase frequency of the transmon quadrature variable a depending on the energy level n_a , with the dependence being a consequence of transmon anharmonicity. In addition to variation in the transmon transition energies, a nonzero anharmonicity ϵ also gives rise to the nonlinear atom-resonator couplings in H_{AC} and H_{A^3C} . Furthermore, when we obtain a superoperator whose coefficient depends on n_a , we adopt the convention that the operator-valued coefficient acts on the diagonal elements of $\bar{\rho}$ before any action of the dissipator, i.e.,

$$\kappa(n_a) D_{A,B} \bar{\rho} \equiv A \kappa(n_a) \bar{\rho} B - \frac{B A \kappa(n_a) \bar{\rho} + \kappa(n_a) \bar{\rho} B A}{2} \quad (\text{H10})$$

with the coefficient $\kappa(n_a)$ being any function of n_a .

1. Ranges of parameters and first-order TCG

At the lowest order (the first order), TCG simply replaces the Hamiltonian $H_I(t)$ with its time-average. To explicitly calculate the TCG superoperators, we assume that the coarse graining time scale τ is much greater than $\frac{1}{\omega_c}$, $\frac{1}{\omega'_a(n_a)}$, and $\frac{1}{|\omega_c - \omega'_a(n_a)|}$

(which are typically below the nanosecond range) while allowing τ to be comparable or smaller than $\frac{1}{|\omega_d - \omega_c|}$ (since the near-resonance situation is most relevant to experimental implementations). As stated in Eq.(101) of the Main Text, we make the following assumptions of the model parameters:

$$\frac{1}{\omega_c}, \frac{1}{\omega'_a(n_a)}, \frac{1}{|\omega_c - \omega'_a(n_a)|} \ll \tau \lesssim \frac{1}{|\omega_d - \omega_c|}, \frac{1}{J(\omega_c)};$$

$$\frac{g}{|\omega_c - \omega'_a(n_a)|} \ll 1; \quad \epsilon < 1; \quad \omega_d \sim \omega_c.$$

With these conditions, we calculate the TCG master equation up to the fourth order and ignore superoperators that are exponentially suppressed by factors smaller than or equal to $e^{-\frac{\omega_c^2 \tau^2}{2}}$, $e^{-\frac{\omega'^2_a(n_a) \tau^2}{2}}$, or $e^{-\frac{(\omega_c - \omega'_a(n_a))^2 \tau^2}{2}}$.

At the first order, TCG simply functions as a band-pass filter around the local oscillator frequency $\omega_{LO} = \omega_d$, replacing the interaction-picture Hamiltonian $H(t)$ with its slowly varying time average $\bar{H}(t)$, which gives us

$$H_{\text{TCG}}^{(1)}(t) := \bar{H}_I(t) = \left[\epsilon_d^* c + \int \frac{d\omega}{2\pi} \mathcal{D}_\omega g_\omega e^{-\frac{(\omega - \omega_d)^2 \tau^2}{2}} e^{i(\omega - \omega_d)t} c B_\omega^\dagger + h.c. \right] - (\omega_d - \omega_c) c^\dagger c \quad (\text{H11})$$

Thus we see that, at the lowest order, time-coarse grained dynamics consists of Hamiltonian-generated coherent transitions with transition frequencies that are not much greater $\frac{1}{\tau}$ in the rotating frame of the local oscillator used to probe the system. In particular, the high-frequency transverse coupling $-g_{ac}(c - c^\dagger)(a - a^\dagger)$ between the transmon and the resonator is “filtered out” from the first-order effective Hamiltonian $H_{\text{TCG}}^{(1)}$, whereas the slow dynamics induced by this coupling (e.g., dispersive shift, Purcell decay, and drive-induced transitions) show up in higher-order effective Hamiltonian terms. A very convenient feature which gets preserved to high orders in TCG is that the density matrix elements diagonal in the transmon Hilbert space will be decoupled from the off-diagonal elements, which makes it much easier, both conceptually and numerically, to keep track of the time evolution of the pointer states.

As we will show in Subsection H2, second-order TCG is sufficient for revealing the following phenomena in the slow dynamics, including:

- shifts in the transmon and resonator transition frequencies due to the capacitive coupling between them;
- effective dispersive coupling between the transmon and the resonator;
- effective direct coupling between the transmon and the bath modes, which is responsible for the Purcell decay of the transmon.

In particular, the emergent transmon-bath coupling gives rise to a Purcell decay rate of the transmon on the order of $g_{ac}^2 J(\omega_a) |\omega_a - \omega_c|^{-2}$, which is on the same order in the coupling constants as the fourth-order TCG superoperators. Therefore, to account for the system dynamics on the time scale of transmon Purcell decay, we perform perturbative time-coarse graining to the fourth order. As will be shown in Subsection H3, the third-order TCG gives rise to drive-induced inter-level transitions of the transmon, with transition rates proportional to ϵ_d^2 once the resonator has stabilized to its steady state. Finally, we discuss the induced resonator anharmonicity from the fourth-order TCG in Subsection H4.

In the following sections, we calculate the TCG master equation order by order, and emphasize on the emergent dynamics revealed at each order. We also obtain numerical solutions of the TCG master equation using the following set of parameters:

$$\epsilon = 0.2 \quad \kappa_c = 0.48 \text{MHz} \quad \frac{\omega_c}{2\pi} = 7 \text{GHz}$$

$$\omega_{01} \equiv \frac{\omega_a(1 - \frac{\epsilon}{4})}{2\pi} = 5 \text{GHz} \quad g_{ac} = 0.3016 \text{GHz}$$

where ω_{01} is the (approximate) transition energy between the ground state and the first excited state of the transmon.

2. Second-order TCG, dispersive transmon-resonator coupling, and emergent transmon-bath decay channel

At the second order in perturbative TCG, we obtain the following corrections to the system effective Hamiltonian:

$$H_{\text{TCG}}^{(2)} = \delta^{(2)} h_0(n_a) + \delta^{(2)} h_1(n_a) c^\dagger c + H_{AB} + H_{A^3B} \quad (\text{H12})$$

with the coefficients

$$\delta^{(2)} h_0(n_a) = g_{ac}^2 \frac{(1 - \frac{\epsilon(n_a+1)}{4}) \omega'_a(n_a) - (1 + 2n_a - \frac{\epsilon(2n_a^2+2n_a+1)}{4}) \omega_d}{(\omega_d + \omega'_a(n_a+1))(\omega_d - \omega'_a(n_a))} \quad (\text{H13})$$

and

$$\delta^{(2)} h_1(n_a) = 2g_{ac}^2 \frac{\left(1 - \frac{\epsilon(1+2n_a)}{2} + \frac{\epsilon^2(1+3n_a+3n_a^2)}{16}\right) \omega_d^2}{(\omega_d^2 - \omega'_a(n_a)^2)(\omega_d^2 - \omega'_a(n_a+1)^2)} \omega_a - 2g_{ac}^2 \frac{\omega'_a(n_a)^2 \omega'_a(n_a+1)^2}{(\omega_d^2 - \omega'_a(n_a)^2)(\omega_d^2 - \omega'_a(n_a+1)^2) \omega_a} \quad (\text{H14})$$

giving us the leading-order energy shifts due to the transmon-resonator coupling, whereas H_{AB} and H_{A^3B} are effective transmon-bath couplings that we would discuss later in this subsection. Here the symbol $\delta^{(i)}$ indicates the i -th order contribution to the corresponding coefficient. In particular, the dispersive shift of the cavity frequency is found to be

$$\chi_{ac}^{(2)} \equiv \delta^{(2)} h_1(0) - \delta^{(2)} h_1(1) = \frac{\epsilon g_{ac}^2 (\omega'_a(1) + \omega'_a(2)) \omega_d^2}{(\omega_d^2 - \omega'_a(1)^2)(\omega_d^2 - \omega'_a(2)^2)} \quad (\text{H15})$$

at the second order in coupling constants.

On the other hand, the effective transmon-bath couplings terms

$$H_{AB} = \int \frac{d\omega \mathcal{D}_\omega}{2\pi} \tilde{g}_\omega[a] a B^\dagger(\omega) e^{i(\omega - \omega'_a(n_a))t} + h.c. \quad (\text{H16})$$

and

$$H_{A^3B} = \int \frac{d\omega \mathcal{D}_\omega}{2\pi} \tilde{g}_\omega[a^3] a^3 B^\dagger(\omega) e^{i(\omega - 3\omega'_a(n_a-1))t} + h.c. \quad (\text{H17})$$

represent single- and three-photon creation/annihilation processes which probe bath modes near the frequencies $\omega'_a(n_a)$ and $3\omega'_a(n_a-1)$ respectively, where the operator-valued effective transmon-bath coupling strengths $\tilde{g}_\omega[a]$ and $\tilde{g}_\omega[a^3]$ are given by

$$\begin{aligned} \tilde{g}_\omega[a] &= -g_\omega g_{ac} \left(1 - \frac{\epsilon n_a}{8}\right) \omega_d \left[\frac{e^{-\frac{(\omega - \omega'_a(n_a))^2}{2} \tau^2}}{\omega_d^2 - \omega'_a(n_a)^2} + \frac{e^{-\frac{(\omega - \omega'_a(n_a))^2}{2} \tau^2} - e^{-\frac{(\omega - \omega_d)^2 + (\omega_d - \omega'_a(n_a))^2}{2} \tau^2}}{\omega_d^2 - \omega^2} \right] \\ &\approx -g_\omega g_{ac} \left(1 - \frac{\epsilon n_a}{8}\right) \frac{2\omega_d e^{-\frac{(\omega - \omega'_a(n_a))^2}{2} \tau^2}}{\omega_d^2 - \omega'_a(n_a)^2} \end{aligned} \quad (\text{H18})$$

and

$$\begin{aligned} \tilde{g}_\omega[a^3] &= -\frac{\epsilon g_\omega g_{ac} \omega_d}{16} \left[\frac{e^{-\frac{(\omega - 3\omega'_a(n_a-1))^2}{2} \tau^2}}{9\omega'_a(n_a-1)^2 - \omega_d^2} + \frac{e^{-\frac{(\omega - 3\omega'_a(n_a-1))^2}{2} \tau^2} - e^{-\frac{(\omega - \omega_d)^2 + (\omega_d - 3\omega'_a(n_a-1))^2}{2} \tau^2}}{\omega^2 - \omega_d^2} \right] \\ &\approx -\frac{\epsilon g_\omega g_{ac}}{8} \frac{\omega_d e^{-\frac{(\omega - 3\omega'_a(n_a-1))^2}{2} \tau^2}}{9\omega'_a(n_a-1)^2 - \omega_d^2}. \end{aligned} \quad (\text{H19})$$

We see that $\tilde{g}_\omega[a]$ and $\tilde{g}_\omega[a^3]$ are approximately gaussian functions centered around the one-photon and three-photon transition frequencies respectively. Tracing out the bath degrees of freedom under the standard Born-Markov approximation at zero temperature, we obtain the Purcell decay superoperators $\kappa_a(n_a) D_{a,a^\dagger}$ and $\kappa_{a^3}(n_a) D_{a^3,a^{\dagger 3}}$ with the decay rates

$$\begin{aligned} \kappa_a(n_a) &= J(\omega'_a(n_a)) \left[\frac{2g_{ac} \left(1 - \frac{\epsilon n_a}{8}\right) \omega_d}{\omega_d^2 - \omega'_a(n_a)^2} \right]^2 \\ \kappa_{a^3}(n_a) &= \frac{\epsilon^2 J(3\omega'_a(n_a-1))}{64} \left[\frac{g_{ac} \omega_d}{9\omega'_a(n_a-1)^2 - \omega_d^2} \right]^2 \end{aligned} \quad (\text{H20})$$

where $J(\omega) := \mathcal{D}_\omega g_\omega^2$ is the power spectral density of the bosonic bath. Notice that the rates of the two Purcell decay processes are independent of the state of the cavity, so the second-order TCG does not explain the drive dependence of the lifetime of the transmon's first-excited state, but rather gives rise to the drive-independent part of it. As would be discussed in Subsection H3, the drive-induced transmon level transitions kick in at the third-order TCG, where they become the most prominent processes over the transmon T_1 time scale for moderately strong drive strength ($\langle n_c \rangle > \sim 1$). On the other hand, the effective transmon-bath couplings would also give rise to Hamiltonian corrections proportional to $c^\dagger c$, $n_a c^\dagger c$, $c^{\dagger 2} c^2$, and $n_a c^{\dagger 2} c^2$, in the same way that the resonator-bath couplings give rise to Lamb shift in the resonator frequency. However, for realistic parameters, the coefficients of those Lamb-like corrections are orders of magnitudes smaller than $1/T_1$ (which is the smallest energy scale we are interested in), and will therefore be ignored in subsequent calculations. Finally, as for the resonator-bath couplings, we obtain from them

the same Lamb shift and resonator dissipator whether we apply TCG before or after the Born-Markov bath tracing procedure. In summary, for our purposes in this model, the bath gives rise to the Lamb shift term

$$\Delta_{\text{Lamb}} c^\dagger c := - \int_0^\infty \frac{d\omega}{2\pi} J(\omega) \left(\frac{1}{\omega - \omega_c} + \frac{1}{\omega + \omega_c} \right) c^\dagger c \quad (\text{H21})$$

in the Hamiltonian, as well as the drive-independent dissipators

$$\kappa_c D_{c,c^\dagger} + \kappa_a(n_a) D_{a,a^\dagger} \quad (\text{H22})$$

with the resonator decay rate $\kappa_c \equiv J(\omega_d)$. Here we have dropped the three-photon dissipator $\kappa_{a^3}(n_a) D_{a^3,a^{\dagger 3}}$ since it is of higher order in ϵ than the transmon potential, and with a drive-independent coefficient that is too small compared with $\kappa_a(n_a)$.

In order to explicitly calculate the Lamb shift, we assume an Ohmic power spectral density with a soft frequency cutoff at $\Lambda \gg \omega_a, \omega_d$:

$$J(\omega) = \frac{\alpha\omega}{1 + \frac{\omega^2}{\Lambda^2}}. \quad (\text{H23})$$

The particular functional form of $J(\omega)$ we use here is inspired by the asymptotic behavior of the power spectral density studied in [23] where the bath is modeled by the modes of a transmission-line resonator coupled to semi-infinite waveguides at both ends. And for numerical simulations presented in subsections H3 and H4, we use the value $\Lambda = 2\pi \cdot 100\text{GHz}$.

It should also be emphasized that in using the effective transmon-bath coupling to calculate the Purcell decay rate, we have implicitly assumed that bath relaxation takes place over a longer time scale than that of the (virtual) transitions between the transmon and the resonator. This condition is reasonably well satisfied by the dispersive couplings we consider here. However, the treatment of couplings to the bath in TCG has to be analyzed on a case-by-case basis in general, depending on the relative scale of the coarse-graining time τ in comparison with the bath relaxation time as well as the time scales of relevant coherent processes.

3. Third-order TCG and drive-induced incoherent transitions

Assuming the conditions in Eq.(101) of the Main Text, we find that the third-order TCG gives rise to two types of corrections to the EME: transmon-level dependent modulation of the drive terms in the Hamiltonian, and drive-induced dissipators acting on the transmon. In fact, we can write the action of the third-order Liouvillian superoperator as

$$\mathcal{L}_3(t)\bar{\rho} = -i[H_{\text{TCG}}^{(3)}, \bar{\rho}] + D_{\text{TCG}}^{(3)}\bar{\rho} \quad (\text{H24})$$

where the third-order TCG Hamiltonian $H_{\text{TCG}}^{(3)}(t)$ and dissipator $D_{\text{TCG}}^{(3)}(t)$ can be written as

$$H_{\text{TCG}}^{(3)} = \delta^{(3)} h_0(n_a) + \delta^{(3)} h_1(n_a) c^\dagger c + \left(\delta^{(3)} h_0^{(-1)}(n_a) c + h.c. \right) \quad (\text{H25})$$

and

$$D_{\text{TCG}}^{(3)} = \delta^{(3)} \kappa_{0,0}^{(-1,0)}(n_a) D_{ac,a^\dagger} + \delta^{(3)} \kappa_{0,0}^{(0,1)}(n_a) D_{a,a^\dagger c} + \delta^{(3)} \gamma_{0,0}^{(-1,0)}(n_a) D_{a^\dagger c,a} + \delta^{(3)} \gamma_{0,0}^{(0,1)}(n_a) D_{a^\dagger,ac} + h.c. \quad (\text{H26})$$

respectively. The third-order corrections in $H_{\text{TCG}}^{(3)}$ all provide small quantitative modifications to the Hamiltonian terms already existing at lower orders in the perturbation, and does not introduce any qualitatively new features to the dynamics. We therefore do not show explicit formulas for $\delta^{(3)} h_0(n_a)$, $\delta^{(3)} h_1(n_a)$, and $\delta^{(3)} h_0^{(-1)}(n_a)$. The dissipators in $D_{\text{TCG}}^{(3)}$, on the other hand, are emergent non-Hamiltonian generators of the time evolution of $\bar{\rho}(t)$. More explicitly, we have

$$\begin{aligned} \delta^{(3)} \kappa_{0,0}^{(-1,0)}(n_a) &= ig_{ac}^2 \epsilon_d^* \frac{1 - \frac{\epsilon n_a}{4}}{(\omega'_a(n_a) + \omega_d)^2} \\ \delta^{(3)} \kappa_{0,0}^{(0,1)}(n_a) &= ig_{ac}^2 \epsilon_d^* \frac{1 - \frac{\epsilon n_a}{4}}{(\omega'_a(n_a) - \omega_d)^2} \\ \delta^{(3)} \gamma_{0,0}^{(-1,0)}(n_a) &= ig_{ac}^2 \epsilon_d^* \frac{1 - \frac{\epsilon(n_a+1)}{4}}{(\omega'_a(n_a+1) - \omega_d)^2} \\ \delta^{(3)} \gamma_{0,0}^{(0,1)}(n_a) &= ig_{ac}^2 \epsilon_d^* \frac{1 - \frac{\epsilon(n_a+1)}{4}}{(\omega'_a(n_a+1) + \omega_d)^2}. \end{aligned} \quad (\text{H27})$$

For a wide range of realistic parameters, the dissipators in $D_{\text{TCG}}^{(3)}$ significantly modify the dynamical behavior of the population of the n -th transmon energy level, here defined as $p_n(t) \equiv \text{Tr}(|n\rangle_a \langle n|_a \bar{\rho}(t))$. In fact, tracing out the resonator degrees of freedom, we find the following drive-induced transition rate from level n to level $(n-1)$:

$$K(n) = 2\text{Re} \left[\left(\delta^{(3)} \kappa_{0,0}^{(-1,0)}(n) + \delta^{(3)} \kappa_{0,0}^{(0,1)}(n) \right) \cdot \langle c \rangle_n \right] \quad (\text{H28})$$

with $\langle c \rangle_n \equiv \text{Tr}(c|n\rangle_a \langle n|_a \bar{\rho}) / \text{Tr}(|n\rangle_a \langle n|_a \bar{\rho})$ being the conditional expectation value of the resonator quadrature variable c when the transmon is at energy level n . Similarly, the drive-induced transition rate from level n to level $(n+1)$ is found to be

$$\Gamma(n) = 2\text{Re} \left[\left(\delta^{(3)} \gamma_{0,0}^{(-1,0)}(n) + \delta^{(3)} \gamma_{0,0}^{(0,1)}(n) \right) \cdot \langle c \rangle_n \right]. \quad (\text{H29})$$

In practical applications of dispersive readout, the lifetime of the resonator is much shorter than that of the transmon, and the quasi-steady state of the resonator will be close to a coherent state with

$$\langle c \rangle_n \approx \frac{\epsilon_d}{-(\tilde{\omega}_c(n) - \omega_d) + \frac{\kappa_c}{2}i} \quad (\text{H30})$$

during all but the very initial period of the transmon time evolution, where $\tilde{\omega}_c(n) \equiv \omega_c + h_1(n)$ is the modified resonator frequency when the transmon is at level n . Therefore, we find the drive-induced relaxation and excitation rates to be approximately

$$\begin{aligned} K(n) &\approx |\epsilon_d|^2 g_{ac}^2 \frac{\kappa_c \left(1 - \frac{\epsilon_n}{4}\right)}{(\tilde{\omega}_c(n) - \omega_d)^2 + \frac{\kappa_c^2}{4}} \cdot \left(\frac{1}{(\omega'_a(n) - \omega_d)^2} + \frac{1}{(\omega'_a(n) + \omega_d)^2} \right) \\ \Gamma(n) &\approx |\epsilon_d|^2 g_{ac}^2 \frac{\kappa_c \left(1 - \frac{\epsilon_{(n+1)}}{4}\right)}{(\tilde{\omega}_c(n) - \omega_d)^2 + \frac{\kappa_c^2}{4}} \cdot \left(\frac{1}{(\omega'_a(n+1) - \omega_d)^2} + \frac{1}{(\omega'_a(n+1) + \omega_d)^2} \right). \end{aligned} \quad (\text{H31})$$

Notice in particular that $K(n)$ (for $n \geq 1$) and $\Gamma(n)$ are always positive regardless of the phase of ϵ_d and the sign of $\omega'_a(n+1) - \omega_d$. This is necessary for the stability of the master equation since negative relaxation/excitation rates would make the greatest p_n blow up exponentially while pushing p_{n-1} and p_{n+1} down below zero. In fact, the positivity of $K(n)$ and $\Gamma(n)$ for arbitrary choices of ϵ_d and $\omega'_a(n+1) - \omega_d$ demonstrates that perturbative TCG is consistent with the assumption that the steady state of the readout resonator is close to a coherent state depending on the state of the transmon, which is the basis for using the resonator states as pointer states in readout experiments.

Also, from Eq.(H31) we know that $K(n)$ and $\Gamma(n)$ are on the same order of magnitude. Therefore, we can compare the drive-induced relaxation rate with the Purcell decay rate to estimate their impact on the transmon dynamics:

$$\frac{K(n)}{\kappa_a(n)} = \frac{|\epsilon_d|^2 \left(1 - \frac{\epsilon_n}{4}\right)}{(\tilde{\omega}_c(n) - \omega_d)^2 + \frac{\kappa_c^2}{4}} \frac{\omega_d^2 + \omega'_a(n)^2}{2\omega_d \omega'_a(n) \left(1 - \frac{\epsilon_n}{8}\right)^2} \approx n_c^{(n)} \frac{\omega_d^2 + \omega'_a(n)^2}{2\omega_d \omega'_a(n)} \quad (\text{H32})$$

where $n_c^{(n)} \approx \frac{|\epsilon_d|^2}{(\tilde{\omega}_c(n) - \omega_d)^2 + \frac{\kappa_c^2}{4}}$ is the conditional expectation value of $n_c \equiv c^\dagger c$ when the transmon is at level n . Since ω_d and $\omega'_a(n)$ are both in the GHz range, the ratios between the drive-induced transition rates and the Purcell decay rate are on the same order of magnitude as $\langle n_c \rangle_n$. If Purcell decay is the dominate decay channel of the transmon, then we expect the readout drive to significantly decrease the T_1 lifetime of the transmon qubit initially prepared in its first excited state as soon as the drive is strong enough to maintain a few photons in the readout resonator. This is consistent with observations from recent experimental studies. In fact, our analysis here show that the ratio between drive-induced transition rates and the Purcell decay rate cannot be easily reduced by tuning the transmon and resonator frequencies or adjusting the transmon-resonator coupling strengths. To minimize the relative strength of drive-induced transitions, one has to fundamentally change the type of atom-resonator couplings (employing longitudinal couplings for example) or engineer the functional form of the spectral density (which works at the price of increasing the resonator relaxation time).

As far as the time-coarse grained dynamics of the transmon is concerned, carrying out the perturbation to the third order is sufficient for capturing all the salient features that one typically observes in a readout experiment. Using the cumulant expansion method introduced in the Appendix(I), we can solve for numerical solutions of the 3rd-order TCG master equation for different drive strengths measured in the steady-state resonator photon number $n_c^{(0)} := \langle c^\dagger c \rangle_{n_a=0}$ when the transmon is in the ground state. As shown in panel (a) of Fig. 14 of the Main Text, the T_1 lifetime decreases by more than a factor of two for relatively small resonator photon occupancy ($n_c^{(0)} \sim 1$) if the intrinsic dissipation of the transmon is negligible in comparison with the Purcell decay rate. In addition, the steady-state population $p_n(\infty)$ is nonzero for any transmon level n due to the drive-induced transitions, as shown in panel (b) of Fig. 14 of the Main Text. Moreover, for sufficiently large drive strength, the steady-state

population of a higher transmon level will exceed that of a lower level, making it impossible to assign a positive effective temperature to the transmon in its steady-state. And even for smaller drive strengths, an effective-temperature description of the steady state is only possible if we combine all the excited states of the transmon into a single level for statistical purposes.

Finally, we may also obtain detailed information about the resonator dynamics from the TCG master equation (see the upper panels in Fig. 16 of the Main Text, but we leave the discussion for Subsection H4.

4. Fourth-order TCG and numerical solutions of the effective master equation (EME)

Similar to the third-order TCG superoperators, the superoperators obtained from the fourth-order TCG can be grouped into Hamiltonian commutators and dissipators:

$$\mathcal{L}_4(t)\bar{\rho} = -i[H_{\text{TCG}}^{(4)}, \bar{\rho}] + D_{\text{TCG}}^{(4)}\bar{\rho}. \quad (\text{H33})$$

Ignoring superoperators with coefficients below the 1Hz level, we can write $H_{\text{TCG}}^{(4)}$ and $D_{\text{TCG}}^{(4)}$ as

$$\begin{aligned} H_{\text{TCG}}^{(4)} = & \delta^{(4)}h_0(n_a) + \delta^{(4)}h_1(n_a)c^\dagger c + \delta^{(4)}h_2(n_a)c^{\dagger 2}c^2 \\ & + (\delta^{(4)}h_0^{(-1)}(n_a)c + \delta^{(4)}h_1^{(-1)}(n_a)c^\dagger c^2 \\ & + \delta^{(4)}h_0^{(-2)}(n_a)c^2 + h.c.) \end{aligned} \quad (\text{H34})$$

and

$$\begin{aligned} D_{\text{TCG}}^{(4)} = & \delta^{(4)}d_{0,0}^{(-2,-1)}(n_a)D_{c^2,c^\dagger} + \delta^{(4)}d_{0,0}^{(1,2)}(n_a)D_{c^\dagger,c^2} \\ & + \delta^{(4)}\kappa_{1,0}^{(-1,-1)}(n_a)D_{ac^\dagger c^2, a^\dagger c^\dagger} + \delta^{(4)}\kappa_{1,0}^{(1,1)}(n_a)D_{ac^\dagger c^2, a^\dagger c} \\ & + \delta^{(4)}\kappa_{0,1}^{(0,0)}(n_a)D_{a, a^\dagger c^\dagger c} + \delta^{(4)}\kappa_{0,0}^{(-1,0)}(n_a)D_{ac, a^\dagger} \\ & + \delta^{(4)}\kappa_{0,1}^{(-1,0)}(n_a)D_{ac, a^\dagger c^\dagger c} + \delta^{(4)}\kappa_{0,0}^{(0,1)}(n_a)D_{a, a^\dagger c} \\ & + \delta^{(4)}\kappa_{1,0}^{(0,1)}(n_a)D_{ac^\dagger c, a^\dagger c} + \delta^{(4)}\gamma_{1,0}^{(-1,-1)}(n_a)D_{a^\dagger c^\dagger c^2, ac^\dagger} \\ & + \delta^{(4)}\gamma_{1,0}^{(1,1)}(n_a)D_{a^\dagger c^\dagger c^2, ac} + \delta^{(4)}\gamma_{0,1}^{(0,0)}(n_a)D_{a^\dagger, ac^\dagger c} \\ & + \delta^{(4)}\gamma_{0,0}^{(-1,0)}(n_a)D_{a^\dagger c, a} + \delta^{(4)}\gamma_{0,1}^{(-1,0)}(n_a)D_{a^\dagger c, ac^\dagger c} \\ & + \delta^{(4)}\gamma_{0,0}^{(0,1)}(n_a)D_{a^\dagger, ac} + \delta^{(4)}\gamma_{1,0}^{(0,1)}(n_a)D_{a^\dagger c^\dagger c, ac} + h.c. \end{aligned} \quad (\text{H35})$$

respectively, where we see that the fourth-order TCG provides some modifications to the coefficients of existing superoperators, while at the same time introducing a few new ones. Noticeably, starting at this order in perturbation theory, we begin to see that the transmon anharmonicity induces the anharmonic self-Kerr potential $h_2(n_a)c^{\dagger 2}c^2$ in the resonator²; also, we begin to see transmon-assisted bi-photon absorption and emission processes emerging from the drive terms in the Hamiltonian³. Similarly, more complicated dissipators are also introduced at this order, but they only generate small and more subtle dependence of the drive-induced transition rates on the resonator state.

Appendix I: Equations of motion for the conditional cumulants

In general, the TCG master equation can be written as

$$\partial_t \bar{\rho}_{ac}(t) = -i[H_{\text{eff}}, \bar{\rho}_{ac}(t)] + D_{\text{eff}}\bar{\rho}_{ac}(t). \quad (\text{I1})$$

As discussed in subsections H1-H4, up to the fourth order in coupling constants and with the local oscillator set at the drive frequency ($\omega_{\text{LO}} = \omega_d$), the effective Hamiltonian can be written as

$$H_{\text{eff}} = h_0(n_a) + h_1(n_a)n_c + h_2(n_a)n_c^2 + \left[h_0^{(-1)}(n_a)c + h_1^{(-1)}(n_a)c^\dagger c^2 + h_0^{(-2)}(n_a)c^2 + h.c. \right] \quad (\text{I2})$$

² The resonator anharmonicity is induced by the transmon anharmonicity in the sense that it vanishes in the $\epsilon \rightarrow 0$ limit.

³ Such processes are transmon-assisted in the sense that the effective bi-photon drive strengths are proportional to $g_{ac}^2|\epsilon_d|^2$.

while the effective jump superoperators can be written as

$$D_{\text{eff}} = D_{\text{eff}}^{(0)} + D_{\text{eff}}^{(-)} + D_{\text{eff}}^{(+)} \quad (13)$$

where $D_{\text{eff}}^{(0)}$, $D_{\text{eff}}^{(-)}$, and $D_{\text{eff}}^{(+)}$ are defined as follows:

$$D_{\text{eff}}^{(0)} = d_{0,0}^{(-1,-1)}(n_a)D_{c,c^\dagger} + \left[d_{0,0}^{(-2,-1)}(n_a)D_{c^2,c^\dagger} + d_{0,0}^{(1,2)}(n_a)D_{c^\dagger,c^2} + h.c. \right] \quad (14)$$

$$\begin{aligned} D_{\text{eff}}^{(-)} = & \kappa_{0,0}^{(0,0)}(n_a)D_{a,a^\dagger} + \left[\kappa_{1,0}^{(-1,-1)}(n_a)D_{ac^\dagger c^2, a^\dagger c^\dagger} + \kappa_{1,0}^{(1,1)}(n_a)D_{ac^\dagger c^2, a^\dagger c} + \kappa_{0,1}^{(0,0)}(n_a)D_{a, a^\dagger c^\dagger c} + h.c. \right] \\ & + \left[\kappa_{0,0}^{(-1,0)}(n_a)D_{ac, a^\dagger} + \kappa_{0,1}^{(-1,0)}(n_a)D_{ac, a^\dagger c^\dagger c} + \kappa_{0,0}^{(0,1)}(n_a)D_{a, a^\dagger c} + \kappa_{1,0}^{(0,1)}(n_a)D_{ac^\dagger c, a^\dagger c} + h.c. \right] \end{aligned} \quad (15)$$

$$\begin{aligned} D_{\text{eff}}^{(+)}(t) = & \gamma_{1,0}^{(-1,-1)}(n_a)D_{a^\dagger c^\dagger c^2, ac^\dagger} + \gamma_{1,0}^{(1,1)}(n_a)D_{a^\dagger c^\dagger c^2, ac} + \gamma_{0,1}^{(0,0)}(n_a)D_{a^\dagger, ac^\dagger c} + \gamma_{0,0}^{(-1,0)}(n_a)D_{a^\dagger c, a} \\ & + \gamma_{0,1}^{(-1,0)}(n_a)D_{a^\dagger c, ac^\dagger c} + \gamma_{0,0}^{(0,1)}(n_a)D_{a^\dagger, ac} + \gamma_{1,0}^{(0,1)}(n_a)D_{a^\dagger c^\dagger c, ac} + h.c. \end{aligned} \quad (16)$$

Here the coefficients may receive contributions from different orders in the TCG perturbative expansion:

$$\begin{aligned} h_0 &= \delta^{(2)}h_0 + \delta^{(3)}h_0 + \delta^{(4)}h_0 + \dots; & h_1 &= -(\omega_d - \omega_c) + \delta^{(2)}h_1 + \delta^{(3)}h_1 + \delta^{(4)}h_1 + \dots; \\ h_2 &= \delta^{(4)}h_2 + \dots; & h_0^{(-1)} &= \epsilon_d^* + \delta^{(3)}h_0^{(-1)} + \delta^{(4)}h_0^{(-1)} + \dots; \\ h_1^{(-1)} &= \delta^{(4)}h_1^{(-1)} + \dots; & h_0^{(-2)} &= \delta^{(4)}h_0^{(-2)} + \dots; \\ d_{0,0}^{(-1,-1)} &= \kappa_c + \dots; & d_{0,0}^{(-2,-1)} &= \delta^{(4)}d_{0,0}^{(-2,-1)} + \dots; & d_{0,0}^{(1,2)} &= \delta^{(4)}d_{0,0}^{(1,2)} + \dots; \\ \kappa_{0,0}^{(0,0)} &= \kappa_a + \dots; & \kappa_{1,0}^{(-1,-1)} &= \delta^{(4)}\kappa_{1,0}^{(-1,-1)} + \dots; & \kappa_{1,0}^{(1,1)} &= \delta^{(4)}\kappa_{1,0}^{(1,1)} + \dots; \\ \kappa_{0,1}^{(0,0)} &= \delta^{(4)}\kappa_{0,1}^{(0,0)} + \dots; & \kappa_{0,1}^{(-1,0)} &= \delta^{(4)}\kappa_{0,1}^{(-1,0)} + \dots; & \kappa_{1,0}^{(0,1)} &= \delta^{(4)}\kappa_{1,0}^{(0,1)} + \dots; \\ \kappa_{0,0}^{(0,1)} &= \delta^{(3)}\kappa_{0,0}^{(0,1)} + \delta^{(4)}\kappa_{0,0}^{(0,1)} + \dots; & \kappa_{0,0}^{(-1,0)} &= \delta^{(3)}\kappa_{0,0}^{(-1,0)} + \delta^{(4)}\kappa_{0,0}^{(-1,0)} + \dots; \\ \gamma_{1,0}^{(0,1)} &= \delta^{(4)}\gamma_{1,0}^{(0,1)} + \dots; & \gamma_{1,0}^{(1,1)} &= \delta^{(4)}\gamma_{1,0}^{(1,1)} + \dots; & \gamma_{0,1}^{(0,0)} &= \delta^{(4)}\gamma_{0,1}^{(0,0)} + \dots; \\ \gamma_{0,0}^{(-1,0)} &= \delta^{(3)}\gamma_{0,0}^{(-1,0)} + \delta^{(4)}\gamma_{0,0}^{(-1,0)} + \dots; & \gamma_{0,1}^{(-1,0)} &= \delta^{(4)}\gamma_{0,1}^{(-1,0)} + \dots; \\ \gamma_{0,0}^{(0,1)} &= \delta^{(3)}\gamma_{0,0}^{(0,1)} + \delta^{(4)}\gamma_{0,0}^{(0,1)} + \dots; & \gamma_{1,0}^{(-1,-1)} &= \delta^{(4)}\gamma_{1,0}^{(-1,-1)} + \dots; \end{aligned} \quad (17)$$

We denote the probability of finding the transmon at energy level n by $p_n(t) := \text{Tr}(\bar{\rho}(t)|n\rangle_a \langle n|_a)$, and define the corresponding reduced density matrix of the resonator to be $\bar{\rho}_{c;n}(t) := \frac{1}{p_n} \text{Tr}_a(\bar{\rho}(t)|n\rangle_a \langle n|_a)$. Notice that after time-coarse graining, p_n can be very small, but in general remains nonzero. The TCG master equation then implies that

$$\begin{aligned}
& \partial_t(p_n \bar{\rho}_{c;n}) \\
&= -ip_n \left[h_1(n)n_c + h_2(n)n_c^2 + \left(h_0^{(-1)}(n)c + h_1^{(-1)}(n)c^\dagger c^2 + h_0^{(-2)}(n)c^2 + h.c. \right), \bar{\rho}_{c;n} \right] \\
&+ p_n d_{0,0}^{(-1,-1)}(n) D_{c,c^\dagger} \bar{\rho}_{c;n} + p_n \left[d_{0,0}^{(-2,-1)}(n) D_{c^2,c^\dagger} + d_{0,0}^{(-2,-1)}(n)^* D_{c,c^\dagger^2} + d_{0,0}^{(1,2)}(n) D_{c^\dagger,c^2} + d_{0,0}^{(1,2)}(n)^* D_{c^\dagger^2,c} \right] \bar{\rho}_{c;n} \\
&- p_n \left(\kappa_{0,0}^{(0,0)}(n) + \gamma_{0,0}^{(0,0)}(n) \right) \bar{\rho}_{c;n} - p_n \left(2\text{Re}(\kappa_{0,1}^{(0,0)}(n) + 2\kappa_{1,0}^{(1,1)}(n)) + 2\text{Re}(\gamma_{0,1}^{(0,0)}(n) + 2\gamma_{1,0}^{(1,1)}(n)) \right) \cdot \frac{c^\dagger c \bar{\rho}_{c;n} + \bar{\rho}_{c;n} c^\dagger c}{2} \\
&- p_n \left(\kappa_{0,0}^{(-1,0)}(n) + \kappa_{0,0}^{(0,1)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,0}^{(-1,0)}(n) + \gamma_{0,0}^{(0,1)}(n) + \gamma_{1,0}^{(0,1)}(n) \right) \frac{c \bar{\rho}_{c;n} + \bar{\rho}_{c;n} c}{2} \\
&- p_n \left(\kappa_{0,0}^{(-1,0)}(n)^* + \kappa_{0,0}^{(0,1)}(n)^* + \kappa_{1,0}^{(0,1)}(n)^* + \gamma_{0,0}^{(-1,0)}(n)^* + \gamma_{0,0}^{(0,1)}(n)^* + \gamma_{1,0}^{(0,1)}(n)^* \right) \frac{c^\dagger \bar{\rho}_{c;n} + \bar{\rho}_{c;n} c^\dagger}{2} \\
&- p_n \left(\kappa_{0,1}^{(-1,0)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,1}^{(-1,0)}(n) + \gamma_{1,0}^{(0,1)}(n) \right) \frac{c^\dagger c^2 \bar{\rho}_{c;n} + \bar{\rho}_{c;n} c^\dagger c^2}{2} \\
&- p_n \left(\kappa_{0,1}^{(-1,0)}(n)^* + \kappa_{1,0}^{(0,1)}(n)^* + \gamma_{0,1}^{(-1,0)}(n)^* + \gamma_{1,0}^{(0,1)}(n)^* \right) \frac{c^{\dagger 2} c \bar{\rho}_{c;n} + \bar{\rho}_{c;n} c^{\dagger 2} c}{2} \\
&+ p_{n+1} \left[\kappa_{0,0}^{(0,0)}(n+1) \bar{\rho}_{c;n+1} \right] + p_{n+1} \left[\kappa_{0,0}^{(-1,0)}(n+1) c \bar{\rho}_{c;n+1} + \kappa_{0,0}^{(0,1)}(n+1) \bar{\rho}_{c;n+1} c \right] \\
&+ p_{n+1} \left[\kappa_{0,0}^{(-1,0)}(n+1)^* \bar{\rho}_{c;n+1} c^\dagger + \kappa_{0,0}^{(0,1)}(n+1)^* c^\dagger \bar{\rho}_{c;n+1} \right] \\
&+ p_{n+1} \left[\kappa_{0,1}^{(-1,0)}(n+1) c \bar{\rho}_{c;n+1} c^\dagger + \kappa_{1,0}^{(0,1)}(n+1) c^\dagger c \bar{\rho}_{c;n+1} + \kappa_{0,1}^{(-1,0)}(n+1)^* c^\dagger c \bar{\rho}_{c;n+1} c^\dagger + \kappa_{1,0}^{(0,1)}(n+1)^* c^\dagger \bar{\rho}_{c;n+1} c^\dagger c \right] \\
&+ p_{n+1} \left[\kappa_{1,0}^{(-1,-1)}(n+1) c^\dagger c^2 \bar{\rho}_{c;n+1} c^\dagger + \kappa_{1,0}^{(1,1)}(n+1) c^{\dagger 2} c \bar{\rho}_{c;n+1} + \kappa_{0,1}^{(0,0)}(n+1) \bar{\rho}_{c;n+1} c^\dagger c \right. \\
&\quad \left. + \kappa_{1,0}^{(-1,-1)}(n+1)^* c \bar{\rho}_{c;n+1} c^{\dagger 2} c + \kappa_{1,0}^{(1,1)}(n+1)^* c^\dagger \bar{\rho}_{c;n+1} c^\dagger c^2 + \kappa_{0,1}^{(0,0)}(n+1)^* c^\dagger c \bar{\rho}_{c;n+1} \right] \\
&+ p_{n-1} \left[\gamma_{0,0}^{(0,0)}(n-1) \bar{\rho}_{c;n-1} \right] + p_{n-1} \left[\gamma_{0,0}^{(-1,0)}(n-1) c \bar{\rho}_{c;n-1} + \gamma_{0,0}^{(0,1)}(n-1) \bar{\rho}_{c;n-1} c \right] \\
&+ p_{n-1} \left[\gamma_{0,0}^{(-1,0)}(n-1)^* \bar{\rho}_{c;n-1} c^\dagger + \gamma_{0,0}^{(0,1)}(n-1)^* c^\dagger \bar{\rho}_{c;n-1} \right] \\
&+ p_{n-1} \left[\gamma_{0,1}^{(-1,0)}(n-1) c \bar{\rho}_{c;n-1} c^\dagger + \gamma_{1,0}^{(0,1)}(n-1) c^\dagger c \bar{\rho}_{c;n-1} + \gamma_{0,1}^{(-1,0)}(n-1)^* c^\dagger c \bar{\rho}_{c;n-1} c^\dagger + \gamma_{1,0}^{(0,1)}(n-1)^* c^\dagger \bar{\rho}_{c;n-1} c^\dagger c \right] \\
&+ p_{n-1} \left[\gamma_{1,0}^{(-1,-1)}(n-1) c^\dagger c^2 \bar{\rho}_{c;n-1} c^\dagger + \gamma_{1,0}^{(1,1)}(n-1) c^{\dagger 2} c \bar{\rho}_{c;n-1} + \gamma_{0,1}^{(0,0)}(n-1) \bar{\rho}_{c;n-1} c^\dagger c \right. \\
&\quad \left. + \gamma_{1,0}^{(-1,-1)}(n-1)^* c \bar{\rho}_{c;n-1} c^{\dagger 2} c + \gamma_{1,0}^{(1,1)}(n-1)^* c^\dagger \bar{\rho}_{c;n-1} c^\dagger c^2 + \gamma_{0,1}^{(0,0)}(n-1)^* c^\dagger c \bar{\rho}_{c;n-1} \right].
\end{aligned} \tag{18}$$

And this equation for the conditional reduced density matrix $\bar{\rho}_{c;n}$ gives rise to the following equations of motion for p_n and the central moments $E_{O_c}(n) := \text{Tr}(\bar{\rho}(t) O_c | n \rangle_a \langle n |_a)$ for any resonator operators O_c :

$$\begin{aligned}
\partial_t p_n &= -p_n \left(\kappa_{0,0}^{(0,0)}(n) + \gamma_{0,0}^{(0,0)}(n) \right) - 2p_n \left(\text{Re}(2\kappa_{1,0}^{(1,1)}(n) + \kappa_{0,1}^{(0,0)}(n)) + \text{Re}(2\gamma_{1,0}^{(1,1)}(n) + \gamma_{0,1}^{(0,0)}(n)) \right) \cdot E_{c^\dagger c}(n) \\
&- 2p_n \text{Re} \left[\left(\kappa_{0,0}^{(-1,0)}(n) + \kappa_{0,0}^{(0,1)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,0}^{(-1,0)}(n) + \gamma_{0,0}^{(0,1)}(n) + \gamma_{1,0}^{(0,1)}(n) \right) C_c(n) \right] \\
&- 2p_n \text{Re} \left[\left(\kappa_{0,1}^{(-1,0)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,1}^{(-1,0)}(n) + \gamma_{1,0}^{(0,1)}(n) \right) E_{c^\dagger c^2}(n) \right] \\
&+ p_{n+1} \kappa_{0,0}^{(0,0)}(n+1) + 2p_{n+1} \text{Re} \left[\kappa_{0,1}^{(0,0)}(n+1) + 2\kappa_{1,0}^{(1,1)}(n+1) \right] E_{c^\dagger c}(n+1) \\
&+ 2p_{n+1} \text{Re} \left[\left(\kappa_{0,0}^{(-1,0)}(n+1) + \kappa_{0,0}^{(0,1)}(n+1) + \kappa_{1,0}^{(0,1)}(n+1) \right) C_c(n+1) \right] \\
&+ 2p_{n+1} \text{Re} \left[\left(\kappa_{0,1}^{(-1,0)}(n+1) + \kappa_{1,0}^{(0,1)}(n+1) \right) E_{c^\dagger c^2}(n+1) \right] \\
&+ p_{n-1} \gamma_{0,0}^{(0,0)}(n-1) + 2p_{n-1} \text{Re} \left[\gamma_{0,1}^{(0,0)}(n-1) + 2\gamma_{1,0}^{(1,1)}(n-1) \right] E_{c^\dagger c}(n-1) \\
&+ 2p_{n-1} \text{Re} \left[\left(\gamma_{0,0}^{(-1,0)}(n-1) + \gamma_{0,0}^{(0,1)}(n-1) + \gamma_{1,0}^{(0,1)}(n-1) \right) C_c(n-1) \right] \\
&+ 2p_{n-1} \text{Re} \left[\left(\gamma_{0,1}^{(-1,0)}(n-1) + \gamma_{1,0}^{(0,1)}(n-1) \right) E_{c^\dagger c^2}(n-1) \right]
\end{aligned} \tag{19}$$

$$\begin{aligned}
& \partial_t(p_n C_c(n)) \\
&= -\frac{1}{2}p_n d_{0,0}^{(-1,-1)}(n)C_c(n) - ip_n \left((h_1(n) + h_2(n))C_c(n) + 2h_2(n)E_{c^\dagger c^2}(n) + 2h_0^{(-2)}(n)^*C_c(n)^* \right) \\
&+ \frac{1}{2}p_n (d_{0,0}^{(1,2)}(n) - d_{0,0}^{(-2,-1)}(n))E_{c^2}(n) - ip_n \left(h_0^{(-1)}(n)^* + h_1^{(-1)}(n)E_{c^2}(n) + 2h_1^{(-1)}(n)^*E_{c^\dagger c}(n) \right) \\
&+ p_n (d_{0,0}^{(1,2)}(n)^* - d_{0,0}^{(-2,-1)}(n)^*)E_{c^\dagger c}(n) + p_n d_{0,0}^{(1,2)}(n)^* - p_n (\kappa_{0,0}^{(0,0)}(n) + \gamma_{0,0}^{(0,0)}(n))C_c(n) \\
&- p_n \left(2\text{Re}(\kappa_{0,1}^{(0,0)}(n) + 2\kappa_{1,0}^{(1,1)}(n)) + 2\text{Re}(\gamma_{0,1}^{(0,0)}(n) + 2\gamma_{1,0}^{(1,1)}(n)) \right) \left(\frac{1}{2}C_c(n) + E_{c^\dagger c^2}(n) \right) \\
&- p_n (\kappa_{0,0}^{(-1,0)}(n) + \kappa_{0,0}^{(0,1)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,0}^{(-1,0)}(n) + \gamma_{0,0}^{(0,1)}(n) + \gamma_{1,0}^{(0,1)}(n))E_{c^2}(n) \\
&- p_n (\kappa_{0,0}^{(-1,0)}(n)^* + \kappa_{0,0}^{(0,1)}(n)^* + \kappa_{1,0}^{(0,1)}(n)^* + \gamma_{0,0}^{(-1,0)}(n)^* + \gamma_{0,0}^{(0,1)}(n)^* + \gamma_{1,0}^{(0,1)}(n)^*) \left(\frac{1}{2} + E_{c^\dagger c}(n) \right) \\
&- p_n (\kappa_{0,1}^{(-1,0)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,1}^{(-1,0)}(n) + \gamma_{1,0}^{(0,1)}(n)) \left(\frac{1}{2}E_{c^2}(n) + E_{c^\dagger c^3} \right) \\
&- p_n (\kappa_{0,1}^{(-1,0)}(n)^* + \kappa_{1,0}^{(0,1)}(n)^* + \gamma_{0,1}^{(-1,0)}(n)^* + \gamma_{1,0}^{(0,1)}(n)^*) (E_{c^\dagger c}(n) + E_{c^\dagger c^2}) \\
&+ p_{n+1} (\kappa_{0,0}^{(0,0)}(n+1) + \kappa_{0,1}^{(0,0)}(n+1)^* + 2\kappa_{1,0}^{(1,1)}(n+1))C_c(n+1) + p_{n+1} (\kappa_{0,0}^{(-1,0)}(n+1) + \kappa_{0,0}^{(0,1)}(n+1))E_{c^2}(n+1) \\
&+ p_{n+1} \left(\kappa_{1,0}^{(-1,-1)}(n+1) + \kappa_{1,0}^{(1,1)}(n+1) + 2\text{Re}(\kappa_{0,1}^{(0,0)}(n+1)) \right) E_{c^\dagger c^2}(n+1) \\
&+ p_{n+1} \left((\kappa_{0,0}^{(-1,0)}(n+1)^* + \kappa_{0,0}^{(0,1)}(n+1)^*)E_{c^\dagger c}(n+1) + \kappa_{0,0}^{(0,1)}(n+1)^* \right) \\
&+ p_{n+1} \left(2\kappa_{1,0}^{(0,1)}(n+1)E_{c^2}(n+1) + (\kappa_{0,1}^{(-1,0)}(n+1) + \kappa_{1,0}^{(0,1)}(n+1))E_{c^\dagger c^3}(n+1) \right) \\
&+ p_{n+1} \left((\kappa_{0,1}^{(-1,0)}(n+1)^* + 2\kappa_{1,0}^{(0,1)}(n+1)^*)E_{c^\dagger c}(n+1) + (\kappa_{0,1}^{(-1,0)}(n+1)^* + \kappa_{1,0}^{(0,1)}(n+1)^*)E_{c^\dagger c^2}(n+1) \right) \\
&+ p_{n-1} (\gamma_{0,0}^{(0,0)}(n-1) + \gamma_{0,1}^{(0,0)}(n-1)^* + 2\gamma_{1,0}^{(1,1)}(n-1))C_c(n-1) + p_{n-1} (\gamma_{0,0}^{(-1,0)}(n-1) + \gamma_{0,0}^{(0,1)}(n-1))E_{c^2}(n-1) \\
&+ p_{n-1} \left(\gamma_{1,0}^{(-1,-1)}(n-1) + \gamma_{1,0}^{(1,1)}(n-1) + 2\text{Re}(\gamma_{0,1}^{(0,0)}(n-1)) \right) E_{c^\dagger c^2}(n-1) \\
&+ p_{n-1} \left((\gamma_{0,0}^{(-1,0)}(n-1)^* + \gamma_{0,0}^{(0,1)}(n-1)^*)E_{c^\dagger c}(n-1) + \gamma_{0,0}^{(0,1)}(n-1)^* \right) \\
&+ p_{n-1} \left(2\gamma_{1,0}^{(0,1)}(n-1)E_{c^2}(n-1) + (\gamma_{0,1}^{(-1,0)}(n-1) + \gamma_{1,0}^{(0,1)}(n-1))E_{c^\dagger c^3}(n-1) \right) \\
&+ p_{n-1} \left((\gamma_{0,1}^{(-1,0)}(n-1)^* + 2\gamma_{1,0}^{(0,1)}(n-1)^*)E_{c^\dagger c}(n-1) + (\gamma_{0,1}^{(-1,0)}(n-1)^* + \gamma_{1,0}^{(0,1)}(n-1)^*)E_{c^\dagger c^2}(n-1) \right)
\end{aligned} \tag{I10}$$

$$\begin{aligned}
& \partial_t \left(p_n E_{c^2}(n) \right) \\
&= -2ip_n \left((h_1(n) + 2h_2(n)) E_{c^2}(n) + 2h_2(n) E_{c^\dagger c^3}(n) + h_1^{(-1)}(n) E_{c^3}(n) + h_0^{(-2)}(n)^* (1 + 2E_{c^\dagger c}(n)) \right) \\
&\quad - 2ip_n \left((h_0^{(-1)}(n)^* + h_1^{(-1)}(n)^*) C_c(n) + 2h_1^{(-1)}(n)^* E_{c^\dagger c^2}(n) \right) \\
&\quad - p_n \left((d_{0,0}^{(-1,-1)}(n) + \kappa_{0,0}^{(0,0)}(n) + \gamma_{0,0}^{(0,0)}(n)) E_{c^2}(n) + (d_{0,0}^{(-2,-1)}(n) + d_{0,0}^{(1,2)}(n)) E_{c^3}(n) \right) \\
&\quad - p_n \left((d_{0,0}^{(-2,-1)}(n)^* + 3d_{0,0}^{(1,2)}(n)^*) C_c(n) + 2(d_{0,0}^{(-2,-1)}(n)^* + d_{0,0}^{(1,2)}(n)^*) E_{c^\dagger c^2}(n) \right) \\
&\quad - p_n \left(2\text{Re}[\kappa_{0,1}^{(0,0)}(n) + 2\kappa_{1,0}^{(1,1)}(n)] + 2\text{Re}[\gamma_{0,1}^{(0,0)}(n) + 2\gamma_{1,0}^{(1,1)}(n)] \right) (E_{c^2}(n) + E_{c^\dagger c^3}(n)) \\
&\quad - p_n (\kappa_{0,0}^{(-1,0)}(n) + \kappa_{0,0}^{(0,1)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,0}^{(-1,0)}(n) + \gamma_{0,0}^{(0,1)}(n) + \gamma_{1,0}^{(0,1)}(n)) E_{c^3}(n) \\
&\quad - p_n (\kappa_{0,0}^{(-1,0)}(n)^* + \kappa_{0,0}^{(0,1)}(n)^* + \kappa_{1,0}^{(0,1)}(n)^* + \gamma_{0,0}^{(-1,0)}(n)^* + \gamma_{0,0}^{(0,1)}(n)^* + \gamma_{1,0}^{(0,1)}(n)^*) (C_c(n) + E_{c^\dagger c^2}(n)) \\
&\quad - p_n (\kappa_{0,1}^{(-1,0)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,1}^{(-1,0)}(n) + \gamma_{1,0}^{(0,1)}(n)) (E_{c^3}(n) + E_{c^\dagger c^4}) \\
&\quad - p_n (\kappa_{0,1}^{(-1,0)}(n)^* + \kappa_{1,0}^{(0,1)}(n)^* + \gamma_{0,1}^{(-1,0)}(n)^* + \gamma_{1,0}^{(0,1)}(n)^*) (C_c(n) + 2E_{c^\dagger c^2}(n) + E_{c^\dagger c^3}) \\
&\quad + p_{n+1} (\kappa_{0,0}^{(0,0)}(n+1)) E_{c^2}(n+1) + 2p_{n+1} (\kappa_{0,1}^{(0,0)}(n+1)^* + 3\kappa_{1,0}^{(1,1)}(n+1)) E_{c^2}(n+1) \\
&\quad + 2p_{n+1} (\text{Re}[\kappa_{0,1}^{(0,0)}(n+1)] + \kappa_{1,0}^{(-1,-1)}(n+1) + \kappa_{1,0}^{(1,1)}(n+1)) E_{c^\dagger c^3}(n+1) \\
&\quad + p_{n+1} (\kappa_{0,0}^{(-1,0)}(n+1) + \kappa_{0,0}^{(0,1)}(n+1)) E_{c^3}(n+1) + 2p_{n+1} \kappa_{0,0}^{(0,1)}(n+1)^* C_c(n+1) \\
&\quad + p_{n+1} (\kappa_{0,0}^{(-1,0)}(n+1)^* + \kappa_{0,0}^{(0,1)}(n+1)^*) E_{c^\dagger c^2}(n+1) \\
&\quad + p_{n+1} \left((\kappa_{0,1}^{(-1,0)}(n+1) + \kappa_{1,0}^{(0,1)}(n+1)) E_{c^\dagger c^4}(n+1) + 3\kappa_{1,0}^{(0,1)}(n+1) E_{c^3}(n+1) \right) \\
&\quad + p_{n+1} \left((2\kappa_{0,1}^{(-1,0)}(n+1)^* + 3\kappa_{1,0}^{(0,1)}(n+1)^*) E_{c^\dagger c^2}(n+1) + (\kappa_{0,1}^{(-1,0)}(n+1)^* + \kappa_{1,0}^{(0,1)}(n+1)^*) E_{c^\dagger c^3}(n+1) \right) \\
&\quad + p_{n-1} (\gamma_{0,0}^{(0,0)}(n-1)) E_{c^2}(n-1) + 2p_{n-1} (\gamma_{0,1}^{(0,0)}(n-1)^* + 3\gamma_{1,0}^{(1,1)}(n-1)) E_{c^2}(n-1) \\
&\quad + 2p_{n-1} (\text{Re}[\gamma_{0,1}^{(0,0)}(n-1)] + \gamma_{1,0}^{(-1,-1)}(n-1) + \gamma_{1,0}^{(1,1)}(n-1)) E_{c^\dagger c^3}(n-1) \\
&\quad + p_{n-1} (\gamma_{0,0}^{(-1,0)}(n-1) + \gamma_{0,0}^{(0,1)}(n-1)) E_{c^3}(n-1) + 2p_{n-1} \gamma_{0,0}^{(0,1)}(n-1)^* C_c(n-1) \\
&\quad + p_{n-1} (\gamma_{0,0}^{(-1,0)}(n-1)^* + \gamma_{0,0}^{(0,1)}(n-1)^*) E_{c^\dagger c^2}(n-1) \\
&\quad + p_{n-1} \left((\gamma_{0,1}^{(-1,0)}(n-1) + \gamma_{1,0}^{(0,1)}(n-1)) E_{c^\dagger c^4}(n-1) + 3\gamma_{1,0}^{(0,1)}(n-1) E_{c^3}(n-1) \right) \\
&\quad + p_{n-1} \left((2\gamma_{0,1}^{(-1,0)}(n-1)^* + 3\gamma_{1,0}^{(0,1)}(n-1)^*) E_{c^\dagger c^2}(n-1) + (\gamma_{0,1}^{(-1,0)}(n-1)^* + \gamma_{1,0}^{(0,1)}(n-1)^*) E_{c^\dagger c^3}(n-1) \right)
\end{aligned} \tag{H11}$$

$$\begin{aligned}
& \partial_t \left(p_n E_{c^\dagger c}(n) \right) \\
&= -2p_n \text{Im} \left[h_0^{(-1)}(n) C_c(n) + h_1^{(-1)}(n) E_{c^\dagger c^2}(n) + 2h_0^{(-2)} E_{c^2}(n) \right] - p_n d_{0,0}^{(-1,-1)}(n) E_{c^\dagger c}(n) \\
&+ 3p_n \text{Re} \left[2d_{0,0}^{(1,2)}(n) C_c(n) + (d_{0,0}^{(1,2)}(n) - d_{0,0}^{(-2,-1)}(n)) E_{c^\dagger c^2}(n) \right] - p_n (\kappa_{0,0}^{(0,0)}(n) + \gamma_{0,0}^{(0,0)}(n)) E_{c^\dagger c}(n) \\
&- 2p_n \text{Re} \left[\kappa_{0,1}^{(0,0)}(n) + 2\kappa_{1,0}^{(1,1)}(n) + \gamma_{0,1}^{(0,0)}(n) + 2\gamma_{1,0}^{(1,1)}(n) \right] (E_{c^\dagger c}(n) + E_{c^\dagger c^2}(n)) \\
&- p_n \text{Re} \left[(\kappa_{0,0}^{(-1,0)}(n) + \kappa_{0,0}^{(0,1)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,0}^{(-1,0)}(n) + \gamma_{0,0}^{(0,1)}(n) + \gamma_{1,0}^{(0,1)}(n)) (C_c(n) + 2E_{c^\dagger c^2}(n)) \right] \\
&- p_n \text{Re} \left[(\kappa_{0,1}^{(-1,0)}(n) + \kappa_{1,0}^{(0,1)}(n) + \gamma_{0,1}^{(-1,0)}(n) + \gamma_{1,0}^{(0,1)}(n)) (3E_{c^\dagger c^2} + 2E_{c^\dagger c^3}) \right] \\
&+ p_{n+1} (\kappa_{0,0}^{(0,0)}(n+1) + 2\text{Re}[\kappa_{0,1}^{(0,0)}(n+1)]) E_{c^\dagger c}(n+1) + 2p_{n+1} \text{Re}[\kappa_{0,1}^{(0,0)}(n+1)] E_{c^\dagger c^2}(n+1) \\
&+ 2p_{n+1} \text{Re}[\kappa_{0,0}^{(0,1)}(n+1) C_c(n+1)] + 2p_{n+1} \text{Re}[(\kappa_{0,0}^{(-1,0)}(n+1) + \kappa_{0,0}^{(0,1)}(n+1)) E_{c^\dagger c^2}(n+1)] \\
&+ 2p_{n+1} \text{Re}[\kappa_{1,0}^{(0,1)}(n+1) C_c(n+1) + (\kappa_{0,1}^{(-1,0)}(n+1) + 3\kappa_{1,0}^{(0,1)}(n+1)) E_{c^\dagger c^2}(n+1)] \\
&+ 2p_{n+1} \text{Re}[(\kappa_{0,1}^{(-1,0)}(n+1) + \kappa_{1,0}^{(0,1)}(n+1)) E_{c^\dagger c^3}(n+1)] \\
&+ p_{n-1} (\gamma_{0,0}^{(0,0)}(n-1) + 2\text{Re}[\gamma_{0,1}^{(0,0)}(n-1)]) E_{c^\dagger c}(n-1) + 2p_{n-1} \text{Re}[\gamma_{0,1}^{(0,0)}(n-1)] E_{c^\dagger c^2}(n-1) \\
&+ 2p_{n-1} \text{Re}[\gamma_{0,0}^{(0,1)}(n-1) C_c(n-1)] + 2p_{n-1} \text{Re}[(\gamma_{0,0}^{(-1,0)}(n-1) + \gamma_{0,0}^{(0,1)}(n-1)) E_{c^\dagger c^2}(n-1)] \\
&+ 2p_{n-1} \text{Re}[\gamma_{1,0}^{(0,1)}(n-1) C_c(n-1) + (\gamma_{0,1}^{(-1,0)}(n-1) + 3\gamma_{1,0}^{(0,1)}(n-1)) E_{c^\dagger c^2}(n-1)] \\
&+ 2p_{n-1} \text{Re}[(\gamma_{0,1}^{(-1,0)}(n-1) + \gamma_{1,0}^{(0,1)}(n-1)) E_{c^\dagger c^3}(n-1)]
\end{aligned} \tag{I12}$$

However, the equations of motion (EOM) above are not closed with a finite number of central moment variables. In order to obtain a closed set of EOM, we assume that the conditional resonator states do not deviate very significantly from coherent states so that we can perform a cumulant expansion to central moments and truncate at a certain order in the cumulant variables. For the numerical simulations in this paper, we make the following truncation at the second-order cumulants to close the EOM with the variables $p_n, C_c(n), C_{c^2}(n)$ and $C_{c^\dagger c}(n)$:

$$\begin{aligned}
C_c(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a \bar{c} \bar{\rho}(t)) & E_{c^2}(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a c^2 \bar{\rho}(t)) = C_c(n)^2 + C_{c^2}(n) \\
E_{c^\dagger c}(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a c^\dagger \bar{c} \bar{\rho}(t)) = |C_c(n)|^2 + C_{c^\dagger c}(n) \\
E_{c^\dagger c^2}(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a c^\dagger c^2 \bar{\rho}(t)) \approx |C_c(n)|^2 C_c(n) + C_c(n)^* C_{c^2}(n) + 2C_{c^\dagger c}(n) C_c(n) \\
E_{c^3}(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a c^3 \bar{\rho}(t)) \approx C_c(n)^3 + 3C_{c^2}(n) C_c(n) \\
E_{c^\dagger c^3}(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a c^\dagger c^3 \bar{\rho}(t)) \approx |C_c(n)|^2 C_c(n)^2 + 3C_{c^\dagger c}(n) C_c(n)^2 + 3C_{c^2}(n) |C_c(n)|^2 + 3C_{c^\dagger c}(n) C_{c^2}(n) \\
E_{c^\dagger c^2 c^2}(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a c^\dagger c^2 c^2 \bar{\rho}(t)) \\
&\approx |C_c(n)|^4 + C_{c^2}(n)^* C_c(n)^2 + C_c(n)^* C_{c^2}(n) + 4C_{c^\dagger c}(n) |C_c(n)|^2 + |C_{c^2}(n)|^2 + 2C_{c^\dagger c}(n)^2 \\
E_{c^\dagger c^4}(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a c^\dagger c^4 \bar{\rho}(t)) \\
&\approx |C_c(n)|^2 C_c(n)^3 + 4C_{c^\dagger c}(n) C_c(n)^3 + 6C_{c^2}(n) |C_c(n)|^2 C_c(n) + 12C_{c^\dagger c}(n) C_{c^2}(n) C_c(n) + 3C_{c^2}(n)^2 C_c(n)^* \\
E_{c^\dagger c^3 c^3}(n) &:= \frac{1}{p_n} \text{Tr}(|n\rangle_a \langle n|_a c^\dagger c^3 c^3 \bar{\rho}(t)) \\
&\approx |C_c(n)|^4 C_c(n) + C_{c^2}(n)^* C_c(n)^3 + 6C_{c^\dagger c}(n) |C_c(n)|^2 C_c(n) + 3C_{c^2}(n) |C_c(n)|^2 C_c(n)^* + 3|C_{c^2}(n)|^2 C_c(n) \\
&\quad + 6C_{c^\dagger c}(n)^2 C_c(n) + 6C_{c^\dagger c}(n) C_{c^2}(n) C_c(n)^*
\end{aligned} \tag{I13}$$

In addition, the EOM are fully closed by truncating the transmon Hilbert space at a certain energy level. In this paper, we make the approximation that $p_n = 0$ for all $n > 6$, which is sufficient for simulating the dynamics corresponding to the first six energy levels of the transmon.

Finally, to calculate the numerical values of the coefficients in the EME, we assume the following experimentally relevant parameters:

$$\begin{aligned}\epsilon &= 0.2; & \omega_c &= 2\pi \times 7\text{GHz}; & \omega_{01} &\equiv \omega_a \left(1 - \frac{\epsilon}{4}\right) = 2\pi \times 5\text{GHz}; \\ g_{ac} &= 9.6 \times 10^{-3} \omega_{01} = 0.3016\text{GHz}; & \kappa_c &= 0.48\text{MHz},\end{aligned}\tag{I14}$$

whereas the spectral density of the bath takes the form

$$J(\omega) = 1.09 \times 10^{-5} \cdot \frac{\omega}{1 + (\frac{\omega}{\Lambda})^2}\tag{I15}$$

with a soft cut-off scale set at $\Lambda = 100 \cdot 2\pi\text{GHz}$. We emphasize that the dynamics of the system is very insensitive to the exact value of Λ which depends on the physical details of the resonator-bath coupling.

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