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Transient fluctuation relations for time-dependent particle transport

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We consider particle transport under the influence of time-varying driving forces, where fluctuation relations connect the statistics of pairs of time reversed evolutions of physical observables. In many “mesoscopic” transport processes, the effective many-particle dynamics is dominantly classical, while the microscopic rates governing particle motion are of quantum-mechanical origin. We here employ the stochastic path integral approach as an optimal tool to probe the fluctuation statistics in such applications. Describing the classical limit of the Keldysh quantum nonequilibrium field theory, the stochastic path integral encapsulates the quantum origin of microscopic particle exchange rates. Dynamically, it is equivalent to a transport master equation which is a formalism general enough to describe many applications of practical interest. We apply the stochastic path integral to derive general functional fluctuation relations for current flow induced by time-varying forces. We show that the successive measurement processes implied by this setup do not put the derivation of *quantum* fluctuation relations in jeopardy. While in many cases the fluctuation relation for a full time-dependent current profile may contain excessive information, we formulate a number of reduced relations, and demonstrate their application to mesoscopic transport. Examples include the distribution of transmitted charge, where we show that the derivation of a fluctuation relation requires the combined monitoring of the statistics of charge *and* work.

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I. INTRODUCTION

Fluctuation relations (FRs) have recently emerged as a new powerful set of concepts in statistical physics.^{1–7} They relate the stochastic fluctuations of systems far from equilibrium to their dissipative properties, thereby generalizing the well-known fluctuation-dissipation theorem,⁸ and provide ways to quantify the degree of irreversibility of nonequilibrium processes. The possibility to formulate exact statements (sometimes referred to as “fluctuation theorems”) about generic nonequilibrium systems – both classical^{2,3,9–11} and quantum^{7,12,13} – has ignited a burgeoning research activity. On the experimental side, first tests have already appeared, e.g., for soft matter^{14–16} or mesoscopic systems.^{17–20}

The concept of FRs is frequently applied to the statistics of variables of thermodynamic significance, e.g., work, heat, or entropy. This is exemplified by the Crooks relation^{10,11}

$$\frac{P(W)}{P_b(-W)} = e^{\beta(W-\Delta F)}, \quad (1.1)$$

where $P(W)$ is the probability that an amount of work W is done on a system in a given driving protocol, i.e., when an external time-dependent force f_t is applied to the system during a time interval $t \in [-\tau, \tau]$. According to the Crooks relation, the ratio to the probability $P_b(-W)$ of negative work done during the “backward” protocol, i.e., when the time-inverted force f_{-t} acts on the system, is given by a Boltzmann-type factor, where $\beta = T^{-1}$ is the inverse temperature ($k_B = 1$ throughout) associated with the initial equilibrium state, and ΔF is

the thermodynamic free energy difference between final and initial state. Relations of this type may be applied to gain access to thermodynamic data (e.g., ΔF) from fluctuation statistics. From Eq. (1.1) one obtains the celebrated Jarzynski equality,⁹

$$\langle e^{-\beta W} \rangle = e^{-\beta \Delta F}, \quad (1.2)$$

where the average is over all process realizations under the force protocol f_t . On average $W \geq \Delta F$, which means that the “sum rule” (1.2) controls the cumulative weight of rare events. Relations of this type provide rigorous bounds on the behavior of thermodynamic observables. Fluctuations around these bounds have been analyzed recently.^{21–23}

The application of external forces in many-particle systems generally leads to transport, with current flow and re-adjustment of particle concentrations. For time-dependent driving forces, FRs for the ensuing current profiles carry particularly rich information. Here, the notion of “transport” is to be interpreted in a very general sense: it may refer to the changing number of individuals in a biological quasi-species model, to the electric current flow in a mesoscopic conductor, to the number of agents in a chemical reaction, etc. Unlike with FRs for “global” (i.e., integrated over time) variables, the time-resolved information on transient current flow is stored in a time-dependent function $I = \{I_t\}$, and one needs to study *functional* probability distributions $P[I]$, rather than functions like $P(W)$. To be specific, transport through a “system” exchanging particles with M “reservoirs” is described by currents $I_{\nu=1,\dots,M}$ flowing out of the system, and $P[I] \equiv P[I_1, \dots, I_M]$. Importantly, due to the discreteness of particle exchange with

the reservoirs, the current flow enhances the noise level of the system. Far off thermal equilibrium, this “shot noise” often becomes the dominant source of fluctuations, and the self-consistent description of the feedback cycle of currents generating noise and noise affecting the current flow becomes an important issue.

Given the present interest in time-dependent mesoscopic transport phenomena, see for instance Refs. 24–26, or in the work statistics under a quantum quench,²⁷ it is important to extend the general constraints imposed by transient FRs to the quantum setting. The above argument, however, needs to be applied with care in the quantum case. In fact, the existing literature on FRs appears to be essentially divided into a classical and a quantum part. This concerns both the systems under consideration as well as methodological aspects. Specifically, quantum theories of FRs^{7,12,13,28–30} mostly rely on quantum measurement processes performed at the beginning and at the end of the protocol. This prescription is not directly suitable to transient situations, where a continuous readout of, say, currents is required. The lack of commutativity of current operators at different times then becomes an issue, and the construction of a general quantum theory of transient FRs may seem a difficult task. Perhaps surprisingly, the derivation of a FR for *quantum* current flow goes through in unaltered form as long as the external driving forces vary on time scales corresponding to classical frequencies. We will discuss this point in some detail below (see also Ref. 31). The situation becomes particularly transparent in the many cases where the quantum system of interest actually operates close to the *semiclassical* limit: involving the dynamics of many particles, the action scales relevant to transport are usually much bigger than $\hbar$. In this case, the quantum Keldysh functional^{32–38} stays close to its classical limit, the stochastic path integral,^{39–42} and the formal lack of commutativity of current operators ceases to be an issue.

In an important early work,³ Bochkov and Kuzovlev (BK) have formulated a general classical FR for current flow. BK relied on a symmetry analysis of the Markovian operators generating the dynamics of the system (see also the discussion in Ref. 30). In contrast, our derivations below are based on a path integral representation, which permits to explore the nonequilibrium fluctuation statistics of observables beyond the rigorous bounds imposed by FRs. Also, it stands to reason that the BK functional relation is too general to be useful in applications. However, the general result can be used to obtain more manageable *derived fluctuation relations*. For example, rather than probing the full profile $I = \{I_t\}$, one may consider the total charge transmitted into the ν th reservoir, $Q_\nu \equiv \int_{-\tau}^t dt I_{\nu,t}$. This is arguably one of the most important global variables characterizing a transport process. Under *stationary* transport conditions, the forward and backward protocols coincide, $P_b(Q) = P(Q)$, and a Crooks FR for charge has been stated in the con-

text of mesoscopic transport,^{7,43}

$$\frac{P(Q)}{P(-Q)} = e^{\beta \sum_\nu f_\nu Q_\nu}. \quad (1.3)$$

This relation imposes nontrivial conditions on the generating function for the full counting statistics (FCS) of charge transport.⁴⁴ In particular, it implies that current cumulants of different order must be linked together.^{28,43,45–47} Equation (1.3) has also been probed experimentally^{19,20} in mesoscopic circuits using a quantum point contact charge detector.⁴⁸ While the case of unidirectional single-electron counting is directly accessible to experiments, recent progress has also been reported for bidirectional counting.^{20,49} All these results apply to stationary regimes. Below we will show that in the experimentally relevant case of *time dependent* forces f_t drastic things happen: the charge FR (1.3) actually breaks down, but a more general FR for the joint probability $P(Q, W)$ can still be formulated, see Eq. (5.2) below. This “generalized Crooks relation” also ensures the validity of Eq. (1.1) for the statistics of work alone. Furthermore, we will use the BK relation to derive cross-relations between nonlinear AC response coefficients, which can be put to an experimental test.

The structure of the remainder of this article is as follows. In Sec. II we discuss classical nonequilibrium transport processes in terms of master equations and the stochastic path integral. We derive the functional FR for currents under a transient driving protocol in Sec. III. The connection to the quantum theory is studied in Sec. IV. In Sec. V we discuss derived fluctuation relations and compare them to numerical simulations for a mesoscopic RC circuit. Some concluding remarks can be found in Sec. VI. Various details of our calculations have been relegated to several Appendices.

II. STOCHASTIC PATH INTEGRAL

A. Master equation

We are interested in the statistical properties of particle currents flowing through a system in contact with M reservoirs. The probability $P_t(n)$ that the system contains n particles at time t is given by the convolution

$$P_t(n) = \sum_{n-\tau} P_t(n|n-\tau) \rho(n-\tau), \quad (2.1)$$

where $P_t(n|n-\tau)$ is the conditional probability to evolve from an initial state $n-\tau$ at $-\tau$ to n at t , and the weight $\rho(n-\tau)$ describes the probability of the initial state. Without much loss of generality, we take ρ to be of Boltzmann form,

$$\rho(n) = e^{-\beta(U(n)-F)}, \quad (2.2)$$

where $U(n)$ determines the internal energy of the system and $F = -T \ln \sum_n \exp(-\beta U(n))$ is a free energy. The

Markovian time evolution of P_t is governed by a one-step master equation,⁵⁰

$$\begin{aligned}\partial_t P_t(n) &= -\hat{H}_g P_t(n), \\ \hat{H}_g(n, \hat{p}) &= \sum_{\nu=1}^M \sum_{\pm} (1 - e^{\mp \hat{p}}) g_{\nu,t}^{\pm}(n),\end{aligned}\quad (2.3)$$

where the explicitly time-dependent rates g_{ν}^{+} (g_{ν}^{-}) control the flux into (out of) the system. The operator $e^{\hat{p}}$ ($e^{-\hat{p}}$) raises (lowers) n by one unit, i.e., we have the commutator $[\hat{p}, \hat{n}] = 1$. We assume that the rates obey the detailed balance condition

$$\frac{g_{\nu,t}^{+}}{g_{\nu,t}^{-}} = e^{-\beta \kappa_{\nu,t}(n)}, \quad \kappa_{\nu,t}(n) \equiv \partial_n U(n) - f_{\nu,t}, \quad (2.4)$$

where the functions f_{ν} describe external driving forces on the system. For concreteness, we consider a cyclic protocol: starting from an equilibrium situation at time $t = -\tau$, $f_{\nu,-\tau} = 0$, the time-dependent protocol $f_{\nu,t} \neq 0$ eventually ends at $f_{\nu,\tau} = 0$. The generalization of the for-

malism below to several types of particles, or to multiple-step master equations is straightforward. However, the generalization to situations where the reservoirs are at different temperatures requires a more substantial extension of the formalism. In this case, one has to account for the energy transfer necessarily accompanying particle transfer. We will briefly comment on this point in Sec. IV C.

The condition (2.4) is less restrictive than it might seem at first sight: it states that the logarithmic ratio of rates is governed by a cost function, $T \ln(g_{\nu}^{+}/g_{\nu}^{-}) = -[E_{\nu}(n+1) - E_{\nu}(n)]$, measuring the difference in “energies” $E_{\nu}(n) = -n f_{\nu} + U(n)$ before and after a particle has entered the system through terminal ν . Note that $E_{\nu}(n)$ contains contributions linear in the driving parameters and the particle number, i.e., the driving couples to the energy balance of individual particles and not to particle interactions. With $U(n)$ introduced in Eq. (2.2) and the notation $\partial_n U(n) \equiv U(n+1) - U(n)$, we obtain Eq. (2.4). Particle interactions then correspond to nonlinearities in $U(n)$.

System	Variable n	$U(n)$	$f_{\nu,t}$
electronic circuits ³³	charge	charging energy	bias voltages
molecular motors ⁵¹	mechanochemical state of motor protein	load potential	ATP concentration
chemical reaction networks ⁵²	number of reaction partners	internal energy	chemostat concentrations
adaptive evolution ⁵³	allele frequencies	log equilibrium dist.	fitness gradients

TABLE I: Examples of application fields for the master equation.

Table I lists several application fields of present interest where the above model of discrete transport applies with little or no modification. For later reference, let us introduce one of the examples above in some more detail: consider a mesoscopic RC circuit, where the system corresponds to a central node (“quantum dot”) with n electrons held by it. The role of the reservoirs is taken by $M = 2$ voltage sources connected to the dot through resistors R_1 and R_2 . The driving of the system by a time-varying bias voltage V_t and its internal energy are given by $f_{\nu,t} = (-)^{\nu+1} e V_t / 2 - e V_{\text{eff}}$ (with $\nu = 1, 2$) and $U(n) = (n - 1/2)^2 E_c$, respectively. Here, $e V_{\text{eff}}$ is the effective chemical potential on the dot that needs to be determined self-consistently, $E_c = e^2 / (2C)$ is the capacitive charging energy of the dot, and we have taken an offset charge corresponding to Coulomb blockade peak conditions.³³ (In the rest of the paper we will set $e = 1$.) In earlier studies,^{16,54–56} circuits of this type have been discussed within the framework of Langevin equations, where the dominant source of fluctuations was thermal noise. In contrast, we wish to include the more general mechanism of noise self-generated by transport. This physics can be described by the master equation (2.3)

with sequential tunneling rates,³³

$$g_{\nu,t}^{\pm}(n) = \frac{1}{R_{\nu}} \frac{\pm \kappa_{\nu,t}(n)}{e^{\pm \beta \kappa_{\nu,t}(n)} - 1}, \quad (2.5)$$

with $\kappa_{\nu,t}$ in Eq. (2.4). The Bose-Einstein function in Eq. (2.5) indicates a degree of quantum-mechanical input, to be discussed in more detail in Sec. IV C. The rates (2.5) comply with the balance relation (2.4). In addition, it is straightforward to perform numerical simulations for the dynamics generated by the master equation, see Sec. V.

B. Path integral and current statistics

To obtain the stochastic path-integral representation^{39,40} of the above master equation, we allude to quantum-mechanical analogies and notice the similarity of Eq. (2.3) to an (imaginary-time) Schrödinger equation. The unit-normalized quantity

$$Z \equiv \sum_n P_{\tau}(n) = 1 \quad (2.6)$$

can thus be written as imaginary-time path integral,⁵⁷

$$Z = \int D(n, p) e^{\int_{-\tau}^{\tau} dt (p \partial_t n - H_g(n, p))} \rho(n_{-\tau}). \quad (2.7)$$

The integration in Eq. (2.7) is over smooth paths $(n, p) = \{(n_t, p_t)\}$, where the auxiliary “momentum” $p_t \in i\mathbb{R}$ is integrated over the imaginary axis and the information on the actual discreteness of the evolution of n_t is encoded in the “Hamiltonian” H_g .

In order to extract information on current flow, we need source fields. Specifically, time-dependent counting fields $\chi_{\nu, t}$ probing the current flow $I_{\nu, t}$ can be introduced by generalization of the Hamiltonian,⁵⁸

$$H_g(n, p) \rightarrow H_g(n, p, \chi) \equiv \sum_{\nu, \pm} \left(1 - e^{\mp(p - i\chi_{\nu})}\right) g_{\nu}^{\pm}(n). \quad (2.8)$$

Cumulants of the currents can then be obtained by functional differentiation,

$$\begin{aligned} \langle\langle I_{\nu_1, t_1} I_{\nu_2, t_2} \dots \rangle\rangle &= \frac{i\delta}{\delta\chi_{\nu_1, t_1}} \frac{i\delta}{\delta\chi_{\nu_2, t_2}} \dots \Big|_{\chi=0} \ln Z[\chi] \\ \Leftrightarrow Z[\chi] &= \left\langle e^{-i\sum_{\nu} \int_{-\tau}^{\tau} dt \chi_{\nu} I_{\nu}} \right\rangle, \end{aligned} \quad (2.9)$$

where $Z[\chi]$ is the generating functional, see Eq. (2.11) below. Heuristically, the identification $\frac{i\delta}{\delta\chi} \leftrightarrow I$ follows from the fact that χ_{ν} enters the theory precisely like a vector potential. Much like in quantum mechanics, differentiation with respect to (w.r.t.) these vector potentials generates currents. More rigorously, the connection follows from a “Ward identity” of the functional integral: temporarily considering the case of identical counting fields, $\chi_{\nu} = \chi$, the above functional derivative obtains $\frac{i\delta}{\delta\chi_t} \ln Z = \sum_{\nu} \langle I_{\nu, t} \rangle$, which we tentatively identify as the total current out of the system. On the other hand, χ may be gauged out of the Hamiltonian by a shift $p \rightarrow p + i\chi$, at the expense of an extra term $i \int dt \chi \partial_t n$ in the action. Differentiation w.r.t. χ in the shifted representation obtains $\frac{i\delta}{\delta\chi_t} \ln Z[\chi] = -\langle \partial_t n_t \rangle$. The equality of the two representations yields the continuity equation,

$$\langle \partial_t n \rangle + \sum_{\nu} \langle I_{\nu} \rangle = 0. \quad (2.10)$$

This shows that the differentiation w.r.t. χ obtains the total current, while differentiation w.r.t. the fields χ_{ν} yields currents through individual interfaces.

The functional partition sum is

$$\begin{aligned} Z[\chi] &= \int D(n, p) e^{-S_g[n, p, \chi]} \rho(n_{-\tau}), \quad (2.11) \\ S_g[n, p, \chi] &= - \int_{-\tau}^{\tau} dt (p \partial_t n - H_g(n, p, \chi)), \end{aligned}$$

with the Hamiltonian (2.8). According to standard rules of probability theory, the probability distribution of currents follows by functional integration over all χ ,

$$P[I] = \int D\chi e^{i\sum_{\nu} \int_{-\tau}^{\tau} dt \chi_{\nu} I_{\nu}} Z[\chi]. \quad (2.12)$$

The discreteness of particle transport is encoded in the exponential dependence of the Hamiltonian on the “phase space momentum” p . Keeping this information is crucial, e.g., to properly resolve the statistics of rare events. When the discreteness does not play an important role, an expansion of $e^{\pm p}$ to quadratic order in p may be justified. This reduces the stochastic path integral to the Martin-Siggia-Rose functional.^{59–61} One may then continue to either integrate over momenta, which leads to the Onsager-Machlup path integral,^{56,62} or decouple the quadratic momentum dependence by an auxiliary “noise field”, which generates an effective Langevin description.^{63,64} In the next section, we will employ the stochastic path integral (2.11) to (re)derive a number of general FRs.

III. TIME REVERSAL AND TRANSIENT FLUCTUATION RELATION FOR CURRENTS

We proceed to derive a variant of the BK fluctuation relation³ for the statistics of transient current flow. The properties of the stochastic path integral (2.11) rely on two fundamental symmetries, namely the continuity equation (2.10) and a symmetry under time reversal. The latter is crucial to all FRs. Our aim is thus to relate the functional Z , describing evolution under the influence of rates $g = \{g_{\nu, t}\}$, to the functional Z_b computed for the time-reversed rates $(\hat{T}g)_t = g_{-t}$, i.e., $Z_b = Z|_{g \rightarrow \hat{T}g}$. Here we have defined a time reversal operator $\hat{T}$ that acts on “scalar” functions $x = (n, g, f)$ as $(\hat{T}x)_t = x_{-t}$, while “vectorial” functions $v = (I, p, \chi)$ transform as $(\hat{T}v)_t = -v_{-t}$. As shown in Appendix A, the action in Eq. (2.11) satisfies the symmetry

$$\begin{aligned} S_g[n, p, \chi] &= S_{\hat{T}g}[\hat{T}n, \hat{T}(p - \beta \partial_n U), \hat{T}(\chi + i\beta f)] \\ &\quad + \beta[U(n_{\tau}) - U(n_{-\tau})]. \end{aligned} \quad (3.1)$$

Next observe that n and p in Eq. (2.11) are just functional integration variables. With the auxiliary relation for arbitrary functionals F ,

$$\begin{aligned} \int D(n, p) F[\hat{T}n, \hat{T}p] &= \int D(\hat{T}n, \hat{T}p) F[\hat{T}n, \hat{T}p] \\ &= \int D(n, p) F[n, p], \end{aligned}$$

substitution of Eq. (3.1) into (2.11) and a shift of the momentum field, $p \rightarrow p + \beta \partial_n U$, yields

$$Z[\chi] = \int D(n, p) e^{-S_{\hat{T}g}[n, p, \hat{T}(\chi + i\beta f)]} \rho(n_{-\tau}).$$

We thus obtain a prototypical FR for the generating functional,

$$Z[\chi] = Z_b[\hat{T}(\chi + i\beta f)]. \quad (3.2)$$

Inserting Eq. (3.2) into (2.12), we arrive at a variant of the Crooks relation, first formulated by BK,³

$$\frac{P[I]}{P_b[\hat{T}I]} = e^{-\beta \int_{-\tau}^{\tau} dt \sum_{\nu} f_{\nu} I_{\nu}}, \quad (3.3)$$

where P_b is the probability distribution computed for time-inverted rates $\hat{T}g$. Integrating over I and using the normalization of $P_b[I]$, we also obtain a variant of the Jarzynski equality,

$$\langle X \rangle = 1, \quad X = \exp \left(\beta \int_{-\tau}^{\tau} dt \sum_{\nu} f_{\nu} I_{\nu} \right). \quad (3.4)$$

Equation (3.3) represents the most general FR relevant to this paper. Before turning to a discussion of its applications, it is worthwhile to link our present classical formalism to the extended framework of a quantum theory of fluctuation statistics. This will be the subject of Sec. IV below. Readers primarily interested in classical transport may skip this section and directly turn to Sec. V.

IV. CONNECTION TO KELDYSH APPROACH

In this section, we show how the stochastic path integral (2.11) corresponds to the $\hbar \rightarrow 0$ limit of the Keldysh quantum nonequilibrium functional. In this way, we will see how rates encoding quantum statistics, cf. Eq. (2.5), may appear as dynamical input to an effectively classical theory of stochastic fluctuations. To be concrete, we focus on the example of the mesoscopic device introduced in section II A, but generalization to other setups is straightforward.

A. Model

We study a quantum dot connected to $M = 2$ Fermi liquid leads ($\nu = 1, 2$) at temperatures well above the dot's mean level spacing and in the “open” limit, $\bar{g}_{\nu} > 1$. The dimensionless conductances $\bar{g}_{\nu} \equiv 2\pi\hbar G_{\nu}$, with $G_{\nu} = R_{\nu}^{-1}$, describe the transparency of the contacts to the electrodes.^{33,36} We assume that the external voltage V_t varies on classical time scales (such as the RC time of the circuit), which are large compared to quantum time scales of the problem. Technically, this means that terms like $\hbar\partial_t V_t$ can be neglected. The quantum nonequilibrium theory corresponds to a Keldysh functional integral,^{32–35}

$$Z = \int D(\phi_c, \phi_q) \exp(-S[\phi_c, \phi_q]/\hbar), \quad (4.1)$$

with time-dependent “classical”, $\phi_{c,t}$, and “quantum”, $\phi_{q,t}$, phase fields. These real-valued fields originate from a Hubbard-Stratonovich decoupling of the Coulomb interaction.³² Note that in Eq. (4.1), the action $S =$

$S_c + S_{\text{tun}}$ is dimensionful, in contrast to the dimensionless action S_g in the classical Eq. (2.11). S_c describes electron-electron interactions (Coulomb blockade) due to the charging energy E_c ,

$$S_c[\phi_c, \phi_q] = i \frac{\hbar^2}{E_c} \int dt \phi_q \partial_t^2 \phi_c, \quad (4.2)$$

while the tunnel action contains the influence of the attached electrodes and is of Ambegaokar-Eckern-Schön form,^{32,37,38}

$$S_{\text{tun}}[\phi_c, \phi_q] = -\hbar \sum_{\nu=1,2} \frac{\bar{g}_{\nu}}{4} \text{Tr} \left(\hat{\Lambda}_{\nu} e^{-i\hat{\phi}} \hat{\Lambda}_{\text{d}} e^{i\hat{\phi}} \right), \quad (4.3)$$

where $\hat{\Lambda}_X$ (with $X = \nu, \text{d}$) and $\hat{\phi}$ are operators in both Keldysh and time space. We have $\hat{\phi} = \{\phi_t\}$ with

$$\phi_t = \text{diag}(\phi_t^+, \phi_t^-), \quad \phi^{\pm} = \phi_c \pm \phi_q, \quad (4.4)$$

where we employ the “contour representation” of Keldysh theory³⁵ throughout. Furthermore, $\hat{\Lambda}_X = \{\Lambda_{X,t,t'}\}$ with a 2×2 matrix $\Lambda_{X,t,t'}$. For classically varying V_t , it is convenient to pass to a Wigner representation,³² whereupon matrices become functions of energy and time, $\{\Lambda_{X,t,t'}\} \rightarrow \{\Lambda_X(t, \epsilon)\}$, and $\text{Tr} \rightarrow (2\pi\hbar)^{-1} \int d\epsilon \int_{-\tau}^{\tau} dt \text{tr}$, with “tr” denoting the trace in Keldysh space. We then have the Keldysh matrix^{32,33,38}

$$\Lambda_X(t, \epsilon) = \begin{pmatrix} 1 - 2n_X & 2n_X \\ 2(1 - n_X) & -(1 - 2n_X) \end{pmatrix}, \quad (4.5)$$

where $n_X = n_X(t, \epsilon)$ is the electron distribution function in the leads ($X = \nu$) and in the dot ($X = \text{d}$). The Fermi-Dirac distribution function of the ν th reservoir is

$$n_{\nu}(t, \epsilon) = n_{\text{F}}(\epsilon; V_{\nu,t}, T) = \frac{1}{e^{(\epsilon - V_{\nu})/T} + 1}, \quad (4.6)$$

where the driving voltages are

$$V_{\nu,t} \equiv (-)^{\nu+1} V_t/2. \quad (4.7)$$

The dot distribution function $n_{\text{d}}(t, \epsilon)$, however, has to be determined self-consistently.

Noting that the product between Wigner “functions” under the trace in Eq. (4.3) has to be understood as the Moyal product³²

$$A * B = AB + \frac{i\hbar}{2} (\partial_{\epsilon} A \partial_t B - \partial_t A \partial_{\epsilon} B) + \mathcal{O}(\hbar^2),$$

we obtain

$$e^{-i\hat{\phi}} \hat{\Lambda}_{\text{d}} e^{i\hat{\phi}} = \begin{pmatrix} 1 - 2n_{\text{d},V_c} & 2e^{-2i\phi_q} n_{\text{d},V_c} \\ 2e^{2i\phi_q} (1 - n_{\text{d},V_c}) & -(1 - 2n_{\text{d},V_c}) \end{pmatrix}, \quad (4.8)$$

where higher-order corrections in $\hbar$ are neglected, $n_{\text{d},V_c}(\epsilon) \equiv n_{\text{d}}(\epsilon - V_c)$, and the dynamically fluctuating voltage on the dot is^{37,38}

$$V_{c,t} \equiv \hbar \partial_t \phi_{c,t}. \quad (4.9)$$

Substituting Eq. (4.8) into Eq. (4.3), we obtain

$$S_{\text{tun}}[V_c, p] = \hbar \sum_{\nu, \pm} \int dt (1 - e^{\mp p}) g_{\nu}^{\pm}(V_c) \quad (4.10)$$

with $p = -2i\phi_q$ and the rates

$$g_{\nu}^{+}(V_c) = G_{\nu} \int d\epsilon n_{\nu}(\epsilon) [1 - n_{d, V_c}(\epsilon)],$$

$$g_{\nu}^{-}(V_c) = G_{\nu} \int d\epsilon n_{d, V_c}(\epsilon) [1 - n_{\nu}(\epsilon)].$$

Finally, introducing a charge-like variable through $n = V_c/2E_c$, adding the (n, p) representation of the charging term, $S_c[n, p] = -\hbar \int dt p \partial_t n$, and defining $S_g[n, p] = S[n, p]/\hbar$, we arrive at an action as in Eq. (2.11), where the Hamiltonian H_g is governed by the rates

$$g_{\nu}^{+}(n) = G_{\nu} \int d\epsilon n_{\nu}(\epsilon + \partial_n U) [1 - n_d(\epsilon)], \quad (4.11)$$

$$g_{\nu}^{-}(n) = G_{\nu} \int d\epsilon [1 - n_{\nu}(\epsilon + \partial_n U)] n_d(\epsilon),$$

with $\partial_n U = 2E_c n$.

B. Model distributions

Importantly, the rates (4.11) do not determine the action of the dot unless we specify the dot distribution function n_d . We here consider three different cases, all of which are physically relevant and conceptually interesting in their own way. Which distribution is ultimately realized depends on the ratio of two time scales, the energy relaxation time due to electron-electron interactions^{65–67} on the dot, $\tau_{ee}(\epsilon) = \hbar E_{\text{Th}}^2 / \epsilon^2 \Delta$, and the time for escape into the leads, $\tau_d = \hbar / \Delta (\bar{g}_1 + \bar{g}_2)$. Here Δ is the dot's single-particle level spacing, ϵ is the characteristic excitation energy of particles in the system, and $E_{\text{Th}} = \hbar / t_{\text{Th}}$ is the Thouless energy, where t_{Th} is the classical time scale before the single-particle dynamics in the dot becomes ergodic, e.g., the diffusion time.

The cases considered here are as follows. (i) In the classical limit, $\hbar \rightarrow 0$, we observe that $\tau_{ee}/\tau_d \rightarrow 0$. This implies that strong relaxation mechanisms on the dot enforce an effective equilibrium Fermi distribution,

$$n_d(t, \epsilon) = n_F(\epsilon; V_{\text{eff}, t}, T_{\text{eff}, t}), \quad (4.12)$$

where the effective chemical potential V_{eff} and the effective temperature T_{eff} are determined by requiring particle current and energy current conservation in the dot-leads composite system,^{33,42,68}

$$V_{\text{eff}, t} = \frac{\bar{g}_1 - \bar{g}_2}{\bar{g}_1 + \bar{g}_2} \frac{V_t}{2},$$

$$T_{\text{eff}, t} = \sqrt{T^2 + \frac{3\bar{g}_1\bar{g}_2}{\pi^2(\bar{g}_1 + \bar{g}_2)^2} V_t^2}. \quad (4.13)$$

This so-called “hot electron distribution” captures the heating of the system under the application of a voltage bias. We stress that Eq. (4.12) holds only when τ_{ee} is short compared to the time scale for variation of V_t . (ii) Alternatively, one may consider a situation where the system is externally cooled to the ambient temperature T . In this case, we have

$$n_d(t, \epsilon) = n_F(\epsilon; V_{\text{eff}, t}, T). \quad (4.14)$$

(iii) In cases where the dwell time τ_d is smaller than the relaxation time τ_{ee} , the effective distribution on the dot is determined by the coupling to the leads rather than by internal relaxation. In the absence of counting fields, $\chi = 0$, this leads to the “double-step distribution” obtained by the weighted superposition of the two lead distributions,

$$n_d(t, \epsilon) = \sum_{\nu} \frac{\bar{g}_{\nu}}{\bar{g}_1 + \bar{g}_2} n_F(\epsilon; V_{\nu, t}, T). \quad (4.15)$$

However, for finite χ , the situation gets more complicated in that the effective dot distribution becomes χ -dependent. The ensuing structures are discussed in Appendix B, where we also show that the FR (3.2), derived within a classical formalism in Sec. III, stays valid in such a quantum-mechanical setting.

C. Fluctuation relations

In the remainder of this section, we address the scenarios (i) and (ii) in some more detail.

First, if the dot is cooled down to ambient temperatures, the dot distribution function is given by the Fermi distribution (4.14). Substituting this function into Eq. (4.11) and doing the energy integrals, we obtain the rates (2.5). These rates obey the detailed balance relation (2.4), which means that the externally cooled setup [case (ii) above] seamlessly fits into the general framework of Secs. II and III. Specifically, the FR (3.3) and the derived relations in Sec. V below hold in full generality.

However, for a dot kept in isolation [case (i)], the situation is different. Here, the temperature realized in the dot may differ strongly from that of the leads, which means that there is no uniquely specified reference temperature to relate to. Temperature mismatch of this type will effectively be realized in many different circumstances: out of equilibrium, transport through a dissipative system generally leads to energy relaxation and, hence, to heating. It stands to reason that the effective temperature will typically scale with the external driving parameters, $T_{\text{eff}} = \mathcal{O}(V_{\nu})$. The resulting effective transport rates then no longer obey a detailed balance relation containing the ambient temperature as a reference scale, which in turn implies that the FR in Eq. (3.3) no longer holds.

Albeit the FR (3.3) is violated, it is still possible to formulate modified FRs that contain crucial information

about the fluctuation statistics of the system. Let us briefly sketch two different approaches to handling this situation. First, one may require that FRs categorically have to relate to ambient temperature, or, more generally, to the temperatures T_ν of the external leads. (In this part, we allow for unequal temperatures in the reservoirs.) While our discussion above shows that the FR (3.3) for particle currents is violated, it is possible to derive joint FRs for particle and energy currents that do apply to the heated dot yet contain the reservoir temperatures T_ν only. These FRs rely on the results for the statistics of charge and energy transfer obtained in Refs. 42,68–70. For the convenience of the reader, we briefly summarize the main conclusions of these references in the language of our paper. The main idea is to generalize the state space of the theory from n to (n, ϵ) , where the continuous variable ϵ represents the energy of the dot. It is then straightforward to derive a generalized master equation for the joint probability $P(n, \epsilon)$, where the rates $g_\nu^\pm(n, \epsilon)$ are determined by the energy integrands of Eq. (4.11), i.e., $g_\nu^\pm(n) = \int d\epsilon g_\nu^\pm(n, \epsilon)$. The corresponding stochastic path integral is given by^{42,69,70}

$$\begin{aligned} Z &= \int D(n, p, \epsilon, \xi) e^{-S_g[n, p, \epsilon, \xi]}, \\ S_g &= - \int_{-\tau}^{\tau} dt [p \partial_t n + \xi \partial_t \epsilon - H_g(n, p, \epsilon, \xi)], \\ H_g &= \sum_{\nu, \pm} \int d\epsilon' \left(1 - e^{\mp(p + \epsilon' \xi)} \right) g_\nu^\pm(n, \epsilon'), \end{aligned} \quad (4.16)$$

where the time-like variable ξ is conjugate to ϵ . In order to probe the joint statistics of particle and energy currents,^{69,70} we couple both p and ξ to counting fields in the respective ν -dependent part of H_g ,

$$p_t \rightarrow p_t - i\chi_{\nu, t}, \quad \xi_t \rightarrow \xi_t - i\lambda_{\nu, t}, \quad (4.17)$$

see also Eq. (2.8). One can then verify that the invariance of the action under time reversal can be effected by a simultaneous transformation of both counting fields,

$$\chi_\nu \rightarrow \hat{T}(\chi_\nu + i\beta_\nu V_\nu), \quad \lambda_\nu \rightarrow \hat{T}(\lambda_\nu - i\beta_\nu),$$

where $\beta_\nu \equiv T_\nu^{-1}$. In this way, the symmetry relation in Eq. (3.1) gets generalized and we obtain the *extended FR*

$$Z[\chi_\nu, \lambda_\nu] = Z_b[\hat{T}(\chi_\nu + i\beta_\nu V_\nu), \hat{T}(\lambda_\nu - i\beta_\nu)]. \quad (4.18)$$

Note that this FR involves the ambient (reservoir) temperatures T_ν only. The price to be paid is that both the particle (χ_ν) and the energy (λ_ν) current into the ν th reservoir have to be monitored. A quantum mechanical stationary version of Eq. (4.18) has recently been discussed in Ref. 28.

A second, and at this stage more heuristic, approach is to *define* time-dependent effective temperatures T_ν^* characterizing the particle exchange with the ν th reservoir through the logarithmic ratio of rates,

$$T_\nu^* \equiv \frac{V_\nu - V_{\text{eff}} - \partial_n U}{\ln(g_\nu^+/g_\nu^-)}. \quad (4.19)$$

We now observe that the FR in Eq. (3.2) holds provided one replaces the global temperature $T = \beta^{-1}$ by the time- and ν -dependent temperatures T_ν^* in Eq. (4.19). This FR contains effective (and potentially unknown) temperatures different from the ambient temperature. Focusing on particle transport alone, it provides a reduced description of the nonequilibrium process. Referring to the applications discussed in Sec. V, one may employ the statistical information encoded in such FRs to *determine* these temperatures. This is of interest for systems where external biasing is expected to generate heating through mechanisms that are not completely understood a priori. A precise formulation, however, is beyond the scope of this article and requires to carefully address several subtleties.⁷¹

In the next section, we will turn back to the general FR (3.3) and discuss its applied consequences in the description of the fluctuation statistics of nonequilibrium transport.

V. APPLICATIONS

In the present approach, the general FR (3.3) and its spin-offs are embedded into the formalism of the stochastic path integral. This implies a lot of freedom in exploring the role of fluctuations beyond the rigorous bounds implied by FRs. For example, the general Jarzynski relation (3.4) states that, for a cyclic protocol, the random variable X averages to unity, $\langle X \rangle = 1$. This identity holds under very general circumstances, and it is in this sense that the *fluctuations* of X contain more telling information. This point has been explored in Ref. 23, where we showed how the statistics of X signifies the crossover from near into far equilibrium situations.

In this paper, we concentrate on the statistical information encoded in the FRs as such. A first aspect to notice is that the *functional* FR (3.3), which is a relation for the “infinitely many” variables $I = \{I_t\}$, contains information that in most applications will be excessive. The applied value of the identity rather lies in its potential as a starting point for the derivation of a wealth of *derived relations*. Technically speaking, one may pass to these reduced identities by taking marginals in the sense of probability theory. Below, we discuss such reduction schemes on a number of examples. To keep the discussion concrete, we will stay with our prototypical mesoscopic circuit as a reference system. Generalization to other systems should be straightforward. We stress that the FR (3.3) holds also in a quantum-mechanical setting, see our discussion above and Ref. 31.

A. Stationary case

Let us begin by discussing what Eqs. (3.2) and (3.3) predict for the specific case of a stationary bias, $V_t = V$. More precisely, we assume a symmetric protocol $V_t = V_{-t}$

which is switched on (off) within a time τ_s that is very short compared to the counting time, $\tau_s \ll \tau$. Then, V_t assumes a constant value, V , during the long time interval $|t| < \tau - \tau_s$. For this voltage bias protocol, we have $Z_b = Z$. Moreover, it makes sense to consider the time averaged current, $I \equiv Q/2\tau$, where $Q = \int dt I_t$ is the charge transmitted during the counting interval 2τ . Taking the limit $\tau_s/\tau \rightarrow 0$, a stationary bias can then be described using the above formalism.

Assuming a mesoscopic two-terminal ($M = 2$) setup for concreteness, let us choose constant counting fields, $\chi_1 = -\chi_2 = \chi/2$, whereupon $Z[\chi]$ reduces to a function $Z(\chi)$, and differentiation w.r.t. χ probes the statistics of I . The reduced form of Eq. (3.2) then recovers the known FR^{7,28,43}

$$Z(\chi) = Z(-\chi + i\beta V), \quad (5.1)$$

which is equivalent to Eq. (1.3) for the probability distribution function $P(Q)$.

It is worthwhile to discuss an important consequence that Eq. (5.1) entails for taking the classical limit of nonequilibrium quantum theories. Within the Keldysh approach to nonequilibrium quantum dynamics it is customary to associate the classical limit with a quadratic expansion in the quantum field, cf. Ref. 34 for a discussion of this point. However, the FR (5.1) implies that this expansion cannot be valid, unless one operates in a near equilibrium setting. To see this, notice that the counting field χ couples additively to the quantum field ϕ_q . Thus, a quadratic action in ϕ_q implies a quadratic $S[\chi]$. Now, consider the most general quadratic *Ansatz*, $\ln Z(\chi) = 2\tau[-i\langle I \rangle \chi + C_2 \chi^2]$, where we noted that differentiation w.r.t. χ at $\chi = 0$ yields the average current, $\langle I \rangle$. Consistency with Eq. (5.1) requires $C_2 = \langle I \rangle / (\beta V)$. For $\langle I \rangle \sim V$, this states that the fluctuations of I (determined by the second order of the expansion in χ) are thermal, $\text{var}(I) \sim T$. Thus, a quadratic expansion in ϕ_q is not capable of describing nonequilibrium noise and cannot be valid in general.

More generally, Eq. (5.1) implies constraints for the nonlinear coefficients describing the response of the k th current cumulant $\langle\langle I^k \rangle\rangle$ to the voltage at order V^l . Since derivatives w.r.t. V can be traded for derivatives w.r.t. χ in Eq. (5.1), different coefficients with the same order $k+l$ are connected.^{7,46,47} The fluctuation-dissipation theorem linking thermal Johnson-Nyquist noise to the linear conductance follows from the lowest-order equation in this hierarchy, $l+k=2$. Higher-order relations with $l+k>2$ represent generalizations to the nonequilibrium. We discuss the extension of these relations to the time-dependent situation in Sec. VC.

B. Generalized Crooks relation

We return now to a general setting with M reservoirs and time-dependent forces. In applications, one is often

interested in the FCS of charge, $Q[I] = \int_{-\tau}^{\tau} dt I_t$, transmitted at one of the terminals. (We drop the reservoir index ν for notational simplicity.) For stationary bias, the probability distribution function $P(Q)$ obeys a FR, see Eq. (1.3), and one may ask whether this relation extends to time-varying driving. To answer this question, we study the statistics of both the transmitted charge $Q[I]$ and the work done on the system, $W_g[I] = \int_{-\tau}^{\tau} dt I_t f_t$ in terms of their joint probability density

$$\begin{aligned} P(Q, W) &\equiv \langle \delta(Q - Q[I]) \delta(W - W_g[I]) \rangle \\ &= \int \frac{d\chi_q d\chi_w}{(2\pi)^2} e^{i(\chi_q Q + \chi_w W)} Z[\chi_q + \chi_w f]. \end{aligned}$$

The second equality follows from the integral representation of the δ -functions, where, by virtue of Eq. (2.9), the generating function Z has to be taken for the particular time-dependent counting field $\chi_t = \chi_q + \chi_w f_t$. We now use the FR (3.2) and integrate over the parameters $\chi_{q,w}$,

$$\begin{aligned} P(Q, W) &= \\ &= \int \frac{d\chi_q d\chi_w}{(2\pi)^2} e^{i(\chi_q Q + \chi_w W)} Z_b[-\chi_q - (\chi_w - i\beta)\hat{T}f] \\ &= \int \frac{d\chi_q d\chi_w}{(2\pi)^2} e^{-i[\chi_q Q + (\chi_w + i\beta)W]} Z_b[\chi_q + \chi_w \hat{T}f] \\ &= e^{\beta W} \left\langle \delta(Q + Q[I]) \delta(W + W_{\hat{T}g}[I]) \right\rangle_b. \end{aligned}$$

Note that the transmitted charge and the work in the backward average are defined with the time-inverted force. In effect, we obtain a generalized Crooks relation coupling the transmitted charge and the work done on the system,

$$\frac{P(Q, W)}{P_b(-Q, -W)} = e^{\beta W}, \quad (5.2)$$

which applies for time-dependent driving forces. The derived transient FR (5.2) connects the charge to the work fluctuation statistics. Note that in a stationary situation, $W \propto Q$, and $P(Q, W)$ reduces to $P(Q)$. In that case, Eq. (5.2) implies the FR (1.3) which now has the same physical content as Eq. (1.1). Turning to the generic time-dependent case, integrating Eq. (5.2) over Q recovers the standard Crooks relation (1.1) for the work distribution function $P(W)$. However, there is no FR for the reduced probability $P(Q) = \int dW P(Q, W)$ anymore, unless the driving force is time-independent.

Figure 1 shows a test of the generalized Crooks relation (5.2) in a numerical simulation of the master equation (2.3). Here, we have considered the asymmetric pulse protocol

$$V_t = V_0 \frac{\gamma t}{t^2 + \gamma^2}, \quad (5.3)$$

where V_0 is the pulse strength and γ the pulse width. A relevant time scale is the RC time, $\Omega^{-1} = RC/2$, where

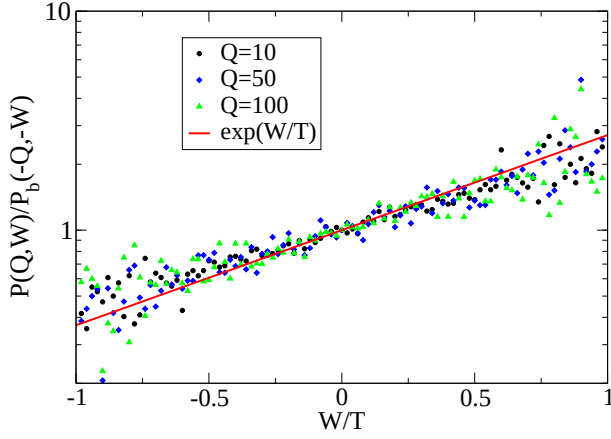


FIG. 1: Numerical results for $P(Q, W)/P_b(-Q, -W)$ vs W/T for several values of the total transferred charge, taken within the respective window $[Q - 1, Q + 1]$. The straight red line is the result predicted by Eq. (5.2). Parameters: $\gamma = 0.01\tau$, $\Omega\tau = 2 \times 10^3$, $V_0\tau = 10$, and $V_0/T = 0.04$.

we choose symmetric contacts, $R_1 = R_2 = R$. We mention in passing that $P_b(-Q, -W) = P(Q, -W)$ for an antisymmetric protocol like Eq. (5.3). Within statistical errors,⁷² the fluctuation statistics of W under a fixed total transferred charge Q , obtained numerically from 4×10^6 independent runs, agrees well with the prediction (5.2).

C. Nonlinear time-dependent transport coefficients

Another way to reduce the information contained in the transient FR for currents, Eq. (3.2), is to employ a series expansion of the k th current cumulant in terms of the driving forces $f_{\nu, t}$,

$$\langle\langle I_{\nu_1, t_1} I_{\nu_2, t_2} \dots \rangle\rangle = \sum_{l=0}^{\infty} \frac{1}{l!} \sum_{\nu'_1, \dots, \nu'_l} \int_{-\tau}^{\tau} dt'_1 \dots dt'_l L_{\nu_1, \dots, \nu_k; \nu'_1, \dots, \nu'_l}^{(k, l)}(t_1, \dots, t_k; t'_1, \dots, t'_l) f_{\nu'_1, t'_1} \dots f_{\nu'_l, t'_l}. \quad (5.4)$$

Together with Eq. (2.9), this defines the time-dependent nonlinear Onsager response coefficients,

$$L_{\nu_1, \dots, \nu_k; \nu'_1, \dots, \nu'_l}^{(k, l)}(t_1, \dots, t_k; t'_1, \dots, t'_l) = \frac{i^k \delta^{(k+l)}}{\delta f_{\nu'_1, t'_1} \dots \delta f_{\nu'_l, t'_l} \delta \chi_{\nu_1, t_1} \dots \delta \chi_{\nu_k, t_k}} \ln Z[\chi] \Big|_{\chi=f=0}. \quad (5.5)$$

They are symmetric under arbitrary separate permutations of the indices (ν_i, t_i) and (ν'_i, t'_i) . Due to the normalization condition $Z[0] = 1$, all coefficients with $k = 0$ identically vanish. Moreover, for $l = 0$, they represent equilibrium correlations. Causality provides another constraint: the response of the system to the driving force $f_{\nu'_i, t'_i}$ is restricted to times $t \geq t'_i$. This implies that Eq. (5.5) must vanish whenever there is at least one $t'_j > \max(t_1, \dots, t_k)$.

Using the FR (3.2), we next show how for a given order $k + l$, different coefficients in Eq. (5.5) are linked together.² The general relation for these time-dependent quantities is rather lengthy and given in App. C, see Eq. (C1). All relations resulting for the four lowest orders ($k + l \leq 4$) are also specified explicitly in App. C. In experimental applications, it is often more useful to probe Fourier coefficients, and we provide here the spectral decomposition of the relations in App. C. We use the Fourier convention $L(t) = \frac{1}{2\tau} \sum_{n \in \mathbb{Z}} e^{-i\pi n t / \tau} L(n)$ (for all time arguments), where the series terminates at the desired accuracy level. Since the coefficients (5.5) are real functions, we have $L(-n) = L^*(n)$. Rather than stating the general formula, we now quote the result for the few lowest orders, $k + l = 2, 3$, and 4.

From Eq. (C2), the *second-order relations* are

$$\begin{aligned} \text{Im } L_{\nu_1, \nu_2}^{(2, 0)}(n_1, n_2) &= 0, \\ \text{Re } L_{\nu_1; \nu'_1}^{(1, 1)}(n_1; n'_1) &= -\frac{\beta}{2} L_{\nu_1, \nu'_1}^{(2, 0)}(n_1, n'_1). \end{aligned} \quad (5.6)$$

Noting that $L^{(1, 1)}$ describes the linear dissipative response of currents to driving, while $L^{(2, 0)}$ is a measure of equilibrium fluctuations, we recognize the second relation in Eq. (5.6) as a spectral representation of the fluctuation-dissipation theorem. Moreover, the symmetry of Eq. (5.5) under permutations of indices implies the Onsager-Casimir reciprocity relation

$$\text{Re } L_{\nu_1; \nu'_1}^{(1, 1)}(n_1; n'_1) = \text{Re } L_{\nu'_1; \nu_1}^{(1, 1)}(n'_1; n_1). \quad (5.7)$$

There is no condition on $\text{Im } L^{(1, 1)}$. Next, from Eq. (C3) we find the *third-order relations*

$$\begin{aligned} \text{Re } L_{\nu_1, \nu_2, \nu_3}^{(3, 0)} &= 0, \\ \text{Im } L_{\nu_1, \nu_2; \nu'_1}^{(2, 1)} &= \frac{\beta}{2} \text{Im } L_{\nu_1, \nu_2, \nu'_1}^{(3, 0)}, \\ \text{Re } L_{\nu_1; \nu'_1, \nu'_2}^{(1, 2)} &= -\frac{\beta}{2} \text{Re} \left(L_{\nu_1, \nu'_1; \nu'_2}^{(2, 1)} + L_{\nu_1, \nu'_2; \nu'_1}^{(2, 1)} \right). \end{aligned} \quad (5.8)$$

For simplicity, we here suppress the Fourier indices (n_i, n'_j) , which can be easily restored from the respective reservoir indices (ν_i, ν'_j) . The last relation in Eq. (5.8)

states how the leading nonlinear response of the current connects to the linear order of the current noise. Finally, in *fourth order*, we get

$$\begin{aligned} \text{Im } L_{\nu_1, \nu_2, \nu_3, \nu_4}^{(4,0)} &= 0, & \text{Re } L_{\nu_1, \nu_2, \nu_3, \nu'_1}^{(3,1)} &= -\frac{\beta}{2} L_{\nu_1, \nu_2, \nu_3, \nu'_1}^{(4,0)}, \\ \text{Im } L_{\nu_1, \nu_2, \nu'_1, \nu'_2}^{(2,2)} &= -\frac{\beta}{2} \text{Im} \left(L_{\nu_1, \nu_2, \nu'_1, \nu'_2}^{(3,1)} + L_{\nu_1, \nu_2, \nu'_2, \nu'_1}^{(3,1)} \right), \\ \text{Re } L_{\nu_1, \nu'_1, \nu'_2, \nu'_3}^{(1,3)} &= -\frac{\beta^3}{2} L_{\nu_1, \nu'_1, \nu'_2, \nu'_3}^{(4,0)} - \frac{\beta^2}{2} \text{Re} \left(L_{\nu_1, \nu'_2, \nu'_3, \nu'_1}^{(3,1)} + L_{\nu_1, \nu'_1, \nu'_3, \nu'_2}^{(3,1)} + L_{\nu_1, \nu'_1, \nu'_2, \nu'_3}^{(3,1)} \right) \\ &\quad - \frac{\beta}{2} \text{Re} \left(L_{\nu_1, \nu'_1, \nu'_2, \nu'_3}^{(2,2)} + L_{\nu_1, \nu'_2, \nu'_1, \nu'_3}^{(2,2)} + L_{\nu_1, \nu'_3, \nu'_1, \nu'_2}^{(2,2)} \right). \end{aligned} \quad (5.9)$$

The utility of such relations in the characterization of stationary transport has been emphasized before.^{7,28,46,47} Here we have applied the concept of cross-relations to time-dependent coefficients. Such relations provide a hierarchy of benchmark criteria for time-varying transport measurements or numerical simulations.

VI. CONCLUSIONS

In recent years, fluctuation relations have been recognized as a potent tool in the characterization of the fluctuation statistics of nonequilibrium systems. Here, we have focused on the adaption of this concept to the fluctuations of transient currents, the motivation being that transport in response to time-varying forces represents the perhaps most direct way of probing the nonequilibrium physics of complex systems. We advocated the stochastic path integral as an optimal tool to describe the emerging feedback mechanism of currents inducing noise which in turn affects the statistics of currents. In its most general form, the degree of irreversibility by which a transport process differs from the time-reversed process is characterized by the functional Crooks relation (3.3). This relation has first been stated in the seminal BK paper.³ The value of the present derivation primarily lies in the linkage of the exact “sum rule” (3.3) to the highly flexible formalism of the stochastic path integral. Indeed, we argued that the fluctuation relation is overly general to be of much use in concrete applications. It is more informative to explore fluctuations around the rigorous bounds imposed by these relations, or to consider derived relations for the statistics of time integrated variables, or Fourier coefficients.

The stochastic path integral also affords an interpretation as the classical limit of the quantum theory of nonequilibrium fluctuations. This connection is useful

both from a conceptual and an applied point of view. Conceptually, it provides a connection between the quantum and the classical theory of fluctuation relations. The former is usually described by means of quantum projector techniques, a theoretical language that is not straightforwardly linked to classical limits. From an applied point of view, the quantum-classical correspondence is of value in that it allows for an integrated description of processes whose stochastic rates follow from a microscopic quantum dynamics. Moreover, we have demonstrated that the FR stays valid in a quantum-mechanical regime.

In this paper, we illustrated many of the concepts above on the prototypical example of a mesoscopic RC circuit. However, the underlying theoretical framework is much more general in nature, and we believe that it can be straightforwardly adjusted to many other different settings.

VII. ACKNOWLEDGEMENTS

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Appendix A: Symmetry of the action

Here we provide the derivation of the symmetry relation (3.1) for the action (2.11) of the stochastic path integral. Using the definition of the time-reversal operator $\hat{T}$ in Sec. III and recalling the definition of $\kappa_{\nu,t}(n)$ in Eq. (2.4), we have

$$S_{\hat{T}g}[\hat{T}n, \hat{T}(p - \beta \partial_n U), \hat{T}(\chi + i\beta f)] = - \int_{-\tau}^{\tau} dt \left[p_{-t} \partial_{-t} n_{-t} + \beta (\partial_n U)_{-t} \partial_{-t} n_{-t} \right. \\ \left. - \sum_{\nu, \pm} \left(1 - e^{\mp[-p_{-t} + i\chi_{\nu, -t} - \beta \kappa_{\nu, -t}(n_{-t})]} \right) g_{\nu, -t}^{\pm} \right].$$

Next we change the time integration variable, $t \rightarrow -t$, and the summation variable $\pm \rightarrow \mp$. Noting that $\int_{-\tau}^{\tau} dt (\partial_n U) \partial_t n = U(n_{\tau}) - U(n_{-\tau})$, we obtain for the above expression

$$S_{\hat{T}g} = -\beta[U(n_{\tau}) - U(n_{-\tau})] - \int_{-\tau}^{\tau} dt \left[p_t \partial_t n_t - \sum_{\nu, \pm} \left(1 - e^{\mp(p_t - i\chi_{\nu, t} + \beta \kappa_{\nu, t}(n_t))} \right) g_{\nu, t}^{\mp} \right].$$

Finally, we employ the detailed balance condition (2.4) and arrive at Eq. (3.1).

Appendix B: Double-step distribution

In this appendix,⁵⁷ we discuss the structure of the tunnel action (4.3) when the effective dot distribution has the double-step form in Eq. (4.15). We assume that the system is driven and monitored on classical time scales such that we can neglect terms like $\hbar \partial_t V_{\nu}$ and $\hbar \partial_t \chi_{\nu}$. The driving voltages V_{ν} were defined in Eq. (4.7). In the present quantum-mechanical context, it is crucial to include the counting fields χ_{ν} from the outset. The Wigner representation of the Λ_X matrices generalizing Eq. (4.5) is given by⁵⁸

$$\tilde{\Lambda}_{\nu}(t, \epsilon) = \begin{pmatrix} 1 - 2n_{\nu} & 2e^{i\chi_{\nu, t} n_{\nu}} \\ 2e^{-i\chi_{\nu, t}(1 - n_{\nu})} & -(1 - 2n_{\nu}) \end{pmatrix}, \\ \hat{\Lambda}_d = \mathcal{N} \sum_{\nu} \frac{\bar{g}_{\nu}}{\bar{g}_1 + \bar{g}_2} \tilde{\Lambda}_{\nu}, \quad (\text{B1})$$

where $n_{\nu}(t, \epsilon)$ is defined in Eq. (4.6). The factor $\mathcal{N}(t, \epsilon)$ ensures the normalization condition,³² $\hat{\Lambda}_d^2 = 1$, and is given by

$$\mathcal{N} = \frac{\bar{g}_1 + \bar{g}_2}{\sqrt{(\bar{g}_1 + \bar{g}_2)^2 + 4\bar{g}_1\bar{g}_2 B_{\chi}}}, \\ B_{\chi} = (e^{i\chi} - 1)n_1(1 - n_2) + (e^{-i\chi} - 1)n_2(1 - n_1),$$

where $\chi \equiv \chi_1 - \chi_2$. Note the symmetry property $B_{-\chi + i\beta V} = B_{\chi}$, which is instrumental to prove FRs. Keeping terms up to order $\hbar \partial_t \phi_q$, instead of Eq. (4.8), the Moyal product expansion yields

$$e^{i\phi} \tilde{\Lambda}_{\nu} e^{-i\phi} = \begin{pmatrix} 1 - 2n_{\nu, V_c + \hbar \partial_t \phi_q} & 2e^{i(\chi_{\nu} + 2\phi_q)n_{\nu, V_c}} \\ 2e^{-i(\chi_{\nu} + 2\phi_q)(1 - n_{\nu, V_c})} & -(1 - 2n_{\nu, V_c - \hbar \partial_t \phi_q}) \end{pmatrix}, \quad (\text{B2})$$

where $n_{\nu, V_c}(\epsilon) \equiv n_{\nu}(\epsilon + V_c)$. Notice that the energy arguments in the diagonal elements are shifted by $\pm \hbar \partial_t \phi_q$.

We now insert Eqs. (B1) and (B2) into Eq. (4.10), and write $S_{\text{tun}} = S_{\text{tun}}^{(1)} + S_{\text{tun}}^{(2)} + S_{\text{tun}}^{(3)}$. The first term reads

$$S_{\text{tun}}^{(1)} = \frac{\hbar}{2} \sum_{\nu\nu', \pm} \int dt \left[-2e^{\pm i(2\phi_q + \chi_{\nu} - \chi_{\nu'})} g_{\nu\nu'}^{\pm}(V_c) \right. \\ \left. + g_{\nu\nu'}^{+}(V_c \pm \hbar \partial_t \phi_q) + g_{\nu\nu'}^{-}(V_c \pm \hbar \partial_t \phi_q) \right], \quad (\text{B3})$$

with the rates

$$g_{\nu\nu'}^{+}(V_c) = \frac{G_{\nu} G_{\nu'}}{G_1 + G_2} \int d\epsilon n_{\nu, V_c}(\epsilon) [1 - n_{\nu'}(\epsilon)], \\ g_{\nu\nu'}^{-}(V_c) = \frac{G_{\nu} G_{\nu'}}{G_1 + G_2} \int d\epsilon [1 - n_{\nu, V_c}(\epsilon)] n_{\nu'}(\epsilon).$$

These rates have the same functional form as the sequential tunneling rates in Eq. (2.5). If ϕ_q fluctuates on classical time scales, i.e., $\hbar \partial_t \phi_q \approx 0$, we see that $S_{\text{tun}}^{(1)}$ has the same structure as the Hamiltonian H_g in Eq. (2.8). Furthermore, $S_{\text{tun}}^{(2)}$ coincides with $S_{\text{tun}}^{(1)}$, except that the rates $g_{\nu\nu'}^{\pm}$ are replaced by the modified rates

$$\tilde{g}_{\nu\nu'}^{+}(V_c) = \frac{G_{\nu} G_{\nu'}}{G_1 + G_2} \int d\epsilon (\mathcal{N}(t, \epsilon) - 1) n_{\nu, V_c} [1 - n_{\nu'}], \\ \tilde{g}_{\nu\nu'}^{-}(V_c) = \frac{G_{\nu} G_{\nu'}}{G_1 + G_2} \int d\epsilon (\mathcal{N}(t, \epsilon) - 1) [1 - n_{\nu, V_c}] n_{\nu'},$$

which are now complicated functions of the dynamical voltage V_c , see Eq. (4.9), of the bias voltage V , and, via the normalization factor $\mathcal{N}$, of the counting field χ . These rates, and thus $S_{\text{tun}}^{(2)}$, vanish in the absence of counting fields, since then $\mathcal{N} = 1$. It is easy to check that they still satisfy the crucial detailed balance condition (2.4), since

$$\frac{\tilde{g}_{\nu\nu'}^{+}(V_c)}{\tilde{g}_{\nu\nu'}^{-}(V_c)} = e^{-\beta(V_c - V_{\nu} + V_{\nu'})}.$$

Finally, $S_{\text{tun}}^{(3)} = -\hbar(G_1 + G_2) \int dt d\epsilon (\mathcal{N}(t, \epsilon) - 1)/2$, which does not contribute to the field dynamics but has to be retained for calculating the current cumulants.

With the fields $\phi^\pm$ in Eq. (4.4), we now observe that S_{tun} satisfies the symmetry property

$$\begin{aligned} S_{\text{tun}} [\phi_t^+, \phi_t^-, \chi_{\nu,t}] = \\ S_{\text{tun}} \left[-\phi_{-t+i\hbar\beta/2}^+, -\phi_{-t-i\hbar\beta/2}^-, -\chi_{\nu,-t} + i\beta V_{\nu,-t} \right], \end{aligned} \quad (\text{B4})$$

which expresses the time reversal invariance of S_{tun} . This relation, together with the invariance of S_c in Eq. (4.2) under the replacement $\phi_t^\pm \rightarrow -\phi_{-t\pm i\hbar\beta/2}^\pm$, when inserted in the Keldysh generating functional (4.1), leads to the quantum generalization of the classical FR (3.2). We notice that in the limit $\hbar \rightarrow 0$, the replacement $\phi_t^\pm \rightarrow -\phi_{-t\pm i\hbar\beta/2}^\pm$ in Eq. (B4) is equivalently written as

$$\begin{aligned} \phi_{q,t} &\rightarrow -\phi_{q,-t} + i\beta V_{c,-t}/2 + \mathcal{O}(\hbar), \\ V_{c,t} &\rightarrow V_{c,-t} + \mathcal{O}(\hbar), \end{aligned}$$

where $\phi_q = (\phi^+ - \phi^-)/2$ and $V_c = \hbar\partial_t(\phi^+ + \phi^-)/2$. Equation (B4) thus recovers the classical relation (3.1) but allows us to extend the validity of the general FR (3.2), which we obtained in Sec. III from the classical generating functional, to the more general setting of the Keldysh quantum generating functional.

Appendix C: Relations between time-dependent response coefficients

In this appendix, we provide some details concerning Sec. V C. The general FR (3.2) for currents under time-dependent driving forces $f_{\nu,t}$ implies that derivatives w.r.t. forces can be exchanged for derivatives w.r.t. counting fields. The time-dependent Onsager coefficients (5.5) can thereby be written as

$$L_{\nu_1, \dots, \nu_k; \nu'_1, \dots, \nu'_l}^{(k,l)}(t_1, \dots, t_k; t'_1, \dots, t'_l) = \prod_{s=1}^k \left(\frac{\delta}{i\delta\chi_{\nu_s, -t_s}} \right) \prod_{j=1}^l \left(i\beta \frac{\delta}{\delta\chi_{\nu'_j, -t'_j}} + \frac{\delta}{\delta f_{\nu'_j, -t'_j}} \right) \ln Z[\chi] \Big|_{\chi=f=0}.$$

Therefore we obtain a connection between coefficients with the same order $k+l$,

$$\begin{aligned} (-1)^k L_{\nu_1, \dots, \nu_k; \nu'_1, \dots, \nu'_l}^{(k,l)}(t_1, \dots, t_k; t'_1, \dots, t'_l) &= \beta^l L_{\nu_1, \dots, \nu_k; \nu'_1, \dots, \nu'_l}^{(k+l,0)}(-t_1, \dots, -t'_l) \\ &+ \beta^{l-1} \sum_{j=1}^l L_{\nu_1, \dots, \nu_k, \nu_1, \dots, \bar{\nu}'_j, \dots, \nu'_l; \nu'_j}^{(k+l-1,1)}(-t_1, \dots, -\bar{t}'_j, \dots, -t'_l; -t'_j) \\ &+ \beta^{l-2} \sum_{j \neq m=1}^l L_{\nu_1, \dots, \nu_k, \nu'_1, \dots, \bar{\nu}'_j, \dots, \bar{\nu}'_m, \dots, \nu'_l; \nu'_j, \nu'_m}^{(k+l-2,2)}(-t_1, \dots, -\bar{t}'_j, \dots, -\bar{t}'_m, \dots, -t'_l; -t'_j, -t'_m) \\ &+ \dots + L_{\nu_1, \dots, \nu_k; \nu'_1, \dots, \nu'_l}^{(k,l)}(-t_1, \dots, -t_k; -t'_1, \dots, -t'_l), \end{aligned} \quad (\text{C1})$$

where $\bar{\nu}'_j$ (resp. $\bar{t}'_j$) means that ν'_j (resp. t'_j) is missing in the string of indices (resp. time arguments).

For instance, the four lowest-order relations resulting from Eq. (C1) are as follows: To first order, $L_{\nu}^{(1,0)}(t) = -L_{\nu}^{(1,0)}(-t)$. The second-order result is

$$\begin{aligned} L_{\nu_1, \nu_2}^{(2,0)}(t_1, t_2) &= L_{\nu_1, \nu_2}^{(2,0)}(-t_1, -t_2) \equiv \hat{T} L_{\nu_1, \nu_2}^{(2,0)}, \\ L_{\nu_1; \nu'_1}^{(1,1)}(t_1; t'_1) &= -\beta L_{\nu_1, \nu'_1}^{(2,0)}(-t_1, -t'_1) - L_{\nu_1; \nu'_1}^{(1,1)}(-t_1, -t'_1) = -\beta \hat{T} L_{\nu_1, \nu'_1}^{(2,0)} - \hat{T} L_{\nu_1; \nu'_1}^{(1,1)}, \end{aligned} \quad (\text{C2})$$

where the time reversal operator $\hat{T}$ inverts all time arguments when acting on a function. For $k+l=3$, we find the relations

$$\begin{aligned} L_{\nu_1, \nu_2, \nu_3}^{(3,0)} &= -\hat{T} L_{\nu_1, \nu_2, \nu_3}^{(3,0)}, \quad L_{\nu_1, \nu_2; \nu'_1}^{(2,1)} = \beta \hat{T} L_{\nu_1, \nu_2, \nu'_1}^{(3,0)} + \hat{T} L_{\nu_1, \nu_2; \nu'_1}^{(2,1)}, \\ L_{\nu_1; \nu'_1, \nu'_2}^{(1,2)} &= -\beta^2 \hat{T} L_{\nu_1, \nu'_1, \nu'_2}^{(3,0)} - \beta \hat{T} \left(L_{\nu_1, \nu'_2; \nu'_1}^{(2,1)} + L_{\nu_1, \nu'_1; \nu'_2}^{(2,1)} \right) - \hat{T} L_{\nu_1; \nu'_1, \nu'_2}^{(1,2)}. \end{aligned} \quad (\text{C3})$$

Finally, the fourth order produces the following relations:

$$\begin{aligned} L_{\nu_1, \nu_2, \nu_3, \nu_4}^{(4,0)} &= \hat{T} L_{\nu_1, \nu_2, \nu_3, \nu_4}^{(4,0)}, \quad L_{\nu_1, \nu_2, \nu_3; \nu'_1}^{(3,1)} = -\beta \hat{T} L_{\nu_1, \nu_2, \nu_3, \nu'_1}^{(4,0)} - \hat{T} L_{\nu_1, \nu_2, \nu_3; \nu'_1}^{(3,1)}, \\ L_{\nu_1, \nu_2; \nu'_1, \nu'_2}^{(2,2)} &= \beta^2 \hat{T} L_{\nu_1, \nu_2, \nu'_1, \nu'_2}^{(4,0)} + \beta \hat{T} \left(L_{\nu_1, \nu_2, \nu'_2; \nu'_1}^{(3,1)} + L_{\nu_1, \nu_2, \nu'_1; \nu'_2}^{(3,1)} \right) + \hat{T} L_{\nu_1, \nu_2; \nu'_1, \nu'_2}^{(2,2)}, \\ L_{\nu_1; \nu'_1, \nu'_2, \nu'_3}^{(1,3)} &= -\beta^3 \hat{T} L_{\nu_1, \nu'_1, \nu'_2, \nu'_3}^{(4,0)} - \beta^2 \hat{T} \left(L_{\nu_1, \nu'_2, \nu'_3; \nu'_1}^{(3,1)} + L_{\nu_1, \nu'_1, \nu'_3; \nu'_2}^{(3,1)} + L_{\nu_1, \nu'_1, \nu'_2; \nu'_3}^{(3,1)} \right) \\ &\quad - \beta \hat{T} \left(L_{\nu_1, \nu'_1; \nu'_2, \nu'_3}^{(2,2)} + L_{\nu_1, \nu'_2; \nu'_1, \nu'_3}^{(2,2)} + L_{\nu_1, \nu'_3; \nu'_1, \nu'_2}^{(2,2)} \right) - \hat{T} L_{\nu_1; \nu'_1, \nu'_2, \nu'_3}^{(1,3)}. \end{aligned} \quad (\text{C4})$$

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⁷¹ The temperatures T_ν^* in Eq. (4.19) may still depend on the variable n . In a quasi-stationary regime with $\partial_t n \approx 0$, we have $n \simeq n(V_\nu, T_\nu)$, and a precise FR involving the effective temperatures T_ν^* can be stated.

⁷² The slight mismatch between the slopes obtained from numerics and from the FR visible in Fig. 1 is caused by the finite size of the window used for the numerical sampling of Q , which in turn is necessary to have reasonable statistical efficiency.