Nonequilibrium quantum transport coefficients and transient dynamics of full counting statistics in the strong-coupling and non-Markovian regimes

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Nonequilibrium transport properties of quantum systems have recently become experimentally accessible in a number of platforms in so-called full-counting experiments that measure transient and steady-state nonequilibrium transport dynamics. We show that the effect of the measurement back-action can be exploited to establish general relationships between transport coefficients in the transient regime which take the form of fluctuation-dissipation theorems in the steady state. This result becomes most conspicuous in the transient dynamics of open quantum systems under strong-coupling to non-Markovian environments in nonequilibrium settings. In order to explore this regime, a new simulation method based in a hierarchy of equations of motion has been developed. We instantiate our proposal with the study of energetic conductance between two baths connected via a few level system.

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I. INTRODUCTION

The experimental ability to probe the statistical properties of quantum transport in mesoscopic systems, such as electrons in nanojunctions [1] or cold atoms [2], has provided insights into the nonequilibrium behavior of quantum systems. Theoretical tools for their description have been developed in the form of so-called full counting statistics (FCS) methods [3,4], which constitute a generalization of the theory of moment and cumulant generating functions and is conceptually based on a two-point measurement scheme [4], so that state projection of the bath after the first measurement is automatically incorporated. The effects of this state collapse quickly fade in Markovian baths, allowing for the derivation of steady-state fluctuation theorems that relate several transport coefficients governing the dynamics. In the linear response regime, these are none other than the celebrated fluctuation-dissipation theorems, such as the Kubo formula, or other properties such as the Onsager-Casimir relations. Far from equilibrium, these theorems can be generalized for nonlinear coefficients [5].

Femtosecond laser pulses and other ultrafast control techniques provide access to the statistical response of quantum systems in the transient regime. The Jarzynski equality [6], the Crooks theorem [7,8], and related relationships confirm that steady-state fluctuation-dissipation theorems need not automatically carry over to the transient regime. An interpretation of these effects as the consequence of measurement back-action in the bath supports the intuition that the failure of steady-state relations must be particularly evident in situations where the system-environment coupling is very large or the environmental evolution is so slow that non-Markovian effects become relevant. In this situation, only an explicit computation of the full dynamics is so far known to provide the correct insight into transport properties.

The behavior of non-Markovian, strong-coupling transport settings is captured in the Levitov-Lesovik formula [9] in the case of noninteracting particles. FCS for non-Markovian settings was studied from a general perspective in [10,11], whereas specific treatments include harmonic chains [12], spin-boson or fermionic models in the perturbative regime [13–20], and general bosonic or fermionic systems for the first and/or second moments of the dynamics [21–25]. Bath statistics of open quantum systems has provided access to universal oscillations in high order cumulants [26] and the Kondo signature in the spin-boson model [27] and fermionic models [28]. FCS measurement strategies are attracting renewed attention [29] together with optimized cumulant evaluation methods [30]. Additionally, discussion of classical and quantum initial correlation effects in shot noise has been addressed in [31] and thermodynamic consistency of FCS simulation methods has been studied in [32].

Here we provide an alternative approach, which consists of quantitatively computing the deviation of the Saito-Utsumi coefficient relations [5] when applied in transient situations. We show that this deviation is directly associated with a physical picture where no part of the system-environment compound is initially subject to measurement, and is expressed in terms of a natural symmetry that affects the cumulant generating function. In order to investigate these effects, we have developed a simulation method that incorporates FCS into the formalism of hierarchy of equations of motion (HEOM) [33], an established method for the simulation of general, multilevel open quantum systems (OQS) that is nonperturbative in the coupling strength and which faithfully represents non-Markovian effects of the environment. With this method we gain access to cumulants of any desired order of environmental energetic and particle observables and arbitrary time dependence of the Hamiltonian may be treated. As a first example, we consider an open quantum system that couples to a bosonic heat bath, although this procedure is equally valid for fermionic baths. This method generalizes previous attempts that involved first moments [34,35] and to our knowledge it is the first time that high order cumulants are simulated with this formalism.

We first introduce the FCS formalism and discuss a generalization thereof that allows for the isolation of the transient measurement back-action. A relationship between the back-action effect and transport coefficients is presented that holds both close to equilibrium and far from it. We further introduce the simulation method developed for the investigation of these transient effects, which is based on

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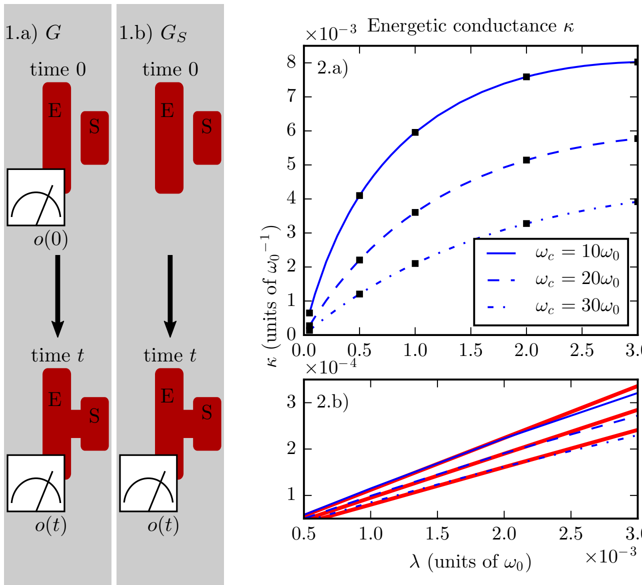

FIG. 1. (1) Depiction of measurement schemes associated with Eq. (1) (1.a) and Eq. (4) (1.b), respectively. In both measurement schemes a separable initial state of the form $\pi(A)$ is assumed. (2) Steady-state energetic conductance as a function of the coupling strength λ and for different spectral density cutoff frequencies ω_c. (2.a) Lines show results computed via numerical derivation for $(T_R - T_L)/T_R = 0.01$. Dots are computed following Eq. (10) with no temperature bias. Other parameters are $T_L = T_R = 10\omega_0$, $J = 0$. All values have been numerically converged by varying the hierarchical depth of the simulation. (2.b) Closeup of the weak coupling limit, where red solid lines reproduce the analytical prediction of the weak coupling theory.

the HEOM formalism. We finally present simulation results on a bosonic transport setting and show that the energetic conductance can be reliably accessed through investigation of measurement back-action effects.

II. STEADY-STATE COEFFICIENT RELATIONS

The formalism for the analysis of full counting experiments constitutes a well-established theoretical framework [4] involving a two point measurement prescription: the value of a specific observable of interest $\hat{O}$ is measured at an initial time $t = 0$, where a result $o(0)$ is obtained, and at a final time $t > 0$, with an outcome $o(t)$ [see Fig. 1(1)]. Repetition of the experiment generates statistics of the measurement difference $\Delta o(t) = o(t) - o(0)$, which can be treated as a stochastic variable. Although more general cases can be considered, let us regard for simplicity a single measured operator and an initial state of the total system $\pi(A) = \rho \otimes \frac{\exp(-A\hat{O})}{\mathrm{Tr}\,\{\exp(-A\hat{O})\}}$, where ρ is an arbitrary state of the subsystem where the measurement has no effect and A is a thermodynamic constraint fixing the initial expected value of $\hat{O}$. An instance of such a setting is a system in contact with several baths, where $\hat{O}$ is the Hamiltonian of one of the baths, A is its initial inverse temperature, and ρ is an arbitrary state of the system and all nonmeasured baths.

Under these conditions, the cumulant generating function (CGF) takes the form [4]

$$G(\chi, A, t) = \ln\langle e^{i\chi\hat{O}(t)} e^{-i\chi\hat{O}(0)}\rangle_A, \tag{1}$$

with $\langle\bullet\rangle_A \equiv \mathrm{Tr}\,\{\bullet\pi(A)\}$. Its Taylor-expansion coefficients in the counting field χ correspond to the cumulants of the measurement difference $\Delta o(t)$. In the steady-state limit, the cumulant generating function for the currents $\mathcal{F}(\chi, A) \equiv \lim_{\tau\to\infty} \frac{1}{\tau} G(\chi, A, \tau)$ often fulfills the symmetry

$$\mathcal{F}(\chi, A) = \mathcal{F}(-\chi + iA, A), \tag{2}$$

also known as fluctuation theorem, so that transport coefficients $L^n_m(A) \equiv \frac{\partial^{m+n}}{\partial(i\chi)^n \partial A^m} \mathcal{F}(\chi, A)\big|_{\chi=0}$ obey the Saito-Utsumi relations [5]

$$L^n_m(A) = \sum_{j=0}^{m} \binom{m}{j} (-1)^{n+j} L^{n+j}_{m-j}(A). \tag{3}$$

Relations such as the Kubo formula and the Onsager-Casimir relations can be recast as specific cases of this equation, in particular when several counting fields χ_k are involved and the associated thermodynamic constraints are close to an equilibrium state $A_k \simeq A$. Nevertheless, Eq. (3) is generally not valid in the transient regime.

III. RELATIONS FOR TRANSIENT TRANSPORT COEFFICIENTS

With the aim of quantifying the error of the fluctuation theorem Eq. (2) in the transient dynamics, one may define the difference between two CGFs:

$$G_S(\chi, A, t) \equiv \ln\langle e^{i\chi\hat{O}(t)}\rangle_A - \ln\langle e^{i\chi\hat{O}(0)}\rangle_A, \tag{4}$$

each associated with a single (S) measurement of the operator $\hat{O}$ given an initial state of the form $\pi(A)$ [see Fig. 1(1)]. Its χ derivatives provide the difference of the cumulants of the measurement outcome between two times t and $t = 0$. Note the subtle difference in the statistical interpretation of functions Eqs. (1) and (4). As shown in Appendix A, both functions are related by the expression

$$G_S(\chi, A, t) = G(\chi, A - i\chi, t). \tag{5}$$

Although this equation relates two physically distinct situations (two different measurement schemes), it bears a resemblance with Eq. (2) that can be exploited to obtain relations similar to Eq. (3) for the objects $J_{(S)}{}^n_m(A,t) \equiv \frac{\partial^{m+n}}{\partial(i\chi)^n \partial A^m} G_{(S)}(\chi, A, t)\big|_{\chi=0}$,

$$J_S{}^n_m(A,t) = \sum_{j=0}^{n} \binom{n}{j} (-1)^j J^{n-j}_{m+j}(A,t). \tag{6}$$

Note that $L^n_m(A) = \lim_{\tau\to\infty} \frac{1}{\tau} J^n_m(A,\tau)$, and this can be used to recover Eq. (3) in the steady state.

This is a powerful relationship that establishes analogies of fluctuation-dissipation theorems on the transient dynamics by quantifying the deviation from the steady-state expressions in terms of the values $J_S{}^n_m(A,t)$, which correspond to measurable quantities in a well defined physical setting. The amplitude of these deviations grows with the strength of the coupling between the measured partition and the rest of the system, and also with the duration of the transient dynamics, which is associated with non-Markovian effects in the language of open quantum systems. We explore below these effects at hand of specific examples.

IV. APPLICATIONS

In order to instantiate the relations in Eq. (6), let us consider an open quantum system of Hamiltonian $\hat{\mathrm{H}}_{\mathrm{S}}$ interacting via operators $\hat{\mathrm{V}}_\nu$ with one or several baths, so that the total Hamiltonian has the form

$$\hat{\mathrm{H}} = \hat{\mathrm{H}}_{\mathrm{S}} + \sum_\nu \hat{\mathrm{V}}_\nu \otimes \hat{\mathrm{B}}_\nu + \hat{\mathrm{H}}_{\mathrm{B}}, \tag{7}$$

$$\hat{\mathrm{H}}_{\mathrm{B}} = \sum_\nu \hat{\mathrm{H}}_\nu = \sum_{\nu,k} \omega_{\nu,k} \hat{\mathrm{a}}^\dagger_{\nu k}\hat{\mathrm{a}}_{\nu k}, \tag{8}$$

where $\hat{\mathrm{B}}_\nu$ is an arbitrary operator of bath ν and $\hat{\mathrm{a}}_{\nu,k}$ and $\hat{\mathrm{a}}^\dagger_{\nu,k}$ are the usual bosonic or fermionic annihilation and creation operators for the mode k of frequency $\omega_{\nu,k}$. This encompasses a broad range of dissipative and transport settings.

The energy of one of the baths $\hat{\mathrm{H}}_\nu$ provides relevant information about the heat flows in the system. The thermodynamic constraint β_ν associated with it is the inverse temperature of the bath, so that Eq. (6) for $m = 0$ takes the form

$$J_{S_0}^{\,n}(\beta_\nu,t) = \sum_{j=0}^{n} \binom{n}{j} (-1)^j J^j_{n-j}(\beta_\nu,t), \tag{9}$$

which relates the cumulants of the bath energies with high order instantaneous energetic conductances. It has the same form as the case $n = 0$ of Eq. (3) except for the *deviation* term $J_{S_0}^{\,n}$ and an extra minus sign for odd n.

For a two-bath setting ($\nu \in \{R,L\}$), we define a first order *energetic transport coefficient* as $\kappa_R(t;T_R,T_L) \equiv \beta_R^2 \frac{\partial}{\partial t} J^1_1(\beta_R,t)$,[1] whose steady-state, equilibrium limit *energetic conductance* $\kappa \equiv \lim_{t\to\infty} \lim_{T_R\to T_L} \kappa_R(t;T_R,T_L)$ can be understood in turn as a generalization of the concept of thermal conductance for the case of strong-coupling and non-Markovian regimes. Equation (9) for $n = 2$ establishes a relationship between the first order energetic transport coefficient and the second order cumulants of the form

$$\kappa_R(t;T_R,T_L) = \frac{\beta_R^2}{2}\frac{\partial}{\partial t}\left[J_0^2(\beta_R,t) - J_{S_0}^{\,2}(\beta_R,t)\right], \tag{10}$$

which is analogous to the Kubo formula except for the deviation term $J_{S_0}^{\,2}$. As in the case of the fluctuation-dissipation theorem, it puts forward the possibility to derive a conductance value from the difference of the fluctuations as obtained from a two measurement scheme and a single measurement scheme. This is a way to circumvent the necessity of a numerical derivative and avoids the accumulation of numerical error implicit in the choice of any small but finite temperature bias.

[1]Please note the special choice of sign, such that subsequent numerical results remain positive.

V. NUMERICAL METHOD

In order to address the strong-coupling and non-Markovian regimes, we developed a hierarchy of equations of motion for the simulation of full counting statistics and high-order moments of relevant bath observables. The technique enables statistical analysis of any bath observable that commutes with its free Hamiltonian term, and we provide examples for the case of the energy of the bath $\hat{\mathrm{H}}_{\mathrm{B}}$. In particular, any linear combination of the operators $\hat{\mathrm{a}}^\dagger_{\nu k}\hat{\mathrm{a}}_{\nu k}$ such as the particle number $\hat{\mathrm{N}}$ can be simulated with this approach. Furthermore, it is flexible enough to generate both the moments corresponding to the two-measurement picture Eq. (1) and the single measurement picture Eq. (4), and we will use it in both modes and apply the relationship Eq. (6) to obtain nonequilibrium transport coefficients.

A central element in the derivation of the method is the counting-field-resolved bath correlation function

$$C_\nu^{jk}(\chi,t) = (-)^{j+k}\left\langle \tilde{\hat{\mathrm{B}}}{}^j_\nu\left[(-)^j\frac{\chi}{2}\right](t)\,\tilde{\hat{\mathrm{B}}}{}^k_\nu\left[(-)^k\frac{\chi}{2}\right](0)\right\rangle, \tag{11}$$

where superindices j and k take two values 0 or 1 and indicate the side an operator acts from: $\hat{\mathrm{A}}^0\rho \equiv \hat{\mathrm{A}}\rho$ or $\hat{\mathrm{A}}^1\rho \equiv \rho\hat{\mathrm{A}}$. The tilde indicates the interaction picture with respect to $\hat{\mathrm{H}}_\nu$, the dressing of an operator by the counting operator $\hat{\mathrm{O}}$ is denoted by $\hat{\mathrm{A}}[\chi](t) = e^{i\chi\hat{\mathrm{O}}}\hat{\mathrm{A}}(t)e^{-i\chi\hat{\mathrm{O}}}$ and the average is over the initial state of the bath. The hierarchy is based on a decomposition of the correlation function $C_\nu^{jk}(\chi,t) = \sum_r c^{jk}_{\nu r}(\chi)\phi_r(t)$ by means of a set of functions $\{\phi_r(t)\}$ whose derivatives are well defined within the set by $\frac{d}{dt}\phi_r(t) = \sum_r \eta_{rs}\phi_s(t)$ [36]. The set size determines the growth of the simulation requirements, traditionally making this method indicated for not too low temperature regimes. Nevertheless, a judicious choice of the basis can circumvent this limitation [36].

We refer the reader to Appendixes B and C for the technical aspects of the method. It involves the propagation of so-called *auxiliary fields*$\sigma^{\{n\}}_{\{m\}}(t)$, labeled by a rank-three tensor $\{n\}$ and a vector $\{m\}$ of nonnegative integer entries whose elements respectively sum up to the hierarchic level n and represent a partition of the moment order m. The zeroth hierarchic level contains the auxiliary field $\sigma^{\{0\}}_{\{0\}}(t)$, corresponding to the system density matrix $\rho(t)$, and $\sigma^{\{0\}}_{\{m\}}(t)$, a linear combination of which [defined in Appendix C, Eq. (C3)] can be traced to obtain the mth moment. The auxiliary fields satisfy the equation

$$\begin{aligned}\frac{d}{dt}\sigma^{\{n\}}_{\{m\}}(t) &= -i\hat{\mathrm{H}}_S^\times\sigma^{\{n\}}_{\{m\}}(t) + \sum_{\nu,r;k=0,1}\Bigg(\hat{\bar{V}}{}^k_{\nu r}\sigma^{\{\dots,n^k_{\nu r}+1,\dots\}}_{\{m\}}(t) \\ &\quad + \sum_s n^k_{\nu r}\eta_{rs}\sigma^{\{\dots,n^k_{\nu r}-1,\dots,n^k_{\nu s}+1,\dots\}}_{\{m\}}(t) \\ &\quad + n^k_{\nu r}\phi_r(0)\hat{\mathrm{V}}^k_\nu\sigma^{\{\dots,n^k_{\nu r}-1,\dots\}}_{\{m\}}(t) \\ &\quad + \sum_q m_q\,\hat{\bar{V}}{}^k_{\nu rq}\sigma^{\{\dots,n^k_{\nu r}+1,\dots\}}_{\{\dots,m_q-1,\dots\}}(t)\Bigg),\end{aligned} \tag{12}$$

where $\hat{\hat{V}}^k_{\nu r} \equiv \sum_{j=0,1} c^{jk}_{\nu r}(0)\hat{V}^j_\nu$, $\hat{\hat{V}}^k_{\nu rq} \equiv \sum_{j=0,1} c^{jk}_{\nu rq}\hat{V}^j_\nu$, and $c^{jk}_{\nu rq} \equiv \frac{d^q}{d(i\chi)^q} c^{jk}_{\nu r}(\chi)|_{\chi=0}$. The structure is identical to the usual hierarchy [33] but for the last term, which connects it to the hierarchy associated with the previous moment, which is compatible with results in the Markovian, weak-coupling case [30]. A cutoff at a maximum hierarchic level $n_{\max}$ is usually justified in terms of numerical convergence and is roughly proportional to the system-bath coupling strength.

In order to demonstrate the numerical validity of Eq. (10), let us consider a two level system with Hamiltonian $\hat{H}_S = \omega_0\sigma_x + J\sigma_z$, where σ_i with $i \in \{x,y,z\}$ are the Pauli matrices, coupled to two bosonic baths $\nu \in \{R,L\}$ via $\hat{V}_\nu = \sigma_z$ and $\hat{B}_\nu = \sum_k \gamma_{\nu k}(\hat{a}_{\nu k} + \hat{a}^\dagger_{\nu k})$, characterized by a spectral density $J_\nu(\omega) = \sum_k \gamma^2_{\nu k}\delta(\omega - \omega_{\nu k})$ and an inverse temperature β_ν. The following results are derived for $\rho(0) = \frac{\sigma_x+1}{2}$ and the choice of an Ohmic spectral density with exponential cutoff $J(\omega) = \frac{\lambda}{\omega_c}\omega e^{-\frac{\omega}{\omega_c}}$, where $\lambda = \int_0^\infty \frac{J(\omega)}{\omega} d\omega$ is the reorganization energy and ω_c is the scale of the cutoff.

Energetic conductance values are shown in Fig. 1(1.b). Following the discussion presented above, two methods are used for their computation. On the one hand, simulations under small temperature bias $(T_R - T_L)/T_R = 0.01$ are run in order to obtain steady-state energy currents, which are related to the energetic conductance through the definition of $J^1_1(\beta_R,t)$. On the other hand, second order moments computed in the two-measurement and the single-measurement schemes for equilibrium conditions ($T_L = T_R$) are further used to derive conductance values as per Eq. (10). Results show excellent agreement between both pictures and also approach predictions from the weak coupling theory and the *noninteracting blip approximation* (NIBA) [13] in their respective regimes of validity. Whereas a linear increase of conductance with the coupling strength λ is expected in the weak coupling limit, a turnover is reproduced for higher coupling strengths. The relationship Eq. (10) is valid also in the transient regime and far from equilibrium as shown in Fig. 2. In this case, the steady-state energetic conductance of a biased two level system ($J \neq 0$) is studied in situations where $T_R - T_L \simeq T_L$. The transient dynamics are also accurately reproduced by Eq. (10), where the oscillating effect is introduced by the tunneling, which acts as an effective driving.

Finally, the transient deviation of high order transport coefficient relations Eq. (3) is shown in Fig. 3 for a range of coupling constants λ and spectral density cutoffs ω_c. It constitutes a quantitative confirmation that the failure of steady-state fluctuation-dissipation theorems is proportional to the coupling strength to the bath and lasts longer for higher degrees of non-Markovianity. Additionally, the sign of the deviation changes for odd orders, as predicted from the comparison of Eqs. (3) and (10).

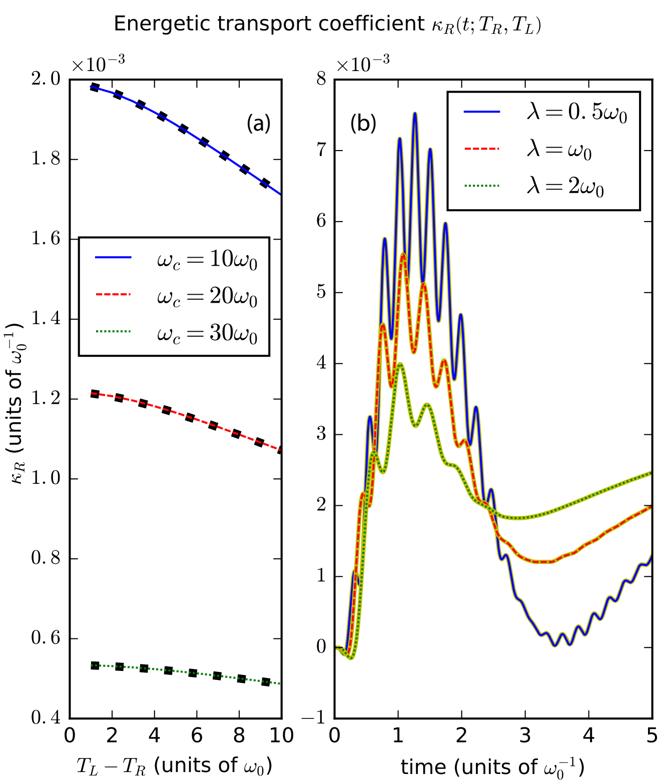

FIG. 2. (a) Steady-state energetic transport coefficient $\lim_{t\to\infty}\kappa_R(t;T_R,T_L)$ as a function of the temperature of the left bath T_L and for different spectral density cutoff frequencies ω_c. Other parameters are $T_R = 10\omega_0$, $J = \omega_0$, $\lambda = \omega_0$. Lines show results computed via numerical derivation of steady-state currents. Dots are computed following Eq. (10). (b) Transient dynamics of the energetic transport coefficient as a function of time and for different reorganization energies λ. Other parameters are $T_R = T_L = 10\omega_0$, $J = \omega_0$, $\omega_c = 3\omega_0$. Results shown in blue are computed via numerical derivation of transient currents. Red lines are computed following Eq. (10).

VI. CONCLUSIONS

When considering the transient transport dynamics of non-Markovian systems, it is possible to quantify deviations from fluctuation-dissipation theorems and the nonlinear Saito-Utsumi relations, which are only valid in the steady state. These deviations have a physical interpretation and are associated with equilibration dynamics of the same system under a different measurement scheme. We demonstrate this relation by developing a tool that allows for the simulation of the full counting statistics of a broad range of bath observables under dissipative and nonequilibrium settings, which is a generalization of the celebrated hierarchy of equations of motion for non-Markovian and strong-coupling settings. By accessing high order cumulants of the bath energy, it is possible to derive energetic conductances and higher order derivatives thereof while, at the same time, avoiding finite bias simulations. This approach is immediately applicable to the study of observables such as the particle number, environments of fermionic nature, more complex and higher dimensional systems, and time-dependent driving.

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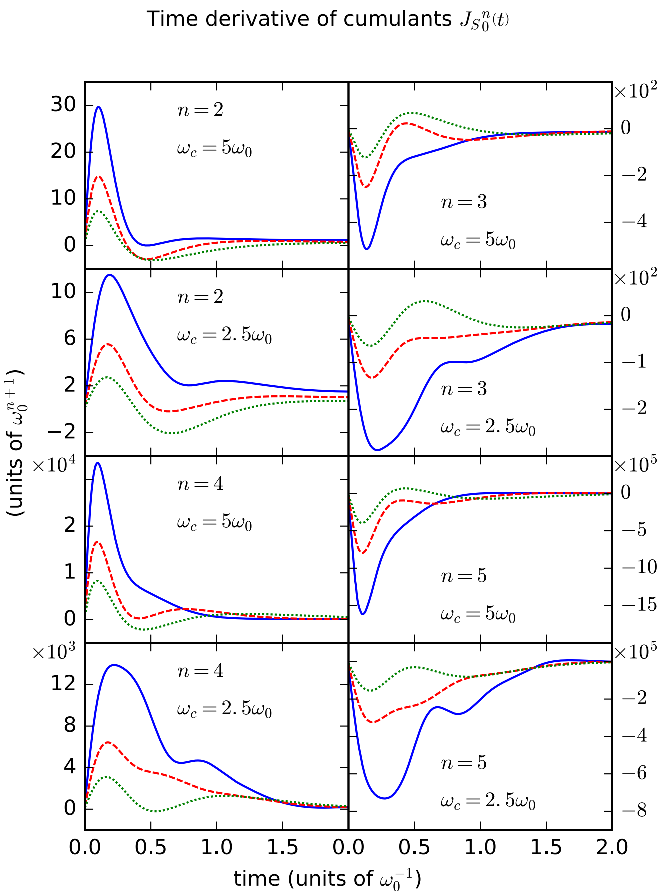

FIG. 3. Transient dynamics of the time derivatives of the first five cumulants of the bath energy $\hat{H}_R$ in the single measurement scheme (S) for different values of the cutoff frequency ω_c and the reorganization energy λ: $\lambda = 0.05\omega_c$ for the dotted lines, $\lambda = 0.1\omega_c$ for the dashed lines, and $\lambda = 0.2\omega_c$ for the solid lines. Other parameter values are $T_L = T_R = 10\omega_0$, $J = 0$.

APPENDIX A: RELEVANT LIMITS OF THE CUMULANT GENERATING FUNCTION

Let us reproduce the definition of the cumulant generating function

$$G(\chi,A,t) \equiv \ln\langle e^{i\chi\hat{O}}(t)e^{-i\chi\hat{O}}(0)\rangle_A$$
$$= \ln \mathrm{Tr}\{e^{i\chi\hat{O}}(t)e^{-i\chi\hat{O}}(0)\pi(A)\}, \quad \text{(A1)}$$

with $\pi(A) = \rho \otimes \frac{e^{-A\hat{O}}}{\mathrm{Tr}\{e^{-A\hat{O}}\}}$ a separable initial state of the total system, where ρ is an arbitrary state of the subsystem where the measurement has no effect and A is a thermodynamic constraint fixing the initial expected value of $\hat{O}$. Evaluating the second variable of the cumulant generating function at $A - i\chi$ one obtains

$$G(\chi,A-i\chi,t) = \ln \mathrm{Tr}\{e^{i\chi\hat{O}}(t)\pi(A)\} - \ln\frac{\mathrm{Tr}\{e^{(-A+i\chi)\hat{O}}\}}{\mathrm{Tr}\{e^{-A\hat{O}}\}}$$
$$= \ln\langle e^{i\chi\hat{O}}(t)\rangle_A - \ln\langle e^{i\chi\hat{O}}(0)\rangle_A. \quad \text{(A2)}$$

One may interpret this function as the difference between two single-measurement cumulant generating functions: one where the measurement takes place at time t and another one where the measurement takes place at time $t = 0$. We denote it by

$$G_S(\chi,A,t) \equiv \ln\langle e^{i\chi\hat{O}}(t)\rangle_A - \ln\langle e^{i\chi\hat{O}}(0)\rangle_A. \quad \text{(A3)}$$

APPENDIX B: COUNTING-FIELD-RESOLVED HIERARCHY OF EQUATIONS OF MOTION

As indicated in the main text [Eqs. (7) and (8)], let us consider a general total Hamiltonian consisting of a system, interaction, and bath parts

$$\hat{H} = \hat{H}_S + \sum_\nu \hat{V}_\nu \otimes \hat{B}_\nu + \hat{H}_B, \quad \text{(B1)}$$
$$\hat{H}_B = \sum_\nu \hat{H}_\nu = \sum_{\nu,k} \omega_{\nu,k}\hat{a}^\dagger_{\nu k}\hat{a}_{\nu k}. \quad \text{(B2)}$$

The index ν labels the baths, $\hat{a}_{\nu,k}$ and $\hat{a}^\dagger_{\nu,k}$ are the usual bosonic or fermionic annihilation and creation operators for the mode k in bath ν. The goal is to derive the full counting cumulant generating function Eq. (1) of an observable $\hat{O}$ of one of the baths which commutes with its free Hamiltonian $\hat{H}_\nu$ and with the initial state of the system and baths $\pi(0) = \rho(0)\bigotimes_\nu \frac{e^{-\beta_\nu\hat{H}_\nu}}{\mathrm{Tr}\{e^{-\beta_\nu\hat{H}_\nu}\}}$. For simplicity we will omit the explicit dependence on the thermodynamic constraint in the derivation, so that the two-measurement cumulant generating function takes the form

$$G(\chi,t) = \ln \mathrm{Tr}\left\{e^{-i\hat{H}[\frac{\chi}{2}]t}\pi(0)e^{i\hat{H}[-\frac{\chi}{2}]t}\right\}, \quad \text{(B3)}$$

where $\hat{A}[\chi](t) = e^{i\chi\hat{O}}\hat{A}(t)e^{-i\chi\hat{O}}$. This problem can be formulated in terms of the solution to a hierarchy of equations of motion for the counting field resolved density matrix

$$\rho(\chi,t) = \mathrm{Tr}_B\left\{e^{-i\hat{H}[\frac{\chi}{2}]t}\pi(0)e^{i\hat{H}[-\frac{\chi}{2}]t}\right\}, \quad \text{(B4)}$$

and the equation $G(\chi,t) = \ln[\mathrm{Tr}\{\rho(\chi,t)\}]$ directly relates both quantities.

For the sake of clarity, we will derive the equations of motion for Eq. (B4). The case associated with Eq. (A3) immediately follows under modification of the initial state of the bath with the imaginary inverse temperature $\beta_\nu - i\chi$. The matrix $\rho(\chi,t)$ satisfies the differential equation

$$\frac{d}{dt}\rho(\chi,t) = -i\,\mathrm{Tr}_B\left\{\hat{H}\left[\frac{\chi}{2}\right]\pi(\chi,t) - \pi(\chi,t)\hat{H}\left[-\frac{\chi}{2}\right]\right\}, \quad \text{(B5)}$$

with $\pi(\chi,t) \equiv e^{-i\hat{H}[\frac{\chi}{2}]t}\pi(0)e^{i\hat{H}[-\frac{\chi}{2}]t}$. The formal solution of Eq. (B5) in the interaction picture with respect to $\hat{H}_S + \hat{H}_B$ (denoted by an overhead tilde) can be obtained by means of Wick's theorem. For simplicity, we will focus on the derivation for the bosonic case, but all steps can be trivially generalized for the fermionic case. Wick's theorem simplifies the calculation of the partial trace of the bath by reducing products of $2n$ operators to n products of pairwise traces

$$\langle \hat{T}\tilde{\hat{B}}_\nu(t_{2n})\tilde{\hat{B}}_\nu(t_{2n-1})\cdots\tilde{\hat{B}}_\nu(t_2)\tilde{\hat{B}}_\nu(t_1)\rangle = \sum_{\text{app}}\prod_{ij}\langle\hat{T}\tilde{\hat{B}}_\nu(t_i)\tilde{\hat{B}}_\nu(t_j)\rangle, \quad \text{(B6)}$$

where the sum is over all possible pairs (app) of indices up to $2n$, $\hat{\mathrm{T}}$ is the time ordering operator and the average may be performed over any Gaussian state. Therefore, the solution of Eq. (B5) in the interaction picture is $\tilde{\rho}(\chi,t) = \tilde{\mathcal{U}}(\chi,t)\rho(0,0)$, with

$$\tilde{\mathcal{U}}(\chi,t) = \prod_{\nu} \prod_{jk=0,1} \exp_{+}\left(\int_0^t ds\, \tilde{\hat{\mathrm{W}}}_{\nu}^{jk}(\chi,s)\right), \quad \text{(B7)}$$

where $\exp_{+}$ stands for the time ordered exponential and

$$\tilde{\hat{\mathrm{W}}}_{\nu}^{jk}(\chi,t) = -\int_0^t ds\, \tilde{\hat{\mathrm{V}}}_{\nu}^{j}(t) C_{\nu}^{jk}(\chi,t-s)\, \tilde{\hat{\mathrm{V}}}_{\nu}^{k}(s). \quad \text{(B8)}$$

Here we introduce the superoperator notation $\hat{\mathrm{A}}^{0}\rho \equiv \hat{\mathrm{A}}\rho$ and $\hat{\mathrm{A}}^{1}\rho \equiv \rho\hat{\mathrm{A}}$ and the bath correlation functions

$$C_{\nu}^{jk}(\chi,t) = (-)^{j+k}\left\langle \tilde{\hat{\mathrm{B}}}_{\nu}^{j}\left[(-)^{j}\frac{\chi}{2}\right](t)\, \tilde{\hat{\mathrm{B}}}_{\nu}^{k}\left[(-)^{k}\frac{\chi}{2}\right](0)\right\rangle, \quad \text{(B9)}$$

where $\langle A\rangle = \mathrm{Tr}\,\{A\pi(0)\}$. Note that this definition should be replaced by $\langle A\rangle = \mathrm{Tr}\,\{A\pi(0)e^{i\chi\hat{\mathrm{O}}}\,\mathrm{Tr}\,\{e^{i\chi\hat{\mathrm{O}}}\pi(0)\}^{-1}\}$ for Eq. (A3).

The HEOM formalism uses an approximate representation of the correlation functions by means of linear combinations of decaying exponential functions. The extended version generalizes the framework to more involved functional bases [36]. In our case, the coefficients of the linear combination are functions dependent on the counting field χ, so that we approximate $C_{\nu}^{jk}(\chi,t) = \sum_r c_{\nu r}^{jk}(\chi)\phi_r(t)$ by means of a set of functions $\{\phi_r(t)\}$ whose derivatives are well defined within the set by $\frac{d}{dt}\phi_r(t) = \sum_n \eta_{rs}\phi_s(t)$, where η is a matrix with complex entries. The form of $c_{\nu r}^{jk}(\chi)$ is general and depends on the specific observable of interest $\hat{\mathrm{O}}$. For instance, in the case $\hat{\mathrm{O}} = \mathrm{H}_{\nu}$ and $\hat{\mathrm{B}}_{\nu} = \sum_k \gamma_{\nu k}(\hat{\mathrm{a}}_{\nu k} + \hat{\mathrm{a}}_{\nu k}^{\dagger})$, $C_{\nu}^{jk}(\chi,t) = C_{\nu}^{jk}(\chi \pm t)$ and the dependence is expected to be similar to that of $\phi_r(t)$. With this representation, it is possible to define the auxiliary objects

$$\tilde{\rho}^{\{n\}}(\chi,t) = \hat{\mathrm{T}} \prod_{\nu,r;k=0,1}\left(\int_0^t ds\phi_r(t-s)\, \tilde{\hat{\mathrm{V}}}_{\nu}^{k}(s)\right)^{n_{\nu r}^{k}} \times \tilde{\mathcal{U}}(\chi,t)\rho(0,0), \quad \text{(B10)}$$

where $\{n\} \equiv \{\ldots,n_{\nu r}^{k},\ldots\}$ is a rank-three tensor of nonnegative integer entries which sum up to n and n is the so-called *hierarchic level*. It is clear that $\tilde{\rho}^{\{0\}}(\chi,t) = \tilde{\rho}(\chi,t)$ and the auxiliary fields satisfy the equation

$$\begin{aligned}\frac{d}{dt}\rho^{\{n\}}(\chi,t) &= -i\hat{\mathrm{H}}_S^{\times}\rho^{\{n\}}(\chi,t) + \sum_{\nu,r;k=0,1}\Bigg(\bar{\hat{\mathrm{V}}}_{\nu r}^{k}\,\rho^{\{\ldots,n_{\nu r}^{k}+1,\ldots\}}(\chi,t) \\ &\quad + \sum_s n_{\nu r}^{k}\eta_{rs}\rho^{\{\ldots,n_{\nu r}^{k}-1,\ldots,n_{\nu s}^{k}+1,\ldots\}}(\chi,t) \\ &\quad + n_{\nu r}^{k}\phi_r(0)\hat{\mathrm{V}}_{\nu}^{k}\rho^{\{\ldots,n_{\nu r}^{k}-1,\ldots\}}(\chi,t)\Bigg),\end{aligned} \quad \text{(B11)}$$

where we have used the notation $\hat{\mathrm{A}}^{\times}\rho \equiv \hat{\mathrm{A}}\rho - \rho\hat{\mathrm{A}}$ and $\bar{\hat{\mathrm{V}}}_{\nu r}^{k} \equiv \sum_{j=0,1} c_{\nu r}^{jk}(\chi)\hat{\mathrm{V}}_{\nu}^{j}$. This is an extension of the usual HEOM formulation [33].

APPENDIX C: HIERARCHY OF EQUATIONS OF MOTION FOR HIGH ORDER STATISTICAL MOMENTS

Although Eq. (B11) can be used on its own to obtain the generating function, in the case where one is interested in specific statistical moments, a specialized hierarchy can be derived. Let us define the object

$$\begin{aligned}\tilde{\sigma}_m(t) &\equiv \frac{\partial^m}{\partial(i\chi)^m}\tilde{\rho}^{\{0\}}(\chi,t)\Big|_{\chi=0} \\ &= \hat{\mathrm{T}}\frac{\partial^m}{\partial(i\chi)^m}\tilde{\mathcal{U}}(\chi,t)\Big|_{\chi=0}\rho(0,0),\end{aligned} \quad \text{(C1)}$$

so that the moment m of the full counting distribution may be obtained by tracing: $\mathrm{Tr}\,\{\tilde{\sigma}_m(t)\}$. It contains correlation functions of the form $\frac{\partial^q}{\partial(i\chi)^q}C_{\nu}^{jk}(\chi,t)|_{\chi=0} \equiv C_{\nu q}^{jk}(t)$, which are well defined in terms of the approximate representation as $C_{\nu q}^{jk}(t) = \sum_r c_{\nu rq}^{jk}\phi_r(t)$ with $c_{\nu rq}^{jk} \equiv \frac{d^q}{d(i\chi)^q}c_{\nu r}^{jk}(\chi)|_{\chi=0}$. In a procedure analogous to the one followed for the obtention of Eq. (B11), we define

$$\tilde{\sigma}_{\{m\}}^{\{n\}}(t) = \hat{\mathrm{T}}\prod_{\nu,k;j=0,1}\left(\int_0^t ds\phi_k(t-s)\,\tilde{\hat{\mathrm{V}}}_{\nu}^{j}(s)\right)^{n_{\nu k}^{j}} \times \prod_q\left(\int_0^t ds\,\tilde{\hat{\mathrm{W}}}_{\nu q}(s)\right)^{m_q}\tilde{\mathcal{U}}(0,t)\rho(0,0), \quad \text{(C2)}$$

where $\{m\} \equiv \{\ldots,m_q,\ldots\}$ is a vector of nonnegative integer entries such that $\sum_q m_q q = m$ and $\tilde{\hat{\mathrm{W}}}_{\nu q}(t) \equiv \sum_{j,k=0,1} -\int_0^t ds\,\tilde{\hat{\mathrm{V}}}_{\nu}^{j}(t)C_{\nu q}^{jk}(t-s)\,\tilde{\hat{\mathrm{V}}}_{\nu}^{k}(s)$. This object satisfies the equation

$$\begin{aligned}\frac{d}{dt}\sigma_{\{m\}}^{\{n\}}(t) &= -i\hat{\mathrm{H}}_S^{\times}\sigma_{\{m\}}^{\{n\}}(t) + \sum_{\nu,r;k=0,1}\Bigg(\bar{\hat{\mathrm{V}}}_{\nu r}^{k}\,\sigma_{\{m\}}^{\{\ldots,n_{\nu r}^{k}+1,\ldots\}}(t) \\ &\quad + \sum_s n_{\nu r}^{k}\eta_{rs}\sigma_{\{m\}}^{\{\ldots,n_{\nu r}^{k}-1,\ldots,n_{\nu s}^{k}+1,\ldots\}}(t) \\ &\quad + n_{\nu r}^{k}\phi_r(0)\hat{\mathrm{V}}_{\nu}^{k}\sigma_{\{m\}}^{\{\ldots,n_{\nu r}^{k}-1,\ldots\}}(t) \\ &\quad + \sum_q m_q\,\bar{\hat{\mathrm{V}}}_{\nu rq}^{k}\,\sigma_{\{\ldots,m_q-1,\ldots\}}^{\{\ldots,n_{\nu r}^{k}+1,\ldots\}}(t)\Bigg),\end{aligned}$$

where $\bar{\hat{\mathrm{V}}}_{\nu rq}^{k} \equiv \sum_{j=0,1} c_{\nu rq}^{jk}\hat{\mathrm{V}}_{\nu}^{j}$. The structure is identical to the usual hierarchy [Eq. (B11)] but for the last term, which connects it to the next tier elements of the hierarchy associated with the previous moment. This can be interpreted as an additional driving that each moment exerts on the next one. This naturally defines a cascade of hierarchies that can be exploited for parallel simulation of several moments with reduced overhead.

The relationship between $\tilde{\sigma}_m(t)$ and the set of $\tilde{\sigma}_{\{m\}}^{\{n\}}(t)$ follows

$$\tilde{\sigma}_m(t) = \sum_{\{m\}} a_{\{m\}}\tilde{\sigma}_{\{m\}}^{\{0\}}(t), \quad \text{(C3)}$$

where the sum is over all partitions $\{m\}$ of m (all vectors m_q with the property $\sum_{q=1}^{m} m_q q = m$) and

$$a_{\{m\}} \equiv \prod_{q=1}^{m} \prod_{j=1}^{m_q} \frac{1}{j} \binom{m - \sum_{r=q}^{m} r m_r + jq}{k}$$

is the number of permutations associated with that partition.

Finally, cumulants recursively relate to moments by means of the formula

$$\langle\langle A^n \rangle\rangle = \langle A^n \rangle - \sum_{m=1}^{n-1} \binom{n-1}{m-1} \langle\langle A^m \rangle\rangle \langle A^{n-m} \rangle. \quad \text{(C4)}$$

Furthermore factorial cumulants are obtained by

$$\langle\langle A^n \rangle\rangle_F = \langle\langle A^n \rangle\rangle - \sum_{m=1}^{n-1} \left\{ n \atop m \right\} \langle\langle A^m \rangle\rangle_F, \quad \text{(C5)}$$

where $\{ n \atop m \}$ are the Stirling numbers of the second kind.

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