Heat distribution in quantum Brownian motion

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We investigate the heat statistics in a relaxation process of quantum Brownian motion described by the Caldeira-Leggett model. By employing the normal mode transformation and the phase-space formulation approach, we can analyze the quantum heat distribution within an exactly dynamical framework beyond the traditional paradigm of Born-Markovian and weak-coupling approximations. It is revealed that the exchange fluctuation theorem for quantum heat generally breaks down in the strongly non-Markovian regime. Our results may improve the understanding about the nonequilibrium thermodynamics of open quantum systems when the usual Markovian treatment is no longer appropriate.

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I. INTRODUCTION

In recent years, much attention has been paid to investigate the thermodynamical character of a small system out of equilibrium [1–7]. In sharp contrast to the equilibrium thermodynamics in classical physics, thermodynamic quantities, such as work, heat, and entropy, in the nonequilibrium thermodynamics commonly exhibit statistical stochasticity and satisfy certain fluctuation theorems [8–17]. Some of them, for example, the celebrated Jarzynski equality for work [8] and the exchange fluctuation theorem (EFT) for heat [12], have been verified in several recent experiments [18–24]. These fluctuation theorems are of fundamental significance in quantum thermodynamics because they can provide insight for us to reexamine the laws of thermodynamics, which were originally established in classical equilibrium thermodynamics at the microscopic level [25–29]. On the other hand, due to the fact that fluctuations are commonly nonnegligible in microscopic engines [30–33], the investigation of these fluctuation theorems may aid in the development of quantum heat machines beyond classical bounds [34].

The EFT for heat between two quantum systems in thermal equilibrium at different temperatures was proposed by Jarzynski and Wójcik in 2004 [12]. In their original paper, they assumed a negligible energy of interaction between the two quantum systems. Such a weak-coupling assumption was also employed in many subsequent studies of heat statistics between a quantum harmonic oscillator and its surrounding heat bath within the dynamical framework of an open quantum system [35–37]. As revealed in Refs. [35,36], the EFT for quantum heat can be still verified in a Born-Markovian relaxation process. In those works, the system-bath interaction is weak instead of completely ignorable. In this sense, the primal EFT proposed by Jarzynski and Wójcik has been greatly generalized.

However, the usual Born-Markovian approximation is no longer available if the system-bath coupling becomes strong. Relatively few studies focus on the statistics of quantum heat in the strong-coupling regime where the non-Markovian memory effect inevitably appears. A very interesting question naturally arises here: Does the EFT for quantum heat still hold in the strongly non-Markovian regime?

To address the above question, one needs to go beyond the limitations of the usual weak-coupling and rotating-wave approximations. In previous works, the polaron transformation method [38], the path integral approach [39,40], and some other numerical tools [41–43] have been employed to investigate the characteristics of quantum heat in strongly coupled open systems. In this paper, we present an alternative way to explore the EFT for heat in the quantum Brownian motion. By using the normal mode transformation and the phase-space formulation approach, we can derive an analytical result and build a clear physical picture on heat statistics. It is demonstrated that the EFT for quantum heat can be well satisfied if the temperature of the heat bath is so high that the quantum system experiences a canonical thermalization, which is in agreement with previous studies [35,36,44]. However, in the low-temperature regime, we find the EFT generally breaks down due to the non-Markovian effect.

II. HEAT STATISTICS IN AN OPEN QUANTUM SYSTEM

In this section, we first briefly outline the definition of quantum heat based on the two-point measurement scheme [35,36,44,45] (Sec. II A). Related to the symmetry relation of the characteristic function, we recall two important EFTs for the quantum heat [12] (Sec. II B). Then, to analytically

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derive the expression of the characteristic function in an open quantum system, we need to present some necessary details of the phase-space formalism approach [46,47] (Sec. II C). Throughout this paper, we set $\hbar = k_B = 1$ for the sake of simplicity.

A. Two-point measurement scheme

There are many arguments on the definition of heat in an open quantum system, especially in the strong-coupling regime [25,48–53]. In this paper, we regard the quantum heat as the energy change of the heat bath plus the system-bath interaction [54]. Such an energy exchange can be detected by measuring the system itself. If the coupling strength is weak, which means the energy of the interaction Hamiltonian becomes ignorable, our result reduces to the situation considered in previous Refs. [35,36,44].

Let us consider a general time-independent open quantum system, whose total Hamiltonian reads $\hat{H}_{\rm tot} = \hat{H}_{\rm s} + \hat{H}_{\rm b} + \hat{H}_{\rm int}$. Here, $\hat{H}_{\rm s}$ and $\hat{H}_{\rm b}$ are, respectively, the Hamiltonian of the quantum system and the heat bath, and $\hat{H}_{\rm int}$ denotes the interaction Hamiltonian. In Ref. [40], the quantum heat in an open quantum system is defined by two-point energy measurements over the heat bath. However, such a definition is hardly realized due to the huge number of degrees of freedom in the heat bath. Notice that no external driving is applied to the whole system, which means no work is performed during the relaxation process. Thus, the energy of the whole system shall be conservative. If one views the heat as the energy change of the heat bath plus the system-bath interaction, it can be quantified by measuring only the quantum system [44]. Using this particular property, we can define the quantum heat as

$$\mathcal{Q}_{mn} = E_{\rm s}^m - E_{\rm s}^n, \tag{1}$$

where $E_{\rm s}^n$ and $E_{\rm s}^m$ are the eigenenergies of the quantum system at the initial time $t = 0$ and the final time $t = \tau$, respectively. The probability of obtaining the energy $E_{\rm s}^n$ at the initial time is

$$\mathcal{P}_n^0 = {\rm Tr}\big[\rho_{\rm tot}(0)\big|E_{\rm s}^n\big\rangle\big\langle E_{\rm s}^n\big|\big], \tag{2}$$

where $|E_{\rm s}^n\rangle$ denotes the eigenstate with respect to the eigenenergy $E_{\rm s}^n$. In our paper, we assume the whole system is initially prepared in a product state, i.e.,

$$\begin{aligned}\rho_{\rm tot}(0) =&\rho_{\rm s}(0)\otimes\rho_{\rm b}(0) = \rho_{\rm s}^{\rm th}\otimes\rho_{\rm b}^{\rm th}\\ =&\frac{e^{-\beta_{\rm s}\hat{H}_{\rm s}}}{Z_{\rm s}(\beta_{\rm s})}\otimes\frac{e^{-\beta_{\rm b}\hat{H}_{\rm b}}}{Z_{\rm b}(\beta_{\rm b})},\end{aligned} \tag{3}$$

where $\beta_{\rm s} = 1/T_{\rm s}$ and $\beta_{\rm b} = 1/T_{\rm b}$ are the inverse temperatures of the quantum system and the heat bath, respectively. Here, $Z_\alpha(\beta_\alpha) \equiv {\rm Tr}(e^{-\beta_\alpha\hat{H}_\alpha})$ with $\alpha = \{{\rm s, b}\}$ are the partition functions. Then, the conditional transition probability of obtaining the energy $E_{\rm s}^m$ at the final time $t = \tau$ is given by

$$\mathcal{P}_{mn}^\tau = {\rm Tr}\big\{\big|E_{\rm s}^m\big\rangle\big\langle E_{\rm s}^m\big|\hat{U}(\tau)\big[\big|E_{\rm s}^n\big\rangle\big\langle E_{\rm s}^n\big|\otimes\rho_{\rm b}^{\rm th}\big]\hat{U}^\dagger(\tau)\big\}, \tag{4}$$

where $\hat{U}(\tau) = e^{-i\tau\hat{H}_{\rm tot}}$ denotes the time-evolution operator of the whole composite system. Thus, the probability distribution function of the quantum heat during the above decoherence process is then computed as

$$\mathcal{P}_\tau(\mathcal{Q}) \equiv \sum_{m,n}\mathcal{P}_{mn}^\tau\mathcal{P}_n^0\delta(\mathcal{Q} - \mathcal{Q}_{mn}). \tag{5}$$

The characteristic function (or the so-called generating function) with respect to $\mathcal{P}_\tau(\mathcal{Q})$ reads [44]

$$\begin{aligned}\chi_\tau(\nu) \equiv& \int_{-\infty}^{+\infty} d\mathcal{Q}e^{i\nu\mathcal{Q}}\mathcal{P}_\tau(\mathcal{Q})\\ =&{\rm Tr}\big[e^{i\nu\hat{H}_{\rm s}}\hat{U}(\tau)e^{-i\nu\hat{H}_{\rm s}}\rho_{\rm tot}(0)\hat{U}^\dagger(\tau)\big].\end{aligned} \tag{6}$$

With the above expression at hand, the kth moment of quantum heat can be calculated by

$$\langle\mathcal{Q}^{\rm k}(\tau)\rangle = (-i)^{\rm k}\frac{\partial^{\rm k}}{\partial\nu^{\rm k}}\chi_\tau(\nu)\bigg|_{\nu=0}. \tag{7}$$

Equations (6) and (7) fully characterize the statistical character of the quantum heat in an open system.

B. EFTs for quantum heat

EFTs for the quantum heat are closely related to the symmetry relation of the characteristic function. If the characteristic function has the following symmetry relation:

$$\chi_\tau(\nu) = \chi_\tau[-i(\beta_{\rm b} - \beta_{\rm s}) - \nu], \tag{8}$$

one can prove the probability distribution function satisfies

$$\frac{\mathcal{P}_\tau(\mathcal{Q})}{\mathcal{P}_\tau(-\mathcal{Q})} = e^{-(\beta_{\rm b}-\beta_{\rm s})\mathcal{Q}}, \tag{9}$$

which is called the EFT in the differential form [12]. Especially, by setting $\nu = 0$, one can find

$$\chi_\tau(0) = \chi_\tau[-i(\beta_{\rm b} - \beta_{\rm s})] = 1, \tag{10}$$

from which the EFT in the integral form [12]

$$\langle e^{(\beta_{\rm b}-\beta_{\rm s})\mathcal{Q}}\rangle = 1 \tag{11}$$

can be established. These EFTs provide insight for us to understand the second law of thermodynamics from statistical perspectives [35,36,44].

C. Phase-space formulation approach

To analytically derive the expression of the characteristic function in the quantum Brownian motion, we employe the phase-space formulation approach reported in Refs. [46,47]. We first rewrite Eq. (6) into

$$\chi_\tau(\nu) = {\rm Tr}\big[e^{i\nu\hat{\mathcal{H}}_{\rm s}(\tau)}\varrho_\nu(0)\big], \tag{12}$$

where $\hat{\mathcal{H}}_{\rm s}(\tau) \equiv \hat{U}^\dagger(\tau)\hat{H}_{\rm s}\hat{U}(\tau)$ denotes the Hamiltonian of the quantum system in the Heisenberg picture, and $\varrho_\nu(0) \equiv e^{-i\nu\hat{H}_{\rm s}}\rho_{\rm tot}(0)$ is introduced as a shifted density operator. Then, using the Weyl symbol, one can express Eq. (12) in the language of the phase-space formulation of quantum mechanics as

$$\chi_\tau(\nu) = \frac{1}{(2\pi)^\epsilon}\int dz[e^{i\nu\hat{\mathcal{H}}_{\rm s}(\tau)}]_w(z)\times[\varrho_\nu(0)]_w(z), \tag{13}$$

where $z = (\boldsymbol{q},\boldsymbol{p})^{\rm T} = (q_1, q_2, \ldots, q_\epsilon, p_1, p_2, \ldots, p_\epsilon)^{\rm T}$ with $\epsilon = {\rm Dim}(\hat{H}_{\rm tot})$ represent the points in the classical phase space. Here, the notation $[\hat{\mathcal{O}}]_w(z)$ denotes the Weyl symbol of a given operator $\hat{\mathcal{O}}$, which is defined by [55]

$$[\hat{\mathcal{O}}]_w(z) = [\hat{\mathcal{O}}]_w(\boldsymbol{q}, \boldsymbol{p}) \equiv \int d\boldsymbol{\xi} \left\langle \boldsymbol{q} - \frac{1}{2}\boldsymbol{\xi} \middle| \hat{\mathcal{O}} \middle| \boldsymbol{q} + \frac{1}{2}\boldsymbol{\xi} \right\rangle e^{i\boldsymbol{p}\cdot\boldsymbol{\xi}}. \quad (14)$$

Thus, as long as the Weyl symbols of $e^{i\nu\hat{\mathcal{H}}_s(\tau)}$ and $\varrho_\nu(0)$ are obtained, the expression of the characteristic function $\chi_\tau(\nu)$ can be accordingly derived by simply performing the integrals over the variable z.

III. CALDEIRA-LEGGETT MODEL

In this section, we first introduce the famous Caldeira-Leggett model [56,57] (Sec. III A), which describes the dissipative dynamics of a quantum harmonic oscillator coupled to a heat bath consisting of a collection of bosonic modes. The reduced dynamics of the Caldeira-Leggett model can be exactly treated by using various methods, for example, the Heisenberg-Langevin equation of motion [58,59], the path-integral approach [60,61], and the stochastic quantum dynamical scheme [62,63] as well as the normal mode transformation technique [64–67]. In this paper, we employe the tool of the normal mode transformation and display the corresponding details in Sec. III B.

A. The Hamiltonian

The quantum Brownian motion is generally described by the Caldeira-Leggett model, whose Hamiltonian is given by

$$\hat{H}_{\rm CL} = \frac{1}{2}\left\{\hat{p}_0^2 + \omega_0^2\hat{q}_0^2 + \sum_{j=1}^{\rm n}\left[\hat{p}_j^2 + \omega_j^2\left(\hat{q}_j - \frac{c_j}{\omega_j^2}\hat{q}_0\right)^2\right]\right\}, \quad (15)$$

where $\hat{q}_0$ ($\hat{q}_j$) and $\hat{p}_0$ ($\hat{p}_j$) are the position and momentum operators of the quantum system (the heat bath) with the corresponding frequency ω_0 (ω_j), respectively. Parameters c_j quantify the system-bath coupling strengths. In this paper, all the masses of these quantum harmonic oscillators are weighted as $M_0 = M_j = 1$.

Commonly, it is very convenient to encode the frequency dependence of the interaction strengths into the so-called spectral density, which is defined by

$$J(\omega) = \frac{\pi}{2}\sum_{j=1}^{\rm n}\frac{c_j^2}{\omega_j}[\delta(\omega_j - \omega) - \delta(\omega_j + \omega)]. \quad (16)$$

The associated friction function $\gamma(t)$ of the Caldeira-Leggett model is introduced as

$$\gamma(t) = \sum_{j=1}^{\rm n}\frac{c_j^2}{\omega_j^2}\cos(\omega_j t). \quad (17)$$

The spectral density considered in this paper is an Ohmic spectral density [58,68]

$$J(\omega) = \eta\omega, \quad (18)$$

where η denotes the coupling strength. Usually, a damping function $f(\omega, \omega_c)$ is included in the expression of $J(\omega)$, such as $J(\omega) = \eta\omega f(\omega, \omega_c)$ with an exponential cutoff $f(\omega, \omega_c) = e^{-\omega/\omega_c}$, a Lorentzian cutoff $f(\omega, \omega_c) = \omega_c^2/(\omega^2 + \omega_c^2)$ or a sudden (Heaviside step) cutoff $f(\omega, \omega_c) = \theta(\omega_c - \omega)$. The introduction of $f(\omega, \omega_c)$ ensures that $J(\omega)$ vanishes when $\omega \to \infty$. Due to the normal mode transformation adopted in our paper, the final results are not directly connected to $J(\omega)$ but to the spectral density of normal modes $I(\Omega)$ [see Eqs. (27) and (28) and Appendix A]. In the main text, we consider the case $f(\omega, \omega_c) = 1$, which can be regarded as the special situation $\omega_c \gg \omega_0$. In Appendix B, we present the results when $f(\omega, \omega_c) = \omega_c^2/(\omega^2 + \omega_c^2)$. The specific form of $f(\omega, \omega_c)$ does not influence our physical conclusions.

The corresponding friction function reads [68]

$$\gamma(t) = 2\eta\delta(t), \quad (19)$$

which is a Dirac-δ function. Although only the Ohmic spectral density is displayed in this paper, it is necessary to point out that our result can be generalized to other cases without difficulty.

B. The normal mode transformation

Since the Caldeira-Leggett Hamiltonian has a quadratic form, it can be diagonalized by an orthogonal rotation and reexpressed as

$$\hat{H}_{\rm CL} = \frac{1}{2}\sum_{j=1}^{N}(\hat{P}_j^2 + \Omega_j^2\hat{Q}_j^2), \quad (20)$$

where the new coordinates $\hat{Q}_j$ are related to the original coordinates via the following normal mode transformation:

$$\begin{pmatrix}\hat{q}_0\\ \hat{q}_1\\ \vdots\\ \hat{q}_{\rm n}\end{pmatrix} = \begin{pmatrix}u_{00} & u_{01} & \cdots & u_{0\rm n}\\ u_{10} & u_{11} & \cdots & u_{1\rm n}\\ \vdots & \vdots & \ddots & \vdots\\ u_{\rm n0} & u_{\rm n1} & \cdots & u_{\rm nn}\end{pmatrix}\begin{pmatrix}\hat{Q}_0\\ \hat{Q}_1\\ \vdots\\ \hat{Q}_{\rm n}\end{pmatrix}. \quad (21)$$

The elements of the transformation matrix given by Eq. (21) are [64–67]

$$u_{j0}^2 = \left[1 + \sum_{l=1}^{\rm n}\frac{c_l^2}{(\Omega_j^2 - \omega_l^2)^2}\right]^{-1}, \; u_{jl} = \frac{c_l}{\Omega_j^2 - \omega_l^2}u_{j0}, \quad (22)$$

with $l = 1, 2, 3, ..., {\rm n}$. The normal mode frequencies Ω_j are determined by the following normal mode eigenvalue equation [64–67]:

$$\Omega_j^2 = \omega_0^2\left[1 + \sum_{l=1}^{\rm n}\frac{c_l^2}{\omega_l^2(\omega_l^2 - \Omega_j^2)}\right]^{-1}. \quad (23)$$

Now Eq. (20) is a sum of separable quantum harmonic oscillators and it is easy to write the equations of motion for each of the normal modes as

$$\hat{Q}_j(t) = \hat{Q}_j(0)\cos(\Omega_j t) + \frac{\hat{P}_j(0)}{\Omega_j}\sin(\Omega_j t) \quad (24)$$

and $\hat{P}_j(t) = d\hat{Q}_j(t)/dt$. These equations of motion are completely identical to those of classical harmonic oscillators. Going back to the original modes, one finds that the time evolutions of the coordinate and the momentum of the quantum system are determined by

$$\hat{q}_0(t) = \sum_{\ell=0}^{n} \dot{\Theta}_\ell(t)\hat{q}_\ell(0) + \sum_{\ell=0}^{n} \Theta_\ell(t)\hat{p}_\ell(0) \tag{25}$$

and $\hat{p}_0(t) = d\hat{q}_0(t)/dt$. Here, the expressions of $\Theta_\ell(t)$ are

$$\Theta_\ell(t) = \sum_{j=0}^{n} u_{j0}u_{j\ell}\frac{\sin(\Omega_j t)}{\Omega_j}. \tag{26}$$

To recast the normal mode representation in the continuum limit, one can define a spectral density of normal modes as

$$I(\Omega) = \frac{\pi}{2}\sum_{j=0}^{n}\frac{u_{j0}^2}{\Omega_j}[\delta(\Omega_j - \Omega) - \delta(\Omega_j + \Omega)], \tag{27}$$

which is related to $J(\omega)$ by [64–67]

$$I(\Omega) = \frac{J(\Omega)}{[\omega_0^2 - \Omega^2 - \Omega \mathrm{Im}\acute{\gamma}(i\Omega)]^2 + J^2(\Omega)}, \tag{28}$$

with $\acute{\gamma}(z)$ being the Laplace transformation of the friction function $\gamma(t)$. The introduction of the spectral density $I(\Omega)$ is very useful when computing the continuum limit of various functions that appear in the analytical expressions of $\chi_\tau(\nu)$ and $\langle\mathcal{Q}(\tau)\rangle$ (see Ref. [67] and Appendix A for details).

IV. RESULTS AND DISCUSSIONS

In this section, we first present our main results, namely, the exact expressions of the characteristic function $\chi_\tau(\nu)$ and the average heat $\langle\mathcal{Q}(\tau)\rangle$ (Sec. IV A). They are complicated in their general forms. To gain a clear physical picture on the quantum heat statistics, we also provide the asymptotic expressions of $\chi_\tau(\nu)$ and $\langle\mathcal{Q}(\tau)\rangle$ in the high-temperature limit (Sec. IV B). Using these approximate expressions as benchmarks, we reexamine the EFT for heat distribution in the quantum Brownian motion without invoking the Born-Markovian and the weak-coupling approximations (Sec. IV C).

A. Analytical expressions of $\chi_\tau(\nu)$ and $\langle\mathcal{Q}(\tau)\rangle$

To compute the characteristic function, one needs to derive the Weyl symbol of the exponential Hamiltonian of the dissipative quantum harmonic oscillator. This key Weyl symbol was already known from several early works [69,70]. Following the detailed expositions in Refs. [69,70], for a quantum harmonic oscillator $\hat{H}_{\rm HO}(\hat{\rm q},\hat{\rm p}) = \frac{1}{2}(\hat{\rm p}^2 + \varpi^2\hat{\rm q}^2)$, the Weyl symbol of $e^{\zeta\hat{H}_{\rm HO}(\hat{\rm q},\hat{\rm p})}$ is given by

$$[e^{\zeta\hat{H}_{\rm HO}(\hat{\rm q},\hat{\rm p})}]_w({\rm q},{\rm p}) = \mathrm{sech}\left(\frac{1}{2}\zeta\varpi\right)\exp\left[\frac{2}{\varpi}\tanh\left(\frac{1}{2}\zeta\varpi\right)H_{\rm HO}(q,p)\right]. \tag{29}$$

Using the above expression and the equation of motion given by Eq. (25), we find

$$\begin{aligned}[e^{i\nu\hat{\mathcal{H}}_s(\tau)}]_w(z) &= \left\{e^{\frac{i}{2}\nu[\hat{p}_0^2(\tau)+\omega_0^2\hat{q}_0^2(\tau)]}\right\}_w(z)\\ &= \frac{1}{\cos\left(\frac{1}{2}\omega_0\nu\right)}\exp\left[i\omega_0\tan\left(\frac{1}{2}\omega_0\nu\right)q_0^2(\tau) + \frac{i}{\omega_0}\tan\left(\frac{1}{2}\nu\omega_0\right)p_0^2(\tau)\right]\\ &= \frac{1}{\cos\left(\frac{1}{2}\nu\omega_0\right)}\exp\left(\frac{i}{2}z^{\rm T}\Lambda_{\nu\tau}z\right),\end{aligned} \tag{30}$$

where $\Lambda_{\nu\tau} = V_{q\tau}V_{q\tau}^{\rm T} + V_{p\tau}V_{p\tau}^{\rm T}$ is a $(2{\rm n}+2)\times(2{\rm n}+2)$ matrix with

$$V_{q\tau} = \sqrt{2\omega_0\tan\left(\frac{1}{2}\omega_0\nu\right)}[\dot{\Theta}_0(\tau), \dot{\Theta}_1(\tau), ..., \dot{\Theta}_{\rm n}(\tau), \Theta_0(\tau), \Theta_1(\tau), ..., \Theta_{\rm n}(\tau)]^{\rm T} \tag{31}$$

and $V_{p\tau} = \dot{V}_{q\tau}/\omega_0$. In the same way, the Weyl symbol of $\varrho_\nu(0)$ is given by

$$[\varrho_\nu(0)]_w(z) = \frac{2\sinh(\frac{1}{2}\beta_{\rm s}\omega_0)}{\cosh(\frac{1}{2}\beta_\nu\omega_0)}\prod_{j=1}^{n}\left[2\tanh\left(\frac{1}{2}\beta_{\rm b}\omega_j\right)\right]\exp\left(-\frac{1}{2}z^{\rm T}\Lambda_{\nu\beta}z\right), \tag{32}$$

where $\beta_\nu \equiv \beta_{\rm s} + i\nu$ and $\Lambda_{\nu\beta}$ is a $(2{\rm n}+2)\times(2{\rm n}+2)$ diagonal matrix

$$\begin{aligned}\Lambda_{\nu\beta} = \mathrm{Diag}\bigg\{&2\omega_0\tanh\left(\frac{\beta_\nu\omega_0}{2}\right), 2\omega_1\tanh\left(\frac{\beta_{\rm b}\omega_1}{2}\right), ..., 2\omega_{\rm n}\tanh\left(\frac{\beta_{\rm b}\omega_{\rm n}}{2}\right),\\ &\times\frac{2}{\omega_0}\tanh\left(\frac{\beta_\nu\omega_0}{2}\right), \frac{2}{\omega_1}\tanh\left(\frac{\beta_{\rm b}\omega_1}{2}\right), ..., \frac{2}{\omega_{\rm n}}\tanh\left(\frac{\beta_{\rm b}\omega_{\rm n}}{2}\right)\bigg\}.\end{aligned} \tag{33}$$

Then, by using the Gaussian integral

$$\int d\boldsymbol{\zeta} e^{-\frac{1}{2}\boldsymbol{\zeta}^{\rm T}\mathbf{Q}\boldsymbol{\zeta}} = \sqrt{\frac{(2\pi)^{\mathrm{Dim}(\mathbf{Q})}}{\mathrm{Det}(\mathbf{Q})}}, \tag{34}$$

one can derive the expression of the characteristic function as [44]

$$\chi_\tau(\nu)=\sqrt{\frac{\mathrm{Det}(\Lambda_{\nu\beta}-i\Lambda_{\nu 0})}{\mathrm{Det}(\Lambda_{\nu\beta}-i\Lambda_{\nu\tau})}}=[\mathrm{Det}(\mathbf{I}_4+i\mathcal{M}_{\nu\tau})]^{-\frac{1}{2}}, \tag{35}$$

where $\mathbf{I}_4$ is a 4×4 identity matrix and

$$\mathcal{M}_{\nu\tau}=\sqrt{(\Lambda_{\nu\beta}-i\Lambda_{\nu 0})^{-1}}(V_{q0},V_{p0},iV_{q\tau},iV_{p\tau})\begin{pmatrix}V_{q0}\\ V_{p0}\\ iV_{q\tau}\\ iV_{p\tau}\end{pmatrix}\sqrt{(\Lambda_{\nu\beta}-i\Lambda_{\nu 0})^{-1}}. \tag{36}$$

Using Eq. (31), one can find 16 matrix elementals of $\mathcal{M}_{\nu\tau}$ are

$$\mathcal{M}_{\nu\tau}=\Xi_\nu\begin{bmatrix}1 & 0 & i\dot{\Theta}_0(\tau) & i\omega_0^{-1}\ddot{\Theta}_0(\tau)\\ 0 & 1 & i\omega_0\Theta_0(\tau) & i\dot{\Theta}_0(\tau)\\ i\dot{\Theta}_0(\tau) & i\omega_0\dot{\Theta}_0(\tau) & -2\Upsilon_{\nu\tau} & \omega_0^{-1}\dot{\Upsilon}_{\nu\tau}\\ i\omega_0^{-1}\ddot{\Theta}_0(\tau) & i\dot{\Theta}_0(\tau) & \omega_0^{-1}\dot{\Upsilon}_{\nu\tau} & -\Phi_{\nu\tau}\end{bmatrix}, \tag{37}$$

where

$$\Xi_\nu=\frac{\tan\left(\frac{1}{2}\nu\omega_0\right)}{\tanh\left(\frac{1}{2}\beta_\nu\omega_0\right)-i\tan\left(\frac{1}{2}\nu\omega_0\right)}, \tag{38}$$

$$\Upsilon_{\nu\tau}=\frac{1}{2}\dot{\Theta}_0^2(\tau)+\frac{1}{2}\omega_0^2\Theta_0^2(\tau)+\frac{\omega_0\tan\left(\frac{1}{2}\omega_0\nu\right)}{2\Xi_\nu}\mathcal{G}(\tau),\ \mathcal{G}(\tau)=\sum_{j=1}^{\mathrm{n}}\frac{\dot{\Theta}_j^2(\tau)+\omega_j^2\Theta_j^2(\tau)}{\omega_j\tanh\left(\frac{1}{2}\beta_\mathrm{b}\omega_j\right)}, \tag{39}$$

$$\Phi_{\nu\tau}=\frac{1}{\omega_0^2}\ddot{\Theta}_0^2(\tau)+\dot{\Theta}_0^2(\tau)+\frac{\tan\left(\frac{1}{2}\omega_0\nu\right)}{\Xi_\nu\omega_0}\mathcal{K}(\tau),\ \mathcal{K}(\tau)=\sum_{j=1}^{\mathrm{n}}\frac{\ddot{\Theta}_j^2(\tau)+\omega_j^2\dot{\Theta}_j^2(\tau)}{\omega_j\tanh\left(\frac{1}{2}\beta_\mathrm{b}\omega_j\right)}. \tag{40}$$

With the expression of the characteristic function at hand, the average heat $\langle\mathcal{Q}(\tau)\rangle$ is given by

$$\langle\mathcal{Q}(\tau)\rangle=\frac{\ddot{\Theta}_0^2(\tau)+2\omega_0^2[\dot{\Theta}_0^2(\tau)-1]+\omega_0^4\Theta_0^2(\tau)}{4\omega_0\tanh(\frac{1}{2}\beta_\mathrm{s}\omega_0)}+\frac{1}{4}\mathcal{K}(\tau)+\frac{1}{4}\omega_0^2\mathcal{G}(\tau). \tag{41}$$

All results of $\Theta_0(\tau)$, $\mathcal{G}(\tau)$, and $\mathcal{K}(\tau)$ can be expressed in terms of the spectral density of normal modes $I(\Omega)$ in the continuum limit (see Appendix A for details).

B. The high-temperature limit

In the exact results of Eqs. (35) and (41), the expressions of $\chi_\tau(\nu)$ and $\langle\mathcal{Q}(\tau)\rangle$ are complicated. We leave them to purely numerical calculations. However, via analyzing the high-temperature behaviors of $\mathcal{G}(\tau)$ and $\mathcal{K}(\tau)$, we can obtain their asymptotic forms, which are helpful for us to build a physical picture on the heat distribution in the quantum Brownian motion. In the high-temperature limit, namely, $\omega_j\beta_\mathrm{b}\to 0$, we have $\coth(\frac{1}{2}\omega_j\beta_\mathrm{b})\simeq 2/(\omega_j\beta_\mathrm{b})$, which results in

$$\mathcal{G}_\mathrm{HT}(\tau)\simeq\frac{2}{\beta_\mathrm{b}\omega_0^2}+\frac{2}{\beta_\mathrm{b}\omega_0^2}e^{-\eta\tau}\times\left[\frac{\eta^2\cos(2\varsigma\tau)}{4\varsigma^2}-\frac{\eta\sin(2\varsigma\tau)}{2\varsigma}-\frac{\omega_0^2}{\varsigma^2}\right], \tag{42}$$

$$\mathcal{K}_\mathrm{HT}(\tau)\simeq\frac{2}{\beta_\mathrm{b}}+\frac{2}{\beta_\mathrm{b}}e^{-\eta\tau}\times\left[\frac{\eta^2\cos(2\varsigma\tau)}{4\varsigma^2}+\frac{\eta\sin(2\varsigma\tau)}{2\varsigma}-\frac{\omega_0^2}{\varsigma^2}\right], \tag{43}$$

where $\varsigma\equiv\sqrt{\omega_0^2-\eta^2/4}$. With these expressions at hand, one can easily prove that the characteristic function $\chi_\tau(\nu)$ satisfies the symmetry relation $\chi_\tau(\nu)=\chi_\tau[-i(\beta_\mathrm{b}-\beta_\mathrm{s})-\nu]$, which means the EFT in the differential form holds at any relaxation time in the high-temperature regime. This result is in good agreement with previous Born-Markovian studies [35,36] because the dissipative quantum harmonic oscillator generally experiences a Markovian decoherence at high temperature [71–73].

Such a Markovian dynamics leads to a canonical thermalization, namely, $\rho_\mathrm{s}(\infty)=\rho_\mathrm{s}^\mathrm{th}$, which validates the EFT in the equilibrium steady state, as demonstrated in many previous references [35,36,44]. To clarify this point, we can take the long-time limit $\omega_0\tau\to\infty$, which leads to $\Theta_0(\infty)=0$ and

$$\mathcal{G}_\mathrm{HT}(\infty)=\frac{2}{\beta_\mathrm{b}\omega_0^2}\simeq\frac{1}{\omega_0}\coth\left(\frac{\beta_\mathrm{b}\omega_0}{2}\right), \tag{44}$$

$$\mathcal{K}_\mathrm{HT}(\infty)=\frac{2}{\beta_\mathrm{b}}\simeq\omega_0\coth\left(\frac{\beta_\mathrm{b}\omega_0}{2}\right). \tag{45}$$

Using these long-time expressions, we immediately find

$$\chi_\infty^\mathrm{HT}(\nu)=\chi_\mathrm{th}(\nu)=\frac{Z_\mathrm{s}(\beta_\mathrm{s}+i\nu)Z_\mathrm{b}(\beta_\mathrm{b}-i\nu)}{Z_\mathrm{s}(\beta_\mathrm{s})Z_\mathrm{b}(\beta_\mathrm{b})}, \tag{46}$$

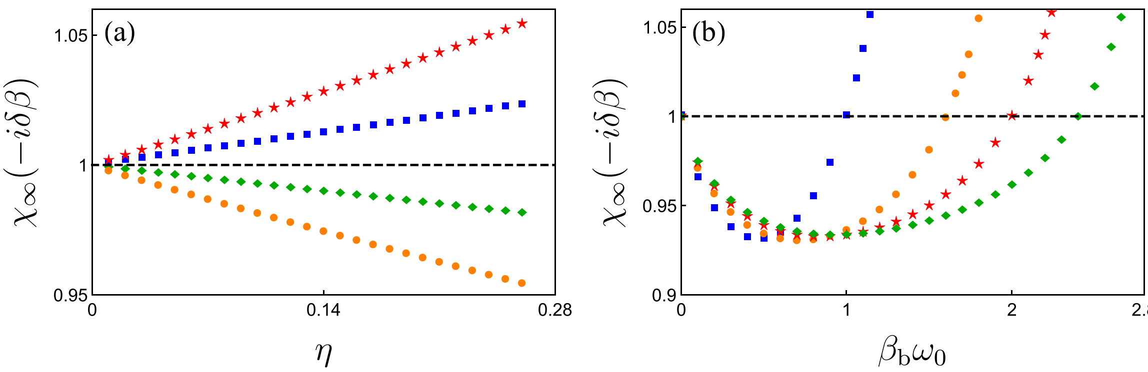

FIG. 1. (a) The characteristic function $\chi_\infty(-i\delta\beta)$ with $\delta\beta \equiv \beta_b - \beta_s$ is plotted as the function of the coupling strength η with different temperatures of the heat bath: $\omega_0\beta_b = 2.4$ (red stars), $\omega_0\beta_b = 2.2$ (blue rectangles), $\omega_0\beta_b = 0.2$ (green diamonds), and $\omega_0\beta_b = 1$ (orange circles). Other parameters are chosen as $\omega_0\beta_s = 2$ and $\omega_0 = 2\ \mathrm{cm}^{-1}$. (b) The characteristic function $\chi_\infty(-i\delta\beta)$ versus the temperature of the heat bath $\omega_0\beta_b$ with different temperatures of the quantum system: $\omega_0\beta_s = 1$ (blue rectangles), $\omega_0\beta_s = 1.6$ (orange circles), $\omega_0\beta_s = 2$ (red stars), and $\omega_0\beta_s = 2.4$ (green diamonds). Other parameters are chosen as $\eta = 0.04$ and $\omega_0 = 2\,\mathrm{cm}^{-1}$.

which is independent of the details of the relaxation dynamics, say, the coupling strength η. And Eq. (46) recovers the conclusion reported in Ref. [44].

On the other hand, with the help of Eqs. (42) and (43), the expression of $\langle \mathcal{Q}(\tau)\rangle$ reduces to

$$\begin{aligned}\langle \mathcal{Q}_{\mathrm{HT}}(\tau)\rangle \simeq{}& \frac{e^{-\eta\tau}}{\beta_b}\left[\frac{e^{\eta\tau}\varsigma^2-\omega_0^2}{\varsigma^2}+\frac{\eta^2\cos(2\varsigma\tau)}{4\varsigma^2}\right]\\ &+\coth\left(\frac{\beta_s\omega_0}{2}\right)\left[\frac{\omega_0(\omega_0^2-e^{\eta\tau}\varsigma^2)}{2\varsigma^2}-\frac{\eta^3\sin(2\varsigma\tau)}{8\omega_0\varsigma}\right.\\ &\left.-\frac{(\omega_0^4-3\varsigma^2\omega_0^2+2\varsigma^2)\cos(2\varsigma\tau)}{2\omega_0\varsigma^2}\right]e^{-\eta\tau}. \end{aligned} \tag{47}$$

From the above expression, one can see the average heat starts from $\langle \mathcal{Q}(0)\rangle = 0$ and gradually approaches to a steady value, which is given by

$$\begin{aligned}\langle \mathcal{Q}_{\mathrm{HT}}(\infty)\rangle &= \langle \mathcal{Q}_{\mathrm{th}}\rangle\\ &= \frac{1}{2}\omega_0\left[\frac{2}{\beta_b\omega_0}-\coth\left(\frac{\beta_s\omega_0}{2}\right)\right]\\ &\simeq \frac{1}{2}\omega_0\left[\coth\left(\frac{\beta_b\omega_0}{2}\right)-\coth\left(\frac{\beta_s\omega_0}{2}\right)\right]. \end{aligned}\tag{48}$$

The above expression of $\langle \mathcal{Q}_{\mathrm{HT}}(\infty)\rangle$ is simply the difference of the mean energies of the quantum harmonic oscillator at temperatures $1/\beta_b$ and $1/\beta_s$. It can be rewritten in terms of the thermal occupation number, namely, $\langle \mathcal{Q}_{\mathrm{HT}}(\infty)\rangle = \omega_0[\bar{n}(\beta_b) - \bar{n}(\beta_s)]$ with $\bar{n}(\beta_\alpha) \equiv (e^{\omega_0\beta_\alpha} - 1)^{-1}$. This result is consistent with that of Ref. [35] and is physically reasonable.

C. Exact numerical results

To justify these high-temperature results in the above subsection, we also provide the exact numerical simulations. In Fig. 1(a), we plot the characteristic function $\chi_\infty[-i(\beta_b - \beta_s)]$ as a function of the dimensionless coupling strength η. One can see $\chi_\infty[-i(\beta_b - \beta_s)] \simeq \chi_\infty(0) = 1$ if η is small, which means the EFT in the integral form is satisfied in the weak-coupling regime. However, as the coupling strength becomes strong, the value of $\chi_\infty[-i(\beta_b - \beta_s)]$ moves away from $\chi_\infty(0) = 1$, which implies the appearance of the noncanonical thermalization as well as the breakdown of the EFT. As demonstrated in many previous studies [73–76], the emergence of a noncanonical distribution is commonly linked to the non-Markovian effect. In this sense, our result suggests the non-Markovianity may invalidate the EFT. Moreover, we display $\chi_\infty[-i(\beta_b - \beta_s)]$ as the function of the temperature of the heat bath in Fig. 1(b). From Fig. 1(b), one can find $\chi_\infty[-i(\beta_b - \beta_s)]$ approaches to $\chi_\infty(0) = 1$ with the increase of the bath temperature, i.e., $\omega_0\beta_b \to 0$. This numerical conclusion is completely consistent with that of the analytical analysis in the high-temperature regime. Exceptions can occur in the vicinities of $\omega_0\beta_b = \omega_0\beta_s$, which corresponds to the trivial case: there is no heat exchange if the system and the bath are at the same temperature.

In Fig. 2, we display the dynamics of $\langle \mathcal{Q}(\tau)\rangle$ with (a) different bath temperatures and (b) different coupling strengths. If the system-bath coupling is not too strong, we find, except some oscillations at the beginning of the time evolution, the analytical results obtained by the hight-temperature approximation [Eq. (47)] are in qualitative agreement with exact numerical simulations. However, the long-time equilibrium state is no longer a canonical Gibbs state in the strong-coupling regimes, which leads to a relatively large deviation from the approximate expression of $\langle \mathcal{Q}_{\mathrm{th}}\rangle$ [Eq. (48)], as plotted in the inset figure of Fig. 2(b).

The oscillations observed in Fig. 2 may be traced back to the non-Markovian effect. In the Markovian regime, the energy, which is presented as the quantum heat in our paper, unidirectionally flows between the system and the bath. However, as reported in previous Refs. [77–79], in the non-Markovian regime, an energy backflow may occur and leads to these oscillations of quantum heat flux. Some previous works [77,78] deem the occurrence of the energy backflow as an evidence of non-Markovianity.

V. CONCLUSION

Before concluding our paper, two important remarks shall be made here.

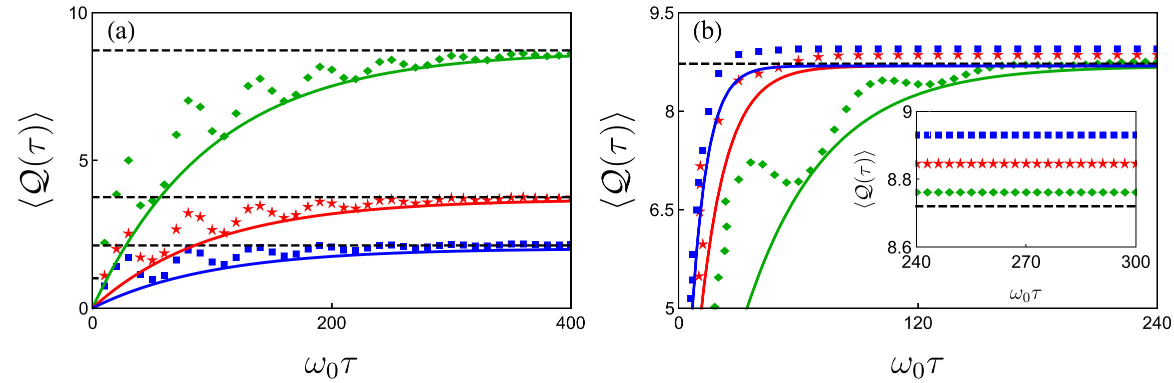

FIG. 2. (a) The dynamics of the average heat $\langle \mathcal{Q}(\tau) \rangle$ is plotted with different temperatures of the heat bath: $\omega_0\beta_b = 0.2$ (green diamonds), $\omega_0\beta_b = 0.4$ (red stars), and $\omega_0\beta_b = 0.6$ (blue rectangles). Other parameters are chosen as $\omega_0\beta_s = 2$ and $\eta = 0.02$. (b) The dynamics of the average heat $\langle \mathcal{Q}(\tau) \rangle$ is plotted with different coupling strengths: $\eta = 0.05$ (green diamonds), $\eta = 0.15$ (red stars), and $\eta = 0.25$ (blue rectangles). The inset figure depicts the steady-state average heat in the long-time regime. Other parameters are chosen as $\omega_0\beta_b = 0.2$, $\omega_0\beta_s = 2$, and $\omega_0 = 2\text{cm}^{-1}$. The solid lines are analytical results predicted by Eq. (47) and the black dashed lines are high-temperature approximate results from the expression of $\langle \mathcal{Q}_{\text{th}} \rangle$ in Eq. (48).

(i) As demonstrated in Sec. IV, the original Jarzynski-Wójcik fluctuation theorem generally breaks down in the strong-coupling regime. This result naturally brings about one question: Does a generalized fluctuation theorem for heat exchange exist in these strong-coupling open systems? Completely different from that of the weak-coupling case, in the strong coupling regime, the joint state of the composite system is so hybridized or entangled that one cannot sufficiently distinguish one from another. In this circumstance, Eq. (3) is no longer applicable and the initial quantum corrections shall be taken into account. In Ref. [80], the authors derived a series of generalized EFTs for initially quantum correlated thermal bipartite system. These fluctuation relations fully capture quantum correlations and quantum coherence, which are commonly neglected in the weak-coupling case. It would be interesting to apply their formalism to the Caldeira-Leggett model considered in this paper.

(ii) In our formalism, the quantum heat is defined as the energy change of the heat bath plus the interaction Hamiltonian. Such an energy transfer can be quantified by measuring only the quantum system, if the whole Caldeira-Leggett model is time-independent. Due to the fact that $\text{Dim}(\hat{H}_s) \ll \text{Dim}(\hat{H}_{\text{tot}})$, the measurements over the quantum system are relatively simple. However, quantum work is generally defined in a time-dependent system. A time-dependent external field, which drives the system out of equilibrium and injects energy into the system, is indispensable. Thus, to investigate the quantum work distribution in the Caldeira-Leggett model, the two-point energy measurement shall be done over the whole composite system $\hat{H}_{\text{tot}}$ [39], which involves a huge number of degrees of freedom. It is still an unknown question whether our scheme is feasible to the case of quantum work. The same problem can be encountered in the study of irreversible entropy production [8,81].

In summary, we investigate the statistics of quantum heat in a non-Markovian decoherence process described by the Caldeira-Leggett model. By using the normal mode transformation and the phase-space formulation approach, we analytically derive the expressions of the characteristic function and the average heat beyond the frequently used Born-Markovian and weak-coupling approximations. At high temperature, the reduced dynamics of the dissipative harmonic oscillator is Markovian and a completely canonical thermalization occurs in the long-time regime. In this case, we find the EFT for quantum heat is fully satisfied at any relaxation time. However, with the decrease of the bath temperature, the dissipative harmonic oscillator experiences a non-Markovian decoherence in the strong-coupling regime, which induces a non-canonical thermalization. Under this circumstance, we find the EFT for quantum heat generally breaks down. The non-Markovian effect on the quantum heat distribution presented in our paper may have potential applications in the study of finite-time quantum heat engines.

ACKNOWLEDGMENTS

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APPENDIX A: CONTINUUM LIMIT

In the continuum limit of normal modes, the explicit expression of $\Theta_0(\tau)$ is

$$\Theta_0(\tau) = \sum_{j=0}^{n} u_{j0}^2 \frac{\sin(\Omega_j \tau)}{\Omega_j} = \frac{2}{\pi} \int_0^\infty d\Omega I(\Omega) \sin(\Omega\tau). \tag{A1}$$

For the Ohmic dissipation considered in the main text, the integral over Ω can be analytically worked out. The expression of $\Theta_0(\tau)$ is then given by

$$\Theta_0(\tau) = \frac{\sin(\varsigma\tau)}{\varsigma} e^{-\frac{1}{2}\eta\tau}, \tag{A2}$$

which decays exponentially.

Next, we derive the expressions of $\mathcal{G}(\tau)$ and $\mathcal{K}(\tau)$ by employing the skills proposed in Ref. [67]. By using the Matsubara expansion of the coth function

$$\coth\left(\frac{\beta_b\omega_j}{2}\right)=\frac{2}{\beta_b\omega_j}+\frac{4}{\beta_b}\sum_{k=1}^{\infty}\frac{\omega_j}{\omega_j^2+v^2k^2},\tag{A3}$$

where $v=2\pi/\beta_b$ and the relation

$$u_{jl}=\frac{c_l}{\Omega_j^2-\omega_l^2}u_{j0},\tag{A4}$$

we have

$$\mathcal{G}(\tau)=\sum_{j=1}^{n}\frac{\dot\Theta_j^2(\tau)+\omega_j^2\Theta_j(\tau)^2}{\omega_j\tanh\left(\frac{1}{2}\beta_b\omega_j\right)}=\frac{2}{\beta_b\omega_j^2}\sum_{j=1}^{n}\left[\sum_{l=1}^{n}\frac{u_{j0}^2\cos(\Omega_\ell\tau)c_j}{\Omega_\ell^2-\omega_j^2}\right]^2+\frac{4}{\beta_b}\sum_{j=1}^{n}\sum_{k=1}^{\infty}\frac{1}{v^2k^2+\omega_j^2}\left[\sum_{l=1}^{n}\frac{u_{j0}^2\cos(\Omega_l\tau)c_j}{\Omega_l^2-\omega_j^2}\right]^2$$
$$+\frac{2}{\beta_b}\sum_{j=1}^{n}\left[\sum_{l=1}^{n}\frac{u_{j0}^2\sin(\Omega_l\tau)c_j}{\Omega_l\left(\Omega_l^2-\omega_j^2\right)}\right]^2+\frac{4\omega_j^2}{\beta_b}\sum_{j=1}^{n}\sum_{k=1}^{\infty}\frac{1}{v^2k^2+\omega_j^2}\left[\sum_{l=1}^{n}\frac{u_{j0}^2\sin(\Omega_l\tau)c_j}{\Omega_l\left(\Omega_l^2-\omega_j^2\right)}\right]^2.\tag{A5}$$

Then, using

$$\left(\sum_\ell a_\ell\right)^2=\sum_\ell a_\ell^2+\sum_{\ell\neq\ell'}a_\ell a_{\ell'},$$

we have

$$\mathcal{G}(\tau)=\frac{2}{\beta_b}\sum_{l=1}^{n}u_{j0}^4\cos^2(\Omega_l\tau)\sum_{j=1}^{n}\frac{c_j^2}{\left(\Omega_l^2-\omega_j^2\right)^2\omega_j^2}+\frac{2}{\beta_b}\sum_{l\neq l'}u_{j0}^2\cos(\Omega_l\tau)u_{l'0}^2\cos(\Omega_{l'}\tau)\sum_{j=1}^{n}\frac{c_j^2}{\left(\Omega_l^2-\omega_j^2\right)\left(\Omega_{l'}^2-\omega_j^2\right)\omega_j^2}$$
$$+\frac{2}{\beta_b}\sum_{l=1}^{n}u_{j0}^4\sin^2(\Omega_l\tau)\sum_{j=1}^{n}\frac{c_j^2}{\Omega_l^2\left(\Omega_l^2-\omega_j^2\right)^2}+\frac{2}{\beta_b}\sum_{l\neq l'}u_{j0}^2\sin(\Omega_l\tau)u_{l'0}^2\sin(\Omega_{l'}\tau)\sum_{j=1}^{n}\frac{c_j^2}{\Omega_l\Omega_{l'}\left(\Omega_l^2-\omega_j^2\right)\left(\Omega_{l'}^2-\omega_j^2\right)}$$
$$+\frac{4}{\beta_b}\sum_{l=1}^{n}u_{j0}^4\cos^2(\Omega_l\tau)\sum_{k=1}^{\infty}\sum_{j=1}^{n}\frac{c_j^2}{\left(\Omega_l^2-\omega_j^2\right)^2\left(v^2k^2+\omega_j^2\right)}$$
$$+\frac{4}{\beta_b}\sum_{l\neq l'}u_{j0}^2\cos^2(\Omega_l\tau)u_{l'0}^2\cos^2(\Omega_{l'}\tau)\sum_{k=1}^{\infty}\sum_{j=1}^{n}\frac{c_j^2}{\left(\Omega_l^2-\omega_j^2\right)\left(\Omega_{l'}^2-\omega_j^2\right)\left(v^2k^2+\omega_j^2\right)}$$
$$+\frac{4}{\beta_b}\sum_{l=1}^{n}u_{j0}^4\sin^2(\Omega_l\tau)\sum_{k=1}^{\infty}\sum_{j=1}^{n}\frac{c_j^2\omega_j^2}{\Omega_l^2\left(\Omega_l^2-\omega_j^2\right)^2\left(v^2k^2+\omega_j^2\right)}$$
$$+\frac{4}{\beta_b}\sum_{l\neq l'}u_{j0}^2\cos^2(\Omega_l\tau)u_{l'0}^2\cos^2(\Omega_{l'}\tau)\sum_{k=1}^{\infty}\sum_{j=1}^{n}\frac{c_j^2\omega_j^2}{\Omega_l\Omega_{l'}\left(\Omega_l^2-\omega_j^2\right)\left(\Omega_{l'}^2-\omega_j^2\right)\left(v^2k^2+\omega_j^2\right)}.\tag{A6}$$

From Eqs. (22) and (23), we note the following two relations:

$$\sum_{j=1}^{n}\frac{c_j^2}{\omega_j^2-\Omega_l^2}=\omega_0^2-\Omega_l^2+\sum_j\frac{c_j^2}{\omega_j^2},\ \sum_{j=1}^{n}\frac{c_j^2}{\left(\omega_j^2-\Omega_l^2\right)^2}=\frac{1}{u_{l0}^2}-1.\tag{A7}$$

On the other hand, from Eq. (17), we find the Laplace transform of the friction function is

$$\acute\gamma(z)=\int_0^\infty dte^{-zt}\gamma(t)=\sum_{j=1}^{n}\frac{c_j^2}{\omega_j^2}\frac{z}{z^2+\omega_j^2},\tag{A8}$$

which leads to

$$\sum_{j=1}^{n}\frac{c_j^2}{z^2+\omega_j^2}=\sum_{j=1}^{n}\frac{c_j^2}{\omega_j^2}-z\acute\gamma(z).\tag{A9}$$

Using Eqs. (A7) and (A9), the summations over j can be worked out:

$$\mathcal{G}(\tau) = \sum_{l=1}^{n} \frac{u_{l0}^2}{\Omega_l \tanh\left(\frac{1}{2}\beta_b\Omega_l\right)} - \frac{2}{\beta_b}\left\{\omega_0^2\left[\sum_{l=1}^{n}\frac{u_{l0}^2\cos(\Omega_l\tau)}{\Omega_l^2}\right]^2 - \Theta_0^2(\tau)\right\}$$
$$- 2\Theta_0(\tau)\sum_{l=1}^{n}\frac{u_{j0}^2\sin(\Omega_l\tau)}{\tanh\left(\frac{1}{2}\beta_b\Omega_l\right)} - \frac{4}{\beta_b}\sum_{k=1}^{\infty}\left[\omega_0^2 + v^2k^2 + vk\acute{\gamma}(vk)\right]\left[\sum_{l=1}^{n}\frac{u_{l0}^2\cos(\Omega_l\tau)}{v^2k^2+\Omega_l^2}\right]^2$$
$$+ \frac{4}{\beta_b}\sum_{k=1}^{\infty}\left[\sum_{l=1}^{n}\frac{u_{l0}^2\Omega_l\sin(\Omega_l\tau)}{v^2k^2+\Omega_l^2}\right]^2 + \frac{4}{\beta_b}\sum_{k=1}^{\infty}v^2k^2\left[\omega_0^2 + vk\acute{\gamma}(vk)\right]\left[\sum_{l=1}^{n}\frac{u_{l0}^2\sin(\Omega_l\tau)}{\Omega_l\left(v^2k^2+\Omega_l^2\right)}\right]^2. \tag{A10}$$

With the help of the spectral density of normal modes, we finally derive

$$\mathcal{G}(\tau) = \frac{2}{\pi}\int_0^\infty d\Omega\frac{I(\Omega)}{\tanh\left(\frac{1}{2}\beta_b\Omega\right)} - \frac{2}{\beta_b}\left\{\omega_0^2\left[\frac{2}{\pi}\int_0^\infty d\Omega\frac{I(\Omega)\cos(\Omega\tau)}{\Omega}\right]^2 - \Theta_0^2(\tau)\right\}$$
$$- 2\Theta_0(\tau)\left[\frac{2}{\pi}\int_0^\infty d\Omega\frac{\Omega I(\Omega)\sin(\Omega\tau)}{\tanh\left(\frac{1}{2}\beta_b\Omega\right)}\right] - \frac{4}{\beta_b}\sum_{k=1}^{\infty}\left[\omega_0^2 + v^2k^2 + vk\acute{\gamma}(vk)\right]\left[\frac{2}{\pi}\int_0^\infty d\Omega\frac{I(\Omega)\Omega\cos(\Omega\tau)}{v^2k^2+\Omega^2}\right]^2$$
$$+ \frac{4}{\beta_b}\sum_{k=1}^{\infty}\left[\frac{2}{\pi}\int_0^\infty d\Omega\frac{I(\Omega)\Omega^2\sin(\Omega\tau)}{v^2k^2+\Omega^2}\right]^2 + \frac{4}{\beta_b}\sum_{k=1}^{\infty}v^2k^2\left[\omega_0^2 + vk\acute{\gamma}(vk)\right]\left[\frac{2}{\pi}\int_0^\infty d\Omega\frac{I(\Omega)\sin(\Omega\tau)}{v^2k^2+\Omega^2}\right]^2. \tag{A11}$$

Following the same steps, one finds

$$\mathcal{K}(\tau) = \frac{2}{\pi}\int_0^\infty d\Omega\frac{I(\Omega)\Omega^2}{\tanh\left(\frac{1}{2}\beta_b\Omega\right)} - \frac{8}{\pi^2}\left[\int_0^\infty d\Omega I(\Omega)\Omega\cos(\Omega\tau)\right]\left[\int_0^\infty d\Omega\frac{\Omega^2 I(\Omega)\cos(\Omega\tau)}{\tanh\left(\frac{1}{2}\beta_b\Omega\right)}\right]$$
$$- \frac{2}{\beta_b}\left\{\omega_0^2\Theta_0^2(\tau) - \left[\frac{2}{\pi}\int_0^\infty d\Omega I(\Omega)\Omega\cos(\Omega\tau)\right]^2\right\} - \frac{16}{\pi^2\beta_b}\sum_{k=1}^{\infty}\left[\omega_0^2 + v^2k^2 + vk\acute{\gamma}(vk)\right]\left[\int_0^\infty d\Omega\frac{I(\Omega)\Omega^2\sin(\Omega\tau)}{v^2k^2+\Omega^2}\right]^2$$
$$+ \frac{16}{\pi^2\beta_b}\sum_{k=1}^{\infty}\left[\int_0^\infty d\Omega\frac{I(\Omega)\Omega^3\cos(\Omega\tau)}{v^2k^2+\Omega^2}\right]^2 + \frac{16}{\pi^2\beta_b}\sum_{k=1}^{\infty}v^2k^2\left[\omega_0^2 + vk\acute{\gamma}(vk)\right]\left[\int_0^\infty d\Omega\frac{I(\Omega)\Omega\cos(\Omega\tau)}{v^2k^2+\Omega^2}\right]^2. \tag{A12}$$

In our numerical simulations, we set the upper bounds as $\Omega_c = 50\omega_0$ for all integrals over Ω and $k_c = 10$ for all sums over k. The increase of the two upper bounds do not change our physical conclusions.

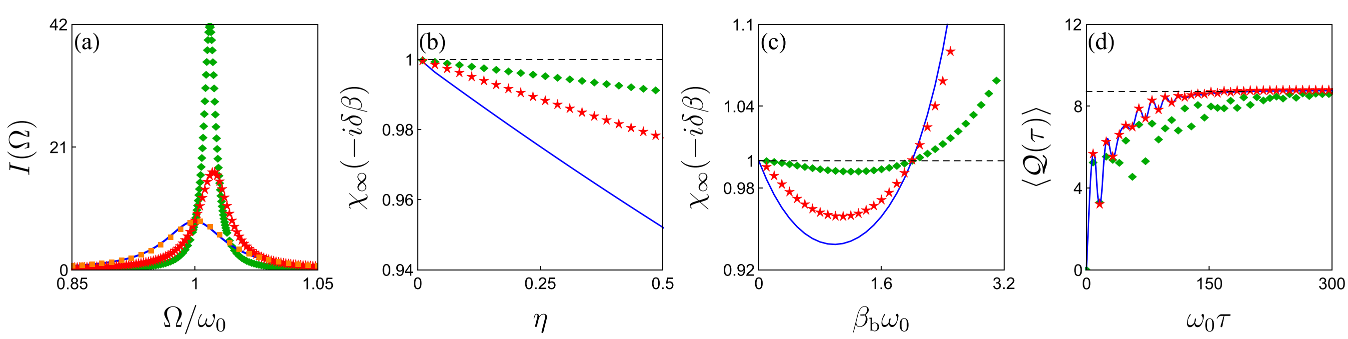

FIG. 3. (a) The spectral density of normal norms $I(\Omega)$ versus Ω with different cutoff frequencies: $\omega_c/\omega_0 = 0.5$ (green diamonds), $\omega_c/\omega_0 = 1$ (red stars), and $\omega_c/\omega_0 = 20$ (orange rectangles). Other parameters are $\eta = 0.06$ and $\omega_0 = 2\text{cm}^{-1}$. The characteristic function $\chi_\infty(-i\delta\beta)$ is plotted as the function of (b) the coupling strength η and (c) the temperature of the heat bath $\omega_0\beta_b$ with different cutoff frequencies: $\omega_c/\omega_0 = 50$ (green diamonds) and $\omega_c/\omega_0 = 500$ (red stars). The temperature of the heat bath in (b) is $\omega_0\beta_b = 0.2$ and the coupling strength in (c) is $\eta = 0.25$. Other parameters are chosen as $\omega_0\beta_s = 2$ and $\omega_0 = 2\text{cm}^{-1}$. (d) The dynamics of the average heat $\langle\mathcal{Q}(\tau)\rangle$ with different cutoff frequencies: $\omega_c/\omega_0 = 0.5$ (green diamonds) and $\omega_c/\omega_0 = 500$ (red stars). Other parameters are chosen as $\eta = 0.05$, $\omega_0\beta_s = 2$, $\omega_0\beta_b = 0.2$, and $\omega_0 = 2\text{cm}^{-1}$. The black dashed line in (d) presents the high-temperature approximate result from Eq. (48). The blue solid lines in (a)–(d) are the results of the special case $f(\omega, \omega_c) = 1$ considered in the main text.

APPENDIX B: LORENTZIAN CUTOFF CASE

In this Appendix, we would like to discuss the case of the Ohmic spectral density with a Lorentzian cutoff, namely, $f(\omega, \omega_c) = \omega_c^2/(\omega^2 + \omega_c^2)$. The corresponding expression of $\acute{\gamma}(z)$ reads $\acute{\gamma}(z) = \eta\omega_c/(z + \omega_c)$. In Fig. 3(a), we plot $I(\Omega)$ with different cutoff frequencies. When ω_c/ω_0 is small, $I(\Omega)$ is underdamped (sharply peaked) while, with the increase of ω_c/ω_0, $I(\Omega)$ becomes overdamped (broad). In the special case $\omega_c \gg \omega_0$, the underdamped spectrum convergences to the one considered in the main text, i.e., the case of $J(\omega) = \eta\omega$. Both underdamped and overdamped baths can result in non-Markovian dynamical effects [82]. With the expression of $I(\Omega)$ at hand, we also compute the characteristic function and the average heat in this Lorentzian-cutoff case. As displayed in Figs. 3(b)–3(d), all the physical conclusions presented in the main text remain unchanged. These results demonstrate our findings are universal to different forms of the original spectral density.

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