Nonlinear quantum Langevin equations for bosonic modes in solid-state systems

Juuso Manninen
Department of Applied Physics, Low Temperature Laboratory,
Aalto University, PO Box 15100, FI-00076 AALTO, Finland

Souvik Agasti and Francesco Massel
Department of Physics and Nanoscience Center, University of Jyväskylä,
P.O. Box 35 (YFL), FI-40014 University of Jyväskylä, Finland

Based on the experimental evidence that impurities contribute to the dissipation properties of solid-state open quantum systems, we provide here a description in terms of nonlinear quantum Langevin equations of the role played by two-level systems in the dynamics of a bosonic degree of freedom. Our starting point is represented by the description of the system/environment coupling in terms of coupling to two separate reservoirs, modelling the interaction with external bosonic modes and two level systems, respectively. Furthermore, we show how this model represents a specific example of a class of open quantum systems that can be described by nonlinear quantum Langevin equations. Our analysis offers a potential explanation of the parametric effects recently observed in circuit-QED cavity optomechanics experiments.

The dynamics of open quantum systems – i.e. quantum systems that can be described as separate entities from their surrounding environment while being somehow coupled to it – is arguably one of the most fundamental problems in quantum mechanics, encompassing concepts such as the measurement paradox [1] and the boundary between quantum and classical physics [2]. On general grounds, the interaction between a quantum system and its environment represents an important aspect of the physics of condensed matter and complex systems, which has been the focus of extensive analysis [3–5], with repercussions in contexts ranging from the energy transport in photosynthetic complexes [6] to the physics of ultracold gases [7–9].

In the description of these systems the inclusion of the role played by coupling to an external environment is necessary, if only because the system has to be coupled to an external measurement apparatus which, from the quantum-dynamical perspective of the system, represents a source of noise and dissipation. At the same time the manipulation of open quantum systems has recently led to the possibility of preparing and detecting quantum states of matter and complex systems, which has been the focus of extensive analysis [3–5], with repercussions in contexts ranging from the energy transport in photosynthetic complexes [6] to the physics of ultracold gases [7–9].

The experimental evidence of nonlinear phenomena related to the coupling between system and environment has emerged and, more importantly for our analysis, the relevance of impurities in this phenomenon has been dis-
discussed. In both setups, it has been shown that the impurities, naturally arising in the material composing the devices, its supports and/or substrate, represent a source of dissipation. These defects can be modelled in terms of TLSs. The reason behind the possibility of modelling impurities in these terms is represented by the fact that each impurity can be construed as quantum systems which exhibit two local energy minima. For instance as a charged impurity that can hop between two defects in the crystal structure, or a dangling bond with two possible configurations.

More specifically, these TLSs exist primarily due to the disordered potential landscape of amorphous materials – e.g. in surface oxides of thin-film circuit electrodes [38], in the tunnel barrier of Josephson junctions [34], and at disordered interfaces [40, 41] – coupling with the bosonic degrees of freedom of the system, either through a purely electromagnetic interaction (optical and circuit QED setups) or a phononic one in the context of nanomechanical systems [22].

In this Letter we show under what conditions, considering a nonlinear coupling between system and a bath of TLSs, it is possible to derive a nonlinear QLE for the dynamics of the degrees of freedom of the system, having in mind a circuit QED setup. In addition, we show how the nonlinear QLEs derived here can represent an explanation to some of the phenomena recently observed in the context of microwave quantum optomechanics [22].

The starting point for our analysis is represented by a bosonic system ($\mathcal{S}$) coupled to an environment ($\mathcal{E}$). The total Hamiltonian of the bipartite system ($\mathcal{S} + \mathcal{E}$) is given by

$$H = H_{\mathcal{S}} + H_{\mathcal{E}} + H_{\mathcal{S} - \mathcal{E}},$$

where $H_{\mathcal{S}} = H_{\mathcal{S}}(c, c^\dagger)$ is the Hamiltonian of the isolated system, exhibiting a generic dependence on the annihilation (creation) operators $c$ ($c^\dagger$) associated with the system, and $H_{\mathcal{E}}$ is the Hamiltonian for the bath.

We assume here that the environment Hamiltonian can be decomposed into two terms, $H_{\mathcal{E}}^B = \sum_k \omega_k b_k b_\dagger_k$ and $H_{\mathcal{E}}^{TLS}$, corresponding to a bath of free bosonic modes, and to a bath of TLSs, respectively. The bosonic bath describes, for instance, the modes of the electromagnetic field of the environment. In our analysis we assume that these modes, while being associated with the noise properties and dissipation of the system, encompass also the external coherent fields driving the system whose properties are encoded in the state of the bath for the modes $b_k$. Our choice is equivalent to considering a coherent driving term for the system Hamiltonian and a purely thermal bath.

In this scenario, we describe the coupling between these modes and the degrees of freedom of the system by the following Hamiltonian

$$H_{\mathcal{S} - \mathcal{B}} = \sum_{k} \Omega_{k} b_{k}^\dagger b_{k} + c_{k}^\dagger c_{k} - \Omega_{k} c_{k} c_{k}^\dagger,$$

where $\Omega_{k}$ is the coupling between the two baths and the system is mediated by the Hamiltonians $H_{\mathcal{S} - \mathcal{B}}$ and $H_{\mathcal{S} - TLS}$, respectively.

This choice for the modeling of TLSs corresponds to the idea that, for each $\Omega_{k}$ multiple TLSs are present that collectively couple with the system $\mathcal{S}$. While for $\Omega_{k} \approx \omega_{S}$ – where $\omega_{S}$ corresponds to a characteristic frequency for the system – the presence of impurities leads to a renormalization of the linewidth associated with the linear response of the system induced by the coupling given in Eq. (2) (see Appendix D); for $\Omega_{k} \approx n \omega_{S}$, nonlinear contributions appear. In our analysis, also in light of the recent investigations concerning the relevance of two-photon emission processes by TLSs [43, 44], when coupled to bosonic modes, we consider the case $n = 2$, representing the lowest-order approximation beyond linear coupling. This assumption appears to be compatible with the usual experimental conditions encountered in the context of circuit QED where microwave cavities operate at frequencies corresponding to few GHz [15, 16, 45] while the energy separation of a TLS relevant for the physics of either of these systems is of the order of 10 GHz [45, 46]. In this case, it is possible to write the system-TLS coupling Hamiltonian as

$$H_{\mathcal{S} - TLS} = \sum_{k} \Omega_{k}^{TLS} \left( J_{k} c_{k}^2 + J_{k}^* c_{k}^* c_{k}^2 \right).$$

If we assume that $|J_{k}| \gg 1$, corresponding to the idea, that, for each value of $k$ multiple TLSs couple to the system $\mathcal{S}$, by resorting to the Holstein-Primakoff (HP) realization of spin operators in terms of bosonic modes, we can replace the spin operators with bosonic ones. This mapping can be performed in two different ways, corresponding to complementary experimental conditions (see Appendix A). If it is assumed that the TLSs mostly reside in their ground state, we have that $J_{k} = -J_{k}$ – where $J_{k}$ is the index of the representation associated with the spin $J_{k}$ – and the HP mapping reads $J_{k} \rightarrow d_{k}^0 d_{k} - J_{k}$, $J_{k}^* \rightarrow d_{k}^0$, $J_{k}^* \rightarrow d_{k}^1$. In this case, the coupling between

![FIG. 1. Cartoon picture of the setup. The system $\mathcal{S}$ is coupled to an environment $\mathcal{E}$, which is constituted by a bosonic bath $\mathcal{E}_{B}$ and a bath of TLSs $\mathcal{E}_{TLS}$. The coupling between the two baths and the system is mediated by the Hamiltonians $H_{\mathcal{S} - B}$ and $H_{\mathcal{S} - TLS}$, respectively.](image)
the system and the TLS bath can be approximated by

$$H_{S-\text{TLS}} = \sum_k g_k^{\text{TLS}} \left( d_k^c c + d_k^e c^\dagger \right).$$  \hfill (4)

with $g_k^{\text{TLS}} = \sqrt{2} j_k b_k^{\text{TLS}}$. On the other hand, if the TLSs mainly reside in their excited state ($J^g \approx +j$) the mapping can be written as $J^g \to j_k^g - d_k^g d_k^e$, $J^e \to d_k^e$, $J^k \to d_k$, leading to the following approximation for $H_{S-\text{TLS}}$

$$H_{S-\text{TLS}} = \sum_k g_k^{\text{TLS}} \left( d_k^e c^2 + d_k^e c^\dagger \right).$$ \hfill (5)

These two different forms of the HP mapping correspond to two different physical situations: in the former case, the TLSs prevalently reside in their ground state corresponding to the idea that the impurities mainly reside in their ground state, implying a low-temperature regime. In this case, the bosonic excitations described by the operators $d_k$, represent (weak) excitations around the ground state. On the other hand, the latter case corresponds to the situation in which the highest-excited (metastable) state of the TLSs is weakly (de-)excited: corresponding, for instance, to the case in which an external drive induces excitations in the TLSs bath, leading to a possible interpretation of the linewidth narrowing observed in circuit QED setups under strong driving conditions \cite{ appendix B} in terms of nonlinear QLEs associated with the saturation of the TLSs. In this picture, the external drive effectively heats the impurities to their excited state, inducing the population inversion for the ensemble of TLSs and a consequent saturation, justifying the $HP_+$ transformation in terms of (weak) de-excitations of the highest-excited state.

As we show in Appendix \cite{ appendix B}, it is possible to derive QLEs for the system, provided that the environment Hamiltonian is described by a set of bosonic operators, coupled linearly to the system degrees of freedom. It is important to note that the requirement of linearity concerning the system/environment Hamiltonian is limited to the bath degrees of freedom, meaning that its most general form can be expressed as

$$H_{S-E} = \sum_k g_k \left[ F^\dagger (c, c^\dagger) e_k + F (c, c^\dagger) e_k^\dagger \right],$$ \hfill (6)

where $e_k$ and $e_k^\dagger$ represent generic bosonic operators associated with the environment degrees of freedom. The form the system/environment coupling represents a sufficient condition for the derivation of a nonlinear QLE, along with the assumption that the modes of the bath are noninteracting. In other terms, it is necessary to assume a linear dependence of the coupling Hamiltonian on the environment degrees of freedom since, in order to derive the QLEs for the system, the solution of the Heisenberg equation of motion for the environment degrees of freedom has to assume a specific form, in which the contribution of the system and the environment operators can be represented as two separate additive terms (see Appendix \cite{ appendix B}).

It is therefore clear that, since the form of $H_{S-B}$ and $H_{S-\text{TLS}}$ can be expressed in the form given by Eq. (6), with $F (c, c^\dagger)$ given by $c, c^2$ and $c^\dagger$, and with $c_k = b_k$ and $c_k = d_k$ for $S - B$, $S - \text{HP}_- \ S - \text{HP}_+$, respectively, we can write the dynamics of the system in terms of a (nonlinear) QLE as

$$\dot{c} = -i [c, H_S] - \frac{\kappa}{2} + \kappa_N c^\dagger c + \sqrt{\kappa_c n} + 2 \sqrt{\kappa c n} \tilde{e}_\text{TLS}^\dagger \quad (7a)$$

$$\dot{c} = -i [c, H_S] - \frac{\kappa}{2} - \kappa_N c^\dagger c + \sqrt{\kappa c n} + 2 \sqrt{\kappa c n} \tilde{e}_\text{TLS} \quad (7b)$$

Eqs. (7a, 7b), obtained considering the system/environment coupling given by $H_{S-\text{HP}_-}$ and $H_{S-\text{HP}_+}$, respectively, are the main result of our analysis: the presence of a TLS bath leads to the appearance of nonlinear dissipative terms $(\pm \kappa_N c^\dagger c)$, and to purely imaginary parametric noise terms $(2 \sqrt{\kappa c n} \tilde{e}_\text{TLS}(t))$. We stress here that these terms are the direct result of the modelling of the bath in terms of two-separate environments ($H_{S-B}$ and $H_{S-\text{HP}_+}$), and do not represent an ad-hoc modification of the linear QLEs that can be derived in the absence of coupling to TLSs. In particular, while the nonlinear dissipation term possibly represents a natural extension to the nonlinear regime of linear QLEs, the parametric noise term is a nontrivial contribution associated with the presence of the TLS bath.

In addition we observe here that, analogously to their linear counterpart, Eqs. (7a, 7b) are time-local, i.e. the dynamics is Markovian. As detailed in Appendix \cite{ appendix B} this property is related to the assumption that, within the range of frequencies of interest, the coupling strength between system and environment is independent of the mode considered (wide band limit approximation) \cite{ appendix B}.

If we further consider a pump-probe representative of a circuit QED setup (e.g. a circuit optomechanical experiment), we can assume that the dynamics given by Eq. (6) is linearized around a strong coherent tone

$$\alpha_p = \alpha_{in} \exp \left[-i \omega_p t \right].$$

The frequency $\omega_p$ is detuned by $\Delta = \omega_p - \omega_c$ from the cavity resonant frequency. As a result of the linearization scheme, we have that the amplitude of the cavity field oscillating at $\omega_p$ is given by the solution of a nonlinear algebraic equation. In Fig. \cite{ appendix B} we have plotted the stationary value of the cavity field for the two choices of the HP mapping ($\text{HP}_+$). As expected, for small values of the driving field $\alpha_{in}$, the stationary solution corresponds to the solution in the absence of nonlinear dissipation. However, for larger values of $\alpha_{in}$ the stationary solution
substantially deviates from the solution of the linear system, with, for the parameters discussed here, a negligible difference between HP_− cases.

Furthermore, the (first-order) dynamics of the fluctuations c = α + a around the stationary value induced by the pump (in a frame rotating at ω_p) is given by

\begin{align}
\dot{a} &= \left[i\Delta - \frac{K}{2} - 2\kappa_N |a|^2\right] a - \kappa_N \alpha^2 a^\dagger + \sqrt{\kappa_a} \alpha + 2\sqrt{\kappa_N} \alpha^\ast a_{\text{TLS}}^\dagger \quad (8a) \\
\dot{\alpha} &= \left[i\Delta - \frac{K}{2} - 2\kappa_N |\alpha|^2\right] \alpha + \kappa_N \alpha^2 a^\dagger + \sqrt{\kappa_a} \alpha + 2\sqrt{\kappa_N} \alpha^\ast a_{\text{TLS}}^\dagger \quad (8b)
\end{align}

HP− and HP+ case respectively (see Appendix C). It is possible to see that Eqs. (8a,8b) include a purely imaginary parametric term, on top of a nonlinear dissipation term implying linewidth broadening or narrowing, depending on the state of the TLSs bath. Recently, in Ref. [22] a term of the same form was introduced as an ad-hoc parameter, in order to match the experimental results of a cavity optomechanical experiment aimed at establishing squeezing below the SQL of a nanomechanical resonator.

Our description, therefore provides a potential explanation of such parametric effects in terms of nonlinear dissipation phenomena associated with the nonlinear coupling to a bath of TLSs. In order to characterize the effect induced by the presence of the nonlinear coupling to TLSs, we evaluate the fluctuation spectrum of the cavity field $S_\theta^0 = 1/2\langle \{X_\theta^0, X_0^\omega\} \rangle$ with $X_\theta^0 = 1/\sqrt{2} \left( a_{\text{TLS}}^\dagger e^{i\theta} + a_{\text{TLS}} e^{-i\theta} \right)$. Assuming thermal fluctuations both for the bosonic and the TLS bath. As hinted by the structure of Eqs. (8a,8b), the presence of a parametric term induces squeezing, which can be experimentally observed by homodyne detection of the output field, in the cavity spectrum for both cases, as it is possible to see from Fig. 3 where it is possible to see how the cavity fluctuation spectrum exhibits a clear dependence on the phase $\theta$.

We have reported here how it is possible to deduce nonlinear QLEs for the dynamics of an open quantum system from a nonlinear system/environment coupling Hamiltonian. Moreover, we have discussed how an effective nonlinear system/environment coupling can emerge in the
presence of impurities modeled as TLSs. Ultimately, we have shown that the TLS-induced nonlinearities can represent a potential explanation for the imaginary para-

metric terms reported in Ref. \cite{22}. This work was supported by the Academy of Finland (Contract No. 275245).

Appendix A: Holstein-Primakoff transformation

We discuss here the Holstein-Primakoff realization allowing us to replace the spin operators \( J_z, J_\pm \) obeying the usual SU(2) commutation relations

\[
\left[ J_z^k, J_\pm^k \right] = ± J_\pm^k, \quad \left[ J_z^k, J_z^k \right] = 2 J_z^k, \tag{A1}
\]

with bosonic operators \( d_k, d_k^\dagger \), for which

\[
\left[ d_k, d_k^\dagger \right] = 1. \tag{A2}
\]

As discussed in the main text, in order to map the spin operators obeying Eq. (A1) with the bosonic operators \( d_k, d_k^\dagger \), we have two possibilities, depending on the physical situation we want to describe. If we assume that \( J_z^k ≥ -j_k \) –this choice is indicated in the main text as HP –, we can consider the following transformation

\[
J_z^k = \hat{n}_k - j_k, \quad J_+^k = d_k^\dagger \sqrt{2j_k - \hat{n}_k}, \quad J_-^k = \sqrt{2j_k - \hat{n}_k} d_k,
\]

where \( \hat{n}_k = d_k^\dagger d_k \). The operators \( J_z^k, J_\pm^k \) can be shown to fulfill the SU(2) commutation relations

\[
\left[ J_z^k, J_\pm^k \right] = \left[ \hat{n}_k, d_k^\dagger \right] \sqrt{2j_k - \hat{n}_k} = J_\pm^k, \quad \left[ J_\pm^k, J_\pm^k \right] = 2j_k - \hat{n}_k \left[ \hat{n}_k, d_k \right] = -J_z^k, \tag{A4a}
\]

\[
\left[ J_\pm^k, J_\mp^k \right] = d_k^\dagger \left( \sqrt{2j_k - \hat{n}_k} \right)^2 d_k - \sqrt{2j_k - \hat{n}_k} \hat{n}_k \sqrt{2j_k - \hat{n}_k} \hat{n}_k = \hat{n}_k (2j_k - \hat{n}_k + 1) - 2j_k + \hat{n}_k - \hat{n}_k (2j_k - \hat{n}_k) = 2J_z^k. \tag{A4b}
\]

In the limit \( j_k → \infty \), we have that

\[
\frac{J_\pm^k}{\sqrt{2j_k}} = d_k^\dagger \left( \frac{2j_k - \hat{n}_k}{2j_k} \right) = d_k^\dagger \left( 1 - \frac{\hat{n}_k}{4j_k} + \ldots \right) \simeq d_k^\dagger, \quad \frac{J_z^k}{\sqrt{2j}} \simeq d_k, \quad \frac{J_\pm^k}{\hat{n}_k} = 1 - \frac{\hat{n}_k}{j_k} \simeq -1. \tag{A5}
\]

Therefore the bosonic excitations described by \( d_k \) and \( d_k^\dagger \) correspond to (small) excitations around the \( J_z^k = -j_k \) state. Conversely we can write

\[
J_z^k = j_k - \hat{n}_k, \quad J_\pm^k = d_k^\dagger \sqrt{2j_k - \hat{n}_k}, \quad J_\pm^k = \sqrt{2j_k - \hat{n}_k} d_k, \tag{A6}
\]

so that when \( j_k → \infty \)

\[
\frac{J_\pm^k}{\sqrt{2j_k}} \simeq d_k, \quad \sqrt{\frac{2}{j_k}} \frac{J_\pm^k}{\sqrt{2j_k}} \simeq d_k^\dagger, \quad \frac{J_\pm^k}{j_k} = 1 - \frac{\hat{n}_k}{j_k} \simeq 1, \tag{A7}
\]

which correspond to the description of small fluctuations around the \( J_z^k = j \) state, indicated as HP + in main text.

Appendix B: QLE for \((c, c^\dagger)\)

We discuss here the form of the QLEs generated by a model for which, following the notation introduced in Eq. (1) of the main text, \( H_S \) is left unspecified, the environment is given by a set of noninteracting bosonic modes described by \( H_E = \sum_k \omega_k e_k^\dagger e_k \), where \( e_k \) (\( e_k^\dagger \)) are the annihilation (creation) operators associated with mode \( k \) and the system/environment coupling is given by the following Hamiltonian

\[
H_{S-E} = \sum_k g_k \left[ F(c, c^\dagger) e_k^\dagger + F^\dagger(c, c^\dagger) e_k \right], \tag{B1}
\]
where \( F(c, c^\dagger) \) is a generic function of the creation and annihilation operators of the system. Since \( H_{S-E} \) is a linear operator with respect to the degrees of freedom of the bath, and \( c_k^{(1)} \) commutes with \( H_S \), we can follow the same strategy employed for the derivation of the linear QLEs \(^{[11]}\) and write the equations of motion (EOM) for the bath field operators in the Heisenberg picture as
\[
\dot{c}_k(t) = -i\omega_k c_k(t) - ig_k F(c, c^\dagger). \tag{B2}
\]
Similarly, the EOM for the system can be written as
\[
\dot{c}(t) = i[H_S, c(t)] + i \sum_k g_k \left( [F, c]c_k^\dagger + [F^\dagger, c]c_k \right). \tag{B3}
\]
Equation (B2) can be solved in terms of an initial condition \( t_0 \), yielding
\[
c_k(t) = e^{-i\omega_k(t-t_0)} c_k(t_0) - ig_k \int_{t_0}^t e^{-i\omega_k(t-t')} F(c(t'), c^\dagger(t')) \, dt'. \tag{B4}
\]
By substituting Eq. (B4) and its Hermitian conjugate into Eq. (B3) we obtain
\[
\dot{c}(t) = i[H_S, c(t)] + i \sum_k g_k \left( [F, c] e^{i\omega_k(t-t_0)} c_k^\dagger(t_0) + ig_k \int_{t_0}^t e^{i\omega_k(t-t')} F^\dagger(t') \, dt' \right)
+ [F^\dagger, c] \left( e^{-i\omega_k(t-t_0)} c_k(t_0) - ig_k \int_{t_0}^t e^{-i\omega_k(t-t')} F(t') \, dt' \right).
\tag{B5}
\]
Like for the purely linear case, we introduce the density of states \( D = \delta \kappa / \partial \omega_k \) (supposing a continuum of states for the bath) and assume that, in the relevant frequency regime, \( g_k \) does not depend on the mode index \( k \). If we define
\[
g_k = \sqrt{\frac{\kappa}{2\pi D}}, \tag{B6}
\]
where \( \kappa \) a the mode-independent constant, we can write Eq. (B5) as
\[
\dot{c}(t) = i[H_S, c(t)] + i \sum_k \sqrt{\frac{\kappa}{2\pi D}} \left( [F, c] e^{i\omega_k(t-t_0)} c_k^\dagger(t_0) + ig_k \int_{t_0}^t \sqrt{\frac{\kappa}{2\pi D}} e^{i\omega_k(t-t')} F^\dagger(t') \, dt' \right)
+ [F^\dagger, c] \left( e^{-i\omega_k(t-t_0)} c_k(t_0) - ig_k \int_{t_0}^t \sqrt{\frac{\kappa}{2\pi D}} e^{-i\omega_k(t-t')} F(t') \, dt' \right).
\tag{B7}
\]
where we have defined \( c_{in}(t) \) as
\[
c_{in}(t) = -\frac{i}{\sqrt{2\pi D}} \sum_k e^{-i\omega_k(t-t_0)} c_k(t_0). \tag{B8}
\]
The definition introduced in Eq. (B6) corresponds to what, in the context of electronic transport is defined as “wide band limit approximation” and, allowing us to write the QLE given in Eq. (B7) in time-local form, can be considered equivalent to the Markov approximation \(^{[17]}\).

Let us focus on the case, discussed in the text, of two separate baths: a bosonic bath with operators \( b_k \) and a bath of TLSs with HP-transformed modes \( d_k \). We define two functions \( F_b \) and \( F_{TLS} \) of the system operators that couple to the bosonic and TLS baths, respectively. The QLE (B7) then reads
\[
\dot{c}(t) = i[H_S, c(t)] + \sqrt{\kappa} \left( [F_b, c] \left( -c_{in} - \frac{\sqrt{\kappa}}{2} F_b^\dagger \right) + [F_b^\dagger, c] \left( -c_{in} + \frac{\sqrt{\kappa}}{2} F_b \right) \right)
+ \sqrt{\kappa N} \left( [F_{TLS}, c] \left( -c_{in}^{TLS} - \frac{\sqrt{\kappa N}}{2} F_{TLS}^\dagger \right) + [F_{TLS}^\dagger, c] \left( -c_{in}^{TLS} + \frac{\sqrt{\kappa N}}{2} F_{TLS} \right) \right). \tag{B9}
\]
Assuming a linear coupling between the system and the bosonic bath and choosing the HP+ mapping for the TLSs, one obtains \( F_b = c \) and \( F_{TLS} = c^2 \). Substituting these into Eq. (B9) gives
\[
\dot{c} = i[H_S, c(t)] - \left( \frac{\kappa}{2} + \kappa N c^2 \right) + \sqrt{\kappa c_{in} + 2\sqrt{\kappa N} c_{in}^{TLS}}. \tag{B10}
\]
which corresponds to Eq. (7a) of the main text. On the contrary, if the HP+ mapping is chosen, one obtains Eq. (7b) with \( F_{TLS} = c^2 \).
Appendix C: Linearization of the quantum Langevin equations

Here we outline the linearization strategy that allows us, in the presence of a strong coherent tone $\alpha_p = \alpha_{in} e^{-i \omega_p t}$, to recast Eqs. (7a,7b) of the main text in terms of equations describing the stationary state (in a frame rotating at $\omega_p$) and the fluctuations around this stationary state, given by Eqs. (8a,8b) of the main text.

Focusing on Eq. (7a)

$$\dot{c} = -i [c, H_S] - \left( \frac{\kappa}{2} + \kappa_N c^\dagger c \right) c + \sqrt{\kappa} c_{in} + 2 \sqrt{\kappa N} c_{TLS}$$

(C1)
in the presence of a strong coherent pump $\alpha_p = \alpha_{in} e^{-i \omega_p t}$, we seek a solution of the form $c = \alpha + a$

$$-i \omega_p \alpha + \dot{a} = -i \omega_c (\alpha + a) - \left( \frac{\kappa}{2} + \kappa_N (\alpha^* + a^\dagger) (\alpha + a) \right) (\alpha + a) + \sqrt{\kappa} (\alpha_{in} + a_{in}) + 2 \sqrt{\kappa N} (\alpha^* + a^\dagger) a_{in}^{TLS},$$

(C2)

where without loss of generality, we have assumed that $H_S = \omega_c c^\dagger c$.

Neglecting the fluctuation terms, we obtain the equation for the steady-state solution

$$0 = i \Delta \alpha - \kappa \alpha - \kappa_N |\alpha|^2 + \sqrt{\kappa} \alpha_{in},$$

(C3)

where $\Delta = \omega_p - \omega_c$. From Eq. (C2) the equation for the fluctuation around the steady-state solution value of $\alpha$ given above is thus expressed as

$$\dot{a} = \left[ i \Delta - \left( \frac{\kappa}{2} + 2 \kappa_N |\alpha|^2 \right) \right] a - \kappa \alpha^2 a^\dagger + \sqrt{\kappa} a_{in} + 2 \sqrt{\kappa N} \alpha^* a_{in}^{TLS}.$$  

(C4)

With a similar procedure one can also show that Eq. (7b) leads to Eq. (8b). Notice that the nonlinear dissipative terms $\mp 2 \kappa_N |\alpha|^2 a$ in Eqs. (8a,8b) lead to the broadening/narrowing of the linewidth associated with the linearized response of the cavity field fluctuations, respectively.

FIG. 4. The total effective dissipation of the linearized models Eq. (8a) (red) and Eq. (8b) (dashed green) that correspond to the cases, where the majority of the TLSs are in the ground state/excited state, respectively. They are compared to the case of pure linear dissipation (black dots). Here we assume the system to be a simple cavity with $H_S = \omega_c c^\dagger c$. In the units of $\kappa$, the parameters are $\Delta = \omega_p - \omega_c = 20$ and $\kappa_N = 1.5 \times 10^{-4}$.

Appendix D: Fluctuation spectrum of the nonlinear model

Assuming that, in addition to the strong coherent tone, the dynamics of the system is affected by thermal fluctuations of both the bosonic and the TLS baths degrees of freedom, we evaluate here the spectrum of these fluctuations focusing
on the HP− case (an analogous derivation holds for the HP+ mapping). The fluctuation spectrum

\[ S_\omega = \frac{1}{2} \langle \{ X_\omega, X_{-\omega}^\theta \} \rangle, \]  

with \( X_\omega^\theta = 1/\sqrt{2} \left( a_{-\omega}^\dagger e^{i\theta} + a_\omega e^{-i\theta} \right) \), can be obtained by Fourier transforming the QLE given by Eq. (8a) and its Hermitian conjugate

\[ \begin{aligned}
&\left[ -i (\omega + \Delta) + \frac{\kappa}{2} + 2\kappa_N |\alpha|^2 \right] a_\omega + \kappa_N \alpha^2 a_{-\omega}^\dagger = \sqrt{\kappa} a_{\text{in},\omega} + 2\sqrt{\kappa_N} \alpha^* a_{\text{TLS}}^\dagger \ 
&\left[ -i (\omega - \Delta) + \frac{\kappa}{2} + 2\kappa_N |\alpha|^2 \right] a_{-\omega}^\dagger + \kappa_N \alpha^2 a_\omega = \sqrt{\kappa} a_{\text{in},-\omega} + 2\sqrt{\kappa_N} \alpha a_{\text{TLS}} \end{aligned} \]  

with the usual convention for the Fourier transform, according to which \( a_t \overset{\text{FT}}{\mapsto} a_\omega \) and \( a_t^\dagger \overset{\text{FT}}{\mapsto} a_{-\omega}^\dagger \).

Defining

\[ A = -i (\omega + \Delta) + \frac{\kappa}{2} + 2\kappa_N |\alpha|^2, \]  

\[ B = \kappa N \alpha^2, \]  

\[ C = -i (\omega - \Delta) + \frac{\kappa}{2} + 2\kappa_N |\alpha|^2, \]

the QLE for the system can be expressed as

\[ \begin{pmatrix} a_\omega \\ a_{-\omega}^\dagger \end{pmatrix} = \frac{1}{AC - |B|^2} \begin{pmatrix} C & -B \\ -B^* & A \end{pmatrix} \begin{pmatrix} \sqrt{\kappa} a_{\text{in},\omega} + 2\sqrt{\kappa_N} \alpha^* a_{\text{TLS}}^\dagger \\ \sqrt{\kappa} a_{\text{in},-\omega} + 2\sqrt{\kappa_N} \alpha a_{\text{TLS}} \end{pmatrix} \]  

and

\[ a_\omega = \chi_d (\omega) a_{\text{in},\omega} + \chi_x (\omega) a_{\text{in},-\omega}^\dagger + \chi_d^{\text{TLS}} (\omega) a_{\text{TLS}}^\dagger a_{\text{in},\omega} + \chi_x^{\text{TLS}} (\omega) a_{\text{in},-\omega}, \]  

\[ a_{-\omega}^\dagger = \chi_x^* (-\omega) a_{\text{in},\omega}^\dagger + \chi_d^* (-\omega) a_{\text{in},-\omega} + \chi_x^{\text{TLS}*} (-\omega) a_{\text{TLS}} + \chi_d^{\text{TLS*}} (-\omega) a_{\text{TLS}}^\dagger \]

where

\[ \chi_d (\omega) = \sqrt{\kappa} C (AC - |B|^2)^{-1}, \]  

\[ \chi_x (\omega) = -\sqrt{\kappa} B (AC - |B|^2)^{-1}, \]  

\[ \chi_d^{\text{TLS}} (\omega) = 2\sqrt{\kappa_N} \alpha^* C (AC - |B|^2)^{-1}, \]  

\[ \chi_x^{\text{TLS}} (\omega) = -2\sqrt{\kappa_N} \alpha B (AC - |B|^2)^{-1}. \]

If we assume that the thermal populations of the baths are given by \( \langle a_{\text{in},\omega} a_{\text{in},\omega}^\dagger \rangle = (n_{\text{th}} + 1) \delta (\omega - \omega') \) and
becomes narrower. Here the thermal populations of the bosonic and TLS baths are

\[ n = \frac{1}{2} \left( 1 + e^{\frac{\pi}{2}} \right) \]

\( n_{\text{th}} = n_{\text{th}}^\dagger = 1, \)

and the spectrum related to HP\(^+\) coupling derived from Eq. (8b) is presented in Fig. 5(b).

\[ \left\langle a_{\text{TLS}} a_{\text{TLS}}^\dagger \right\rangle = (n_{\text{TLS}} + 1) \delta (\omega - \omega'), \]

the cavity spectrum can be written as

\[ S_\omega = \frac{1}{4} \left[ \left| \chi_d (\omega) \right|^2 + \left| \chi_x (\omega) \right|^2 \right] \left\langle \left\{ a_{\text{TLS}}, a_{\text{TLS}}^\dagger \right\} \right\rangle + \left( \left| \chi_d (-\omega) \right|^2 + \left| \chi_x (-\omega) \right|^2 \right) \left\langle \left\{ a_{\text{TLS}}^\dagger, a_{\text{TLS}} \right\} \right\rangle \]

\[ + \frac{1}{4} \left[ \left| \chi_d (\omega) \chi_x (-\omega) e^{-i2\theta} + \chi_d^* (\omega) \chi_x^* (-\omega) e^{i2\theta} \right| \left\langle \left\{ a_{\text{TLS}}, a_{\text{TLS}}^\dagger \right\} \right\rangle + \left| \chi_d (-\omega) \chi_x (\omega) e^{-i2\theta} + \chi_d^* (-\omega) \chi_x^* (\omega) e^{i2\theta} \right| \left\langle \left\{ a_{\text{TLS}}^\dagger, a_{\text{TLS}} \right\} \right\rangle \right] \]

\[ + \frac{1}{4} \left[ \left| \chi_d (\omega) \chi_x^T (\omega) e^{-i2\theta} + \chi_d^T (\omega) \chi_x (\omega) e^{i2\theta} \right| \left\langle \left\{ a_{\text{TLS}}^\dagger, a_{\text{TLS}} \right\} \right\rangle + \left| \chi_d (-\omega) \chi_x^T (-\omega) e^{-i2\theta} + \chi_d^T (-\omega) \chi_x (-\omega) e^{i2\theta} \right| \left\langle \left\{ a_{\text{TLS}}, a_{\text{TLS}}^\dagger \right\} \right\rangle \right] \]

\[ = \frac{1}{2} \left[ \left| \chi_d (\omega) \right|^2 + \left| \chi_d (-\omega) \right|^2 + \left| \chi_x (\omega) \right|^2 + \left| \chi_x (-\omega) \right|^2 \right] \left( n_{\text{th}} + \frac{1}{2} \right) \]

\[ + \frac{1}{2} \left[ \left| \chi_d (\omega) \chi_x (-\omega) + \chi_d (-\omega) \chi_x (\omega) \right| \left( n_{\text{th}} + \frac{1}{2} \right) \right] \]

\[ + \frac{1}{2} \left( 1 + e^{\frac{\pi}{2}} \right) \left| \chi_d (\omega) \chi_x^T (\omega) + \chi_d^T (\omega) \chi_x (\omega) \right| \left( n_{\text{th}} + \frac{1}{2} \right) \]

where \( \phi_{\text{TLS}} = \arg \left( \chi_d^T (\omega) \chi_x (\omega) + \chi_d (\omega) \chi_x^T (\omega) \right). \)

In Fig. 5(a) we have plotted the cavity spectrum for the HP\(^-\), and the spectrum related to HP\(^+\) coupling derived from Eq. (8b) is presented in Fig. 5(b).

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