

Supplementary Materials

I. HAMILTONIANS OF VARIOUS SYSTEM-ENVIRONMENT COUPLINGS

A general open quantum system is defined as a principal system interacting with its environment (reservoirs) through the exchange of matter, energy and information between them [1]. A general Hamiltonian describing the coupling between the system and its environment can be formally expressed as

$$H_{SB} = \sum_{ik} (V_{ik} A_i^\dagger B_k + \text{H.c.}), \quad (1)$$

where the V_{ik} are coupling strengths, and A_i^\dagger and B_k are arbitrary operators acting on the system and the environment, respectively. The explicit form of H_{SB} is determined by the particular system and its environment that one is concerned.

Consider first the exchange of matter between the system and the environment. Since matter is built with fermions (electrons and nuclei or, more fundamentally, quarks), we assume that both the system and its environment are made of fermions. Let a_i, a_i^\dagger and b_k, b_k^\dagger denote the creation and annihilation operators of fermions, which satisfy the standard fermionic anticommutation relationships. Then the most basic process for the underlying matter exchange between the system and its environment can be modelled through the following Hamiltonian:

$$H_{SB} = \sum_{ik} (V_{ik} a_i^\dagger b_k + V_{ik}^* b_k^\dagger a_i). \quad (2)$$

This Hamiltonian is indeed the typical one describing various electron transport phenomena in nanoelectronics and spintronics for mesoscopic physics [2]. The above Hamiltonian for the exchange of matter between the system and its environment can also be applied to the case where both the system and the environment are made of bosons as composite particles of fermions. Correspondingly, the creation and annihilation operators, a_i, a_i^\dagger and b_k, b_k^\dagger , would then obey bosonic commutation relationships. If the system and its environment are made of massless particles, such as photons, the Hamiltonian in Eq. (2) would describe the photon-photon exchanges (corresponding to energy exchanges only) between the system and the environment [3, 4]. Accompanying the exchange of matter and/or energy between the system and its environment, there is also information exchange through the coupling Hamiltonian (2).

The next order contribution to the exchange of matter and energy, between the system and its environment, originates from particle-particle interactions. Correspondingly, the Hamiltonian for the coupled system-environment through particle-particle interactions can be expressed as

$$H'_{SB} = \sum_{ijkl} (V_{ijkl}^{(1)} a_i^\dagger a_j^\dagger b_l b_k + V_{ijkl}^{(2)} a_i^\dagger b_l^\dagger b_k a_j + \text{H.c.}), \quad (3)$$

where the first term plus its hermitian conjugate correspond to particle-pair exchanges between the system and its environment, and the second term only involves energy exchanges (particle-particle scatterings) between them. Including the interacting Hamiltonian of Eq. (3) makes the system and its environment become a typical interacting many-body problem. Except for weak particle-particle interactions, where the perturbation approach can be applied, the problem with Eq. (3) becomes unsolvable in general, just like strongly-correlated electronic systems in condensed matter physics and low-energy quantum chromodynamics in particle physics. Since non-Markovian dynamics is manifested significantly when the system strongly couples to its environment, we should not start with these systems whose dynamics can only be partially solved by the use of perturbation expansions.

The above discussions focus on open quantum systems where both the system and its environment are made of the same type of particles, either fermions or bosons. However, we also often have the case where the system is a fermionic system and its environment is made of bosons. A typical example of this type is a system coupled to its environment through matter-light interaction, where the system-environment coupling is determined by electron-photon interactions under the fundamental theory of quantum electrodynamics,

$$H_{e-p} = -e \int d^3x \psi^\dagger(x) \gamma_\mu \psi(x) A^\mu(x) = \sum_{pq} (V_{pq} a_{p+q}^\dagger a_p b_q + V_{pq}^* a_p^\dagger a_{p+q} b_q^\dagger), \quad (4)$$

where $\psi(x)$ is the electron field, $A^\mu(x)$ is the vector potential of the electromagnetic field, e is the electron charge, and the γ_μ are the Dirac γ -matrices [5]. In the second equality of Eq. (4), we have ignored the antiparticle component

(positron) of the electron field because it has no contribution in the nonrelativistic regime. A two-level system coupled to an electromagnetic field can in principle be derived from (4), and the electron-phonon coupling between the system and its environment has a similar coupling Hamiltonian. The dynamics of the system with such electron-photon interaction has also only been solved within the approach of perturbation expansions, and therefore it is also not our main concern in this work.

As a conclusion, in the main text of this work, we concentrate on the dynamics of open quantum systems with the Hamiltonian of the system-environment couplings, Eq. (2), which describes the underlying exchanges of matter and/or energy between the system and its environment, and can be nonperturbatively solved with the recently developed exact master equations [3, 6].

II. SPECTRAL DENSITIES FOR OPEN QUANTUM SYSTEMS

As we have shown in the main text of this work, the environment may consist of many different reservoirs, and each reservoir and its coupling to the system are fully characterized by the spectral density matrix $J_{ij}(\omega)$:

$$J_{ij}(\omega) \equiv 2\pi \sum_k V_{ik} V_{jk}^* \delta(\omega - \varepsilon_k) = 2\pi \varrho(\omega) V_i(\omega) V_j^*(\omega), \quad (5)$$

where $\varrho(\omega)$ is the density of states of the reservoir, and $V_i(\omega)$ is the system-reservoir coupling constant in the frequency domain. For most reservoirs, its density of states is unknown, and in principle, one may extract the density of states for each reservoir either from experiments or from numerical calculations. However, the complete information about the effect of the environment on the system is encapsulated in the spectral density $J(\omega)$ [7], neither the density of states of the reservoir nor the system-reservoir coupling strength alone. To explore the general dynamics of open quantum systems, it is particularly useful to model the spectral density from *a priori* microscopic knowledge. For thermal bosonic (photon and phonon) reservoirs, Leggett *et al.* [7] introduced the most general spectral density as a smooth function of ω with a form of ω^s up to some frequency cutoff ω_c :

$$J(\omega) = 2\pi\eta\omega \left(\frac{\omega}{\omega_c}\right)^{s-1} \exp\left(-\frac{\omega}{\omega_c}\right), \quad (6)$$

where $s = 1$ corresponds to the normal Ohmic dissipation of the damped dynamics; $s > 1$ corresponds to the super-Ohmic dissipation, in particular, $s = 3$ and $s = 5$ have been used to characterize a defect in a three-dimensional solid interacting with acoustic phonons; $s < 1$ denotes the sub-Ohmic dissipation, where the low-frequency noise plays the dominant role. The spectral density (6) covers a large class of bosonic thermal reservoirs widely used in various investigations of open quantum systems. In the main text of this work, we present for the first time the exact analytical solution of non-Markovian dissipation dynamics for this general spectral density.

For fermionic reservoirs, in particular for electronic nanostructures, the most complicated spectral density commonly used is the Lorentzian-type spectral density [6, 8]

$$J(\omega) = \frac{\Gamma d^2}{(\omega - \omega_c)^2 + d^2}. \quad (7)$$

This is also a very popular spectral density that has often been used in the studies of non-Markovian dynamics for a two-level system coupled to a photonic reservoir [9–11]. In mesoscopic systems, one usually takes the very special limit of (7), namely the wide-band limit ($d \rightarrow \infty$) to analyze various quantum transport and quantum decoherence phenomena [2]. In our study, we take this spectral density with an explicit cutoff to simulate the band structure in electronic nanostructures, see Eq. (14) in the main text. There is another type of reservoir, called structured reservoirs, that has recently attracted a considerable attention in the current study of nanophotonics and nanoelectronics. A typical example is photonic crystals. The spectral density used to study quantum optics in photonic crystals, originally introduced by S. John and other authors [12], is discussed in the third example of the main text, as a very special case. In the main text, we have not only presented the general analytical structure of non-Markovian dynamics for open quantum systems with the basic system-environment coupling Hamiltonian (2), but also provided the explicit solutions for various spectral densities that are commonly used in investigations of open quantum systems.

III. EQUILIBRIUM FLUCTUATION-DISSIPATION THEOREM AND BORN-MARKOVIAN MASTER EQUATION

To see clearly how the nonequilibrium fluctuation-dissipation theorem, given in the main text of this work, is reduced to the equilibrium fluctuation-dissipation theorem, we should derive the Born-Markovian master equation

from the exact master equation, using a perturbation approximation up to second order in the coupling between the system and the environment, where the environment is considered to be in equilibrium. To be explicit, we consider a single-level system weakly coupled to a reservoir. The Dyson equation of the Green function $u(t, t_0)$ can be rewritten as

$$\dot{u}(t, t_0)u^{-1}(t, t_0) = -i\epsilon_s - \int_{t_0}^t d\tau g(t, \tau)u(\tau, t_0)u^{-1}(t, t_0) \quad (8)$$

Since the self-energy correlations $g(\tau, \tau') \sim J(\omega) \sim \sum_k |V_k|^2$, taking a perturbation expansion up to second-order on the coupling V_k means that the Green functions $u(\tau, t_0)$ and $u^{-1}(t, t_0)$ in the right hand side of Eq. (8) should be approximated only up to zero-order, i.e., $u_0(\tau, t_0) = \exp\{-i\epsilon_s(\tau - t_0)\}$ and $u_0^{-1}(t, t_0) = \exp\{i\epsilon_s(t - t_0)\}$. This leads to

$$\dot{u}(t, t_0)u^{-1}(t, t_0) \simeq -i\epsilon_s - \int_{t_0}^t d\tau \int \frac{d\omega}{2\pi} J(\omega) \exp\{-i(\omega - \epsilon_s)(t - \tau)\}, \quad (9)$$

Up to the same order in the perturbation expansion, Eq.(6) in the main text gives

$$\dot{v}(t, t) \simeq 2 \int_{t_0}^t d\tau \int \frac{d\omega}{2\pi} J(\omega) f(\omega) \cos[(\omega - \epsilon_s)(t - \tau)]. \quad (10)$$

Then the renormalized energy, as well as the dissipation and fluctuation coefficients in the exact master equation are reduced to

$$\epsilon'_s(t) = -\text{Im}[\dot{u}(t, t_0)u^{-1}(t, t_0)] \simeq \omega_c - \int_{t_0}^t d\tau \int \frac{d\omega}{2\pi} J(\omega) \sin[(\omega - \epsilon_s)(t - \tau)], \quad (11a)$$

$$\gamma(t) = -\text{Re}[\dot{u}(t, t_0)u^{-1}(t, t_0)] \simeq \int_{t_0}^t d\tau \int \frac{d\omega}{2\pi} J(\omega) \cos[(\omega - \epsilon_s)(t - \tau)], \quad (11b)$$

$$\tilde{\gamma}(t) \simeq \dot{v}(t, t) \simeq 2 \int_{t_0}^t d\tau \int \frac{d\omega}{2\pi} J(\omega) f(\omega) \cos[(\omega - \epsilon_s)(t - \tau)]. \quad (11c)$$

Substituting these coefficients into the master equation reproduces the master equation under the Born and Markov approximations [4]. Setting $t_0 = 0$ and taking the steady-state limit, $t \rightarrow \infty$, Equations (11b) and (11c) are the Fourier transform of the the dissipation and fluctuation coefficients, which give the fluctuation-dissipation theorem in the frequency domain in the second-quantization formalism [13]:

$$\tilde{\gamma}(\omega) = 2f(\omega)\gamma(\omega). \quad (12)$$

Note that in the second-quantization formalism, the fluctuation is defined in terms of $\langle a^\dagger(t)a(\tau) \rangle$. In usual quantum mechanics, the fluctuation is defined as $\chi(t, \tau) = \langle q(t)q(\tau) \rangle$, where $q(t) \sim [a^\dagger(t) + a(t)]/\sqrt{2}$. In equilibrium, $\langle q(t)q(\tau) \rangle = \langle a^\dagger(t)a(\tau) \rangle + 1/2$. Then we have

$$\tilde{\chi}(\omega) = 2[f(\omega) + 1/2]\gamma(\omega) = \coth\left(\frac{\beta\omega}{2}\right)\gamma(\omega), \quad (13)$$

where $f(\omega) = [e^{\beta\omega} - 1]^{-1}$ has been used for a bosonic bath. This is the standard quantum mechanical fluctuation-dissipation theorem.

It may be worth pointing out that the exact master equation of Eq. (2) in the main text can also be rewritten in terms of the standard Lindblad form:

$$\frac{d\rho(t)}{dt} = \frac{1}{i}[\tilde{H}_S(t), \rho(t)] + \sum_{ij} \tilde{\gamma}_{ij}(t)L_{a_i^\dagger, a_j}[\rho(t)] + \sum_{ij} [2\gamma_{ij}(t) \pm \tilde{\gamma}_{ij}(t)]L_{a_j, a_i^\dagger}[\rho(t)], \quad (14a)$$

$$L_{a_i, a_j^\dagger}[\rho(t)] \equiv a_i\rho(t)a_j^\dagger - \frac{1}{2}a_j^\dagger a_i\rho(t) - \frac{1}{2}\rho(t)a_j^\dagger a_i, \quad (14b)$$

where the $+/-$ signs correspond to boson/fermion systems. However, the two non-unitary terms in the exact master equation given in the main text describe the dissipation and fluctuation separately, and the dissipation and fluctuation dynamics are connected by the intrinsic fluctuation-dissipation theorem. The above Lindblad form of the exact master equation has an excellent symmetric superoperator form but it mixes the dissipation and fluctuation such that the fluctuation-dissipation theorem is not manifested. Therefore when the coefficients in the Lindblad master equation cannot be derived microscopically, one has to add additional constraints on these coefficients to ensure the complete positivity of the reduced density matrix during the time evolution.

IV. DERIVATION OF THE ANALYTICAL SOLUTION OF THE GREEN FUNCTION $u(t, t_0)$

Here we present a detailed derivation of the Green function $u(t, t_0)$ for the second example of the application given in the main text, i.e. the single-electron transistor. There, the source and the drain are treated as the environment with the modified Lorentzian spectral density given by Eq. (14),

We solve the Dyson equation for the Green function $u(t, t_0)$ by using the Laplace transformation $F(s) = \int_{t_0}^{\infty} dt f(t) \exp\{-s(t - t_0)\}$. Here it is more convenient to use the modified Laplace transformation and its inverse by replacing the variable s by $-iz$,

$$F(z) = \tilde{\mathcal{L}}\{f(t)\} \equiv \int_{t_0}^{\infty} dt f(t) \exp\{iz(t - t_0)\}, \quad (15a)$$

$$f(t) = \tilde{\mathcal{L}}^{-1}\{F(z)\} \equiv \frac{1}{2\pi} \int_{-\infty+i\lambda}^{\infty+i\lambda} dz F(z) \exp\{-iz(t - t_0)\}. \quad (15b)$$

Eq. (15b) is the modified Bromwich integral, where λ is an arbitrary positive number greater than the imaginary part of all the poles of $F(z)$. The result is independent of λ . Applying the modified Laplace transformation to the Dyson equation, we obtain

$$U(z) = \frac{i}{z - \omega_c - \Sigma(z)}, \quad (16)$$

where $\Sigma(z)$ is the Laplace transformation of the self-energy

$$\begin{aligned} \Sigma(z) &= -i \int_0^{\infty} d\tau g(\tau) e^{iz\tau} = 2 \int_{\omega_c - \Omega}^{\omega_c + \Omega} \frac{d\omega}{2\pi} \frac{\Gamma d^2}{(\omega - \omega_c)^2 + d^2} \cdot \frac{1}{z - \omega} \\ &= \frac{1}{\pi} \left[\frac{\Gamma d^2}{(z - \omega_c)^2 + d^2} \log \left(\frac{\omega_c - \Omega - z}{\omega_c + \Omega - z} \right) + \frac{2(z - \omega_c)\Gamma d}{(z - \omega_c)^2 + d^2} \tan^{-1} \left(\frac{\Omega}{d} \right) \right]. \end{aligned} \quad (17)$$

For $\Omega \rightarrow \infty$, and $\text{Im}\{z\} > 0$, we have $\tan^{-1}(\Omega/d) \rightarrow \pi/2$ and $\log(\omega_c - \Omega - z) - \log(\omega_c + \Omega - z) \rightarrow -\pi i$. Then Eq. (17) is reduced to

$$\Sigma(z) = \frac{\Gamma d}{z - \omega_c + id} \quad \text{for } \Omega \rightarrow \infty, \quad (18)$$

which is the result of the usual Lorentzian spectral density without cut-off, except for a factor 2 coming from the two symmetric reservoirs, the source and the drain.

Due to the cutoff (band structure), for $\omega_c - \Omega < z < \omega_c + \Omega$, we have

$$\lim_{\epsilon \rightarrow 0^+} \Sigma(z \pm i\epsilon) = \Delta(z) \mp iJ(z) \quad (19)$$

with

$$\Delta(z) = \frac{1}{\pi} \left[\frac{\Gamma d^2}{(z - \omega_c)^2 + d^2} \log \frac{z - (\omega_c - \Omega)}{(\omega_c + \Omega) - z} + \frac{2(z - \omega_c)\Gamma d}{(z - \omega_c)^2 + d^2} \tan^{-1} \frac{\Omega}{d} \right]. \quad (20)$$

Thus, the analytic solution for the Green function $u(t - t_0)$ is

$$u(t - t_0) = \sum_j \mathcal{Z}_j \exp\{-i\omega'_j(t - t_0)\} + \frac{1}{\pi} \int_{\omega_c - \Omega}^{\omega_c + \Omega} d\omega \frac{J(\omega)}{[\omega - \varepsilon_s - \Delta(\omega)]^2 + J^2(\omega)} \exp\{-i\omega(t - t_0)\}. \quad (21)$$

The localized bound modes are determined by

$$\omega'_j - \varepsilon_s = \Sigma(\omega'_j) \quad (22)$$

Note that $\Sigma(\omega)$ is monotonically decreasing for $|\omega - \omega_c| > \Omega$ (outside the band), and its boundary values are $\Sigma(\omega_c \pm \Omega) = \pm\infty$ and $\Sigma(\pm\infty) = 0$. Thus, there must exist two poles. One pole is located in $(\omega_c + \Omega, \infty)$ and the other one in $(-\infty, \omega_c - \Omega)$. The corresponding residue is given by

$$\mathcal{Z}_j = \frac{1}{1 - \Sigma'(\omega'_j)}. \quad (23)$$

Equations (20)-(23) are some of the results presented in the main text.

To reproduce the solution of the usual Lorentzian spectral density, let $\Omega \rightarrow \infty$. Then the localized bound modes vanish (they can only exist in the region with $J(z) = 0$), and Eq. (20) becomes

$$\Delta(z) = \frac{(z - \omega_c)\Gamma d}{(z - \omega_c)^2 + d^2} = \frac{z - \omega_c}{d} J(z) \quad (24)$$

and the solution (21) is reduced to

$$\begin{aligned} u(t - t_0) &= \frac{1}{\pi} \int_{-\infty}^{\infty} dz \frac{\Gamma d^2}{[(z - \varepsilon_s)(z - \omega_c) - \Gamma d]^2 + (z - \varepsilon_s)^2 d^2} \exp\{-iz(t - t_0)\} \\ &= \frac{1}{\pi} \int_{-\infty}^{\infty} dz \left[\frac{i(z - \omega_c + id)}{(z - \varepsilon_s)(z - \omega_c) - \Gamma d + i(z - \varepsilon_s)d} - \frac{i(z - \omega_c - id)}{(z - \varepsilon_s)(z - \omega_c) - \Gamma d - i(z - \varepsilon_s)d} \right] e^{-iz(t-t_0)}. \end{aligned} \quad (25)$$

The first term corresponds exactly to the inverse Laplace transform of $U(z)$ with the self-energy of (18). Thus, equation (25) apparently gives a different solution from taking $\Omega \rightarrow \infty$ at beginning. However, note that each term in Eq. (25) has two poles. The two poles of the first term are located in the lower-half complex plane,

$$z_{\pm} = \frac{\varepsilon_s + \omega_c - id \pm \omega_{\delta}}{2} \quad (26)$$

with $\omega_{\delta} = \sqrt{(\delta + id)^2 + 4\Gamma d}$, and $\delta = \varepsilon_s - \omega_c$. The two poles of the second term, which are the complex conjugates of (26), are located in the upper-half plane. Because the integrand is divergent on the infinity boundary of the upper-half plane, we must take the contour integral along the lower-half plane, which gives the solution of $u(t, t_0)$ by

$$\begin{aligned} u(t - t_0) &= \frac{1}{\pi} \int_{-\infty}^{\infty} dz \frac{i(z - \omega_c + id)}{(z - \varepsilon_s)(z - \omega_c) - \Gamma d + i(z - \varepsilon_s)d} e^{-iz(t-t_0)} \\ &= \exp\left\{-\frac{1}{2} [i(\varepsilon_s + \omega_c) + d] (t - t_0)\right\} \left\{ \frac{\delta + \omega_{\delta} + id}{2\omega_{\delta}} e^{-\frac{i}{2}\omega_{\delta}(t-t_0)} - \frac{\delta - \omega_{\delta} + id}{2\omega_{\delta}} e^{\frac{i}{2}\omega_{\delta}(t-t_0)} \right\}. \end{aligned} \quad (27)$$

This provides the consistency for the results with and without having an explicit cutoff. For the resonance, $\delta = \varepsilon_s - \omega_c = 0$, the above solution is reduced to

$$u(t - t_0) = \exp\left\{-\left(i\varepsilon + \frac{d}{2}\right)(t - t_0)\right\} \times \begin{cases} \left[\cosh \frac{\varepsilon_{\Gamma}}{2}(t - t_0) + \frac{d}{\varepsilon_{\Gamma}} \sinh \frac{\varepsilon_{\Gamma}}{2}(t - t_0)\right], & d > 4\Gamma \\ [1 + \Gamma(t - t_0)], & d = 4\Gamma \\ \left[\cos \frac{\varepsilon_{\Gamma}}{2}(t - t_0) + \frac{d}{\varepsilon_{\Gamma}} \sin \frac{\varepsilon_{\Gamma}}{2}(t - t_0)\right], & d < 4\Gamma \end{cases} \quad (28)$$

with $\varepsilon_{\Gamma} = \sqrt{d|d - 4\Gamma|}$. By replacing $2\Gamma \rightarrow \Gamma$ for a single reservoir, the solution (27) recovers the result given by Kofman *et al.* in [9], and the resonant solutions (28) also reproduce the results for the usual Lorentzian spectral density that have been used in the studies of non-Markovian dynamics [10, 11]. It shows that when the spectral density is broad enough (here the half-bandwidth of the spectral density is greater than twice of its height, $d > 4\Gamma$), the Green function $u(t, t_0)$ gives only simple exponential decays. Otherwise, the exponential decays are modified with oscillations (sort of non-exponential decays). The resulting dissipation coefficient varies sequentially between positive and negative values. However, when the spectral density contains a band gap, the localized bound modes significantly change the dissipation behavior, and additional non-Markovian dynamics shows up, as we have shown in the main text.

Applying the same procedure to the Ohmic-type spectral density and the spectral density of the photonic crystals gives the general analytical solutions of other examples in the main text, and we should not repeat these detailed derivation here.

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