

Nonequilibrium fluctuations for linear diffusion dynamics

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We present the theoretical study on nonequilibrium (NEQ) fluctuations for diffusion dynamics in high dimensions driven by a linear drift force. We consider a general situation in which NEQ is caused by two conditions: (i) drift force not derivable from a potential function, and (ii) diffusion matrix not proportional to the unit matrix, implying nonidentical and correlated multidimensional noise. The former is a well-known NEQ source and the latter can be realized in the presence of multiple heat reservoirs or multiple noise sources. We develop a statistical mechanical theory based on generalized thermodynamic quantities such as energy, work, and heat. The NEQ fluctuation theorems are reproduced successfully. We also find the time-dependent probability distribution function exactly as well as the NEQ work production distribution $P(\mathcal{W})$ in terms of solutions of nonlinear differential equations. In addition, we compute low-order cumulants of the NEQ work production explicitly. In two dimensions, we carry out numerical simulations to check out our analytic results and also to get $P(\mathcal{W})$. We find an interesting dynamic phase transition in the exponential tail shape of $P(\mathcal{W})$, associated with a singularity found in solutions of the nonlinear differential equation. Finally, we discuss possible realizations in experiments.

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I. INTRODUCTION

There has been great interest in nonequilibrium (NEQ) statistical mechanics for the last decades since the discovery of the fluctuation theorem for entropy production. The first discovery was made on a deterministic NEQ dynamics governed by the SLLD equations [1–3]. Later on, the fluctuation theorems of various types were found to be a universal feature for a wide class of NEQ systems, which are governed by both deterministic [4,5] and stochastic dynamics [6–11]. Jarzynski found an interesting relation between NEQ work and equilibrium free energy [12], which was later proved to be a special case of the Crooks fluctuation theorems [7,13]. Since then, a number of stimulated studies have been published up to now regarding the fluctuation theorems and related phenomena [14–20].

The diffusion dynamics is distinguished from the Brownian dynamics. The former has only positionlike state variables that have even parity under the time reversal, while the latter has pairs of position and momentum with even and odd parities, respectively. In this work, we consider the diffusion dynamics with two important conditions which drive the system into a NEQ steady state (NESS): (i) drift force not derivable from a potential function, and (ii) nonidentical and correlated noise. These two conditions can be realized only in high dimensions (not possible in one dimension). Unusual results were reported for the dynamics with the combination of these two conditions [21–23]. In particular it was found that the zero mass limit and the overdamping limit are different in reducing the Kramers equation to the Fokker-Planck equation.

Kwon, Ao, and Thouless [24] studied the diffusion dynamics with a linear drift force in high dimensions and found the probability distribution function (PDF) for the NESS exactly. They found that a circulating probability current can exist at the steady state, violating the detailed balance. One example is a nonzero torque generated in a nano heat engine in contact with two different heat reservoirs [25]. For a nonlinear drift force, it has recently been found via a perturbation theory that there exists an additional current that is absent for the linear case, due to the combination of the force nonlinearity and the multidimensional noise correlation [26]. It moves the probability maximum away from the fixed point at which the force is zero. This novel current has the same origin with the noise-induced current, transporting drugs or molecules in biological systems, studied by Prost *et al.* [27] and Doering *et al.* [28].

We revisit the diffusion dynamics with a linear drift force from the point of view of the NEQ fluctuation theorem, as one of a few analytically solvable cases far from equilibrium. We consider the Langevin equation

$$\dot{\mathbf{q}} = \mathbf{f}(\mathbf{q}) + \boldsymbol{\xi}, \quad (1)$$

where $\mathbf{q} = (q_1, q_2, \dots, q_d)^T$ is a state vector in d dimensions with the superscript T denoting the transpose of a given vector or matrix. We restrict ourselves to the case in which \mathbf{q} has even parity under the time reversal, i.e., there are no momentumlike variables. $\boldsymbol{\xi} = (\xi_1, \xi_2, \dots, \xi_d)^T$ is a white noise vector with zero mean satisfying $\langle \xi_i(t) \xi_j^T(t') \rangle = 2D \delta(t - t')$ where $\langle \dots \rangle$ is the noise average. D is defined as a $d \times d$ diffusion matrix that is symmetric, positive definite, and \mathbf{q} independent. The first NEQ condition (i) is given by the drift force $\mathbf{f} \neq -\nabla\Phi(\mathbf{q})$ where Φ is a scalar function of \mathbf{q} . Condition (ii) leads to the case where the diffusion matrix is not proportional to the unit matrix, $D \not\propto I$, in contrast to the conventional thermal noise with $D \propto (k_B T)I$.

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This Langevin equation describes a general stochastic system far from equilibrium without conventional energy or temperature. So we first define *generalized* thermodynamic quantities, energy, work, and heat, properly. With these definitions, we successfully reproduce the NEQ fluctuation theorems. We can also get analytic expressions for many interesting quantities such as the time-dependent PDF $P(\mathbf{q}, t)$, the two-time correlation functions, and the NEQ work production distribution $P(\mathcal{W})$.

The Jarzynski equality can be shown directly and the cumulants for the NEQ work production \mathcal{W} are calculated explicitly up to the second order. More interestingly, we find the exponential tail shape of $P(\mathcal{W})$ with a power-law prefactor, which undergoes a dynamic phase transition as the time increases. This phase transition turns out to be associated with a singularity of solutions of a nonlinear differential equation (NLDE). We solve this NLDE numerically to reveal the details of the exponential tail shape of $P(\mathcal{W})$ and also its dynamic phase transition.

This paper is organized as follows. In Sec. II, the generalized thermodynamic quantities are defined with the corresponding fluctuation theorems. In Sec. III, we obtain $P(\mathbf{q}, t)$ and the two-time correlation functions exactly. In Sec. IV, we derive the analytic expression for the generating function of $P(\mathcal{W})$ in terms of solutions of the NLDE. In Sec. V, we calculate the cumulants of the work production. In Sec. VI, we take the two-dimensional diffusion dynamics as a simple example and calculate the generating function by solving the NLDE numerically. The dynamic phase transition of the tail shape of $P(\mathcal{W})$ is discussed. We also present the results from the direct numerical integration of the Langevin equation, which agree with the analytical results. Finally, in Sec. VII, we summarize our results and discuss interesting features found for the linear diffusion dynamics in high dimensions and the possibility of realization in experiments.

II. FLUCTUATION THEOREMS FOR THE DIFFUSION DYNAMICS IN HIGH DIMENSIONS

We consider the Fokker-Planck equation for the diffusion dynamics associated with the Langevin equation in Eq. (1),

$$\frac{\partial P(\mathbf{q}, t)}{\partial t} = \nabla \cdot [-\mathbf{f}(\mathbf{q}) + \mathbf{D} \cdot \nabla] P(\mathbf{q}, t), \quad (2)$$

where we use the dot notation for the product of a vector and a matrix, or between vectors, but not between matrices. For example, we write $\mathbf{q}^T \cdot \mathbf{A} \mathbf{B} \cdot \mathbf{q}$ where \mathbf{A}, \mathbf{B} are matrices.

Writing the steady state solution as $P_{st} \propto e^{-\Phi_{st}(\mathbf{q})}$, we define equilibrium as the steady state satisfying the detailed balance such that

$$\Pi(\mathbf{q}', t'; \mathbf{q}, t) e^{-\Phi_{st}(\mathbf{q})} = \Pi(\mathbf{q}, t; \mathbf{q}', t') e^{-\Phi_{st}(\mathbf{q}')}, \quad (3)$$

where $\Pi(\mathbf{q}', t'; \mathbf{q}, t)$ is the conditional probability for the transition from state \mathbf{q} at time t to state \mathbf{q}' at time t' .

We can show that the necessary and sufficient condition for the detailed balance reads as

$$\mathbf{f}(\mathbf{q}) = -\mathbf{D} \cdot \nabla \Phi_{st}(\mathbf{q}), \quad (4)$$

i.e., the vanishing probability current $\mathbf{j} = (\mathbf{f} - \mathbf{D} \cdot \nabla) P_{st} = 0$ at the steady state. Defining the force matrix \mathbf{F} as

$$F_{\alpha\beta} = -\nabla_{\beta} f_{\alpha}, \quad (5)$$

with $\nabla_{\beta} \equiv \frac{\partial}{\partial q_{\beta}}$, we can rewrite the detailed balance condition as

$$\mathbf{F} \mathbf{D} = (\mathbf{F} \mathbf{D})^T = \mathbf{D} \mathbf{F}^T, \quad (6)$$

where $\nabla_{\alpha} \nabla_{\beta} \Phi_{st} = \nabla_{\beta} \nabla_{\alpha} \Phi_{st}$ is used.

The detailed balance condition is always satisfied in one dimension. In higher dimensions, however, this does not hold in general due to two possible sources. One is the asymmetry of \mathbf{F} ($\mathbf{F} \neq \mathbf{F}^T$), which happens when \mathbf{f} is not derivable from a scalar potential. Another comes from the diffusion matrix, which is not proportional to the unit matrix, so the noises are not identical in components ($D_{\alpha\alpha} \neq D_{\beta\beta}$ for $\alpha \neq \beta$) and also may be correlated ($D_{\alpha\beta} \neq 0$).

We note that the detailed balance condition is satisfied (subsequently, equilibrium can be achieved) not only with symmetric \mathbf{F} and $\mathbf{D} \propto \mathbf{I}$, but also with the specific combination of general \mathbf{F} and $\mathbf{D} \not\propto \mathbf{I}$ satisfying Eq. (6). In equilibrium, the steady-state distribution should be given by the Boltzmann distribution, so Φ_{st} can be interpreted as *energy* (we set $k_B T \equiv 1$ for convenience). Then it is natural to define a generalized *force* as the negative derivative of the energy function, i.e., $\mathbf{D}^{-1} \cdot \mathbf{f} = -\nabla \Phi_{st}$ from Eq. (4), especially for $\mathbf{D} \propto \mathbf{I}$.

In general, the detailed balance condition is not satisfied and the system is driven into a NESS. In this case, Eq. (4) should be modified as

$$\mathbf{D}^{-1} \cdot \mathbf{f} = -\nabla \Phi(\mathbf{q}) + \mathbf{g}(\mathbf{q}), \quad (7)$$

with nonzero $\mathbf{g}(\mathbf{q})$ which cannot be derivable from a scalar function [$\mathbf{g}(\mathbf{q}) \neq -\nabla \Psi(\mathbf{q})$]. We interpret \mathbf{g} as a generalized NEQ driving force. However, there is no unique way of defining the NEQ driving force \mathbf{g} as well as the energylike function Φ . As the NESS need not be governed by the Boltzmann distribution, the energy function $\Phi(\mathbf{q})$ does not have to be the same as $\Phi_{st}(\mathbf{q})$ in general. In fact, one may choose an arbitrary $\Phi(\mathbf{q})$ and $\mathbf{g}(\mathbf{q})$ accordingly in the system described only by the stochastic equations like Eqs. (1) and (2). We will come back to this issue later.

Following the path integral formalism of Onsager and Machlup [29], the conditional probability that the system evolves along a path $\mathbf{q}(\tau)$ for $0 \leq \tau \leq t$ starting from an initial state $\mathbf{q}(0)$ is given by

$$\Pi[\mathbf{q}(t)]_0^t \propto e^{-(1/4) \int_0^t d\tau [\dot{\mathbf{q}}(\tau) - \mathbf{f}(\mathbf{q}(\tau))]^T \cdot \mathbf{D}^{-1} \cdot [\dot{\mathbf{q}}(\tau) - \mathbf{f}(\mathbf{q}(\tau))]} \quad (8)$$

The time-reverse path is given by $\bar{\mathbf{q}}(\tau) = \mathbf{q}(t - \tau)$ with the initial state $\bar{\mathbf{q}}(0) = \mathbf{q}(t)$. Then the ratio of the conditional probability for the forward path $\mathbf{q}(\tau)$ to that for the reverse path $\bar{\mathbf{q}}(\tau)$ is found as

$$\begin{aligned} \frac{\Pi[\mathbf{q}(t)]_0^t}{\Pi[\bar{\mathbf{q}}(t)]_0^t} &= e^{\int_0^t d\tau \dot{\mathbf{q}}^T \cdot \mathbf{D}^{-1} \cdot \mathbf{f}} = e^{-\int_0^t d\tau \dot{\mathbf{q}}^T \cdot (\nabla \Phi - \mathbf{g})} \\ &= e^{-\Delta \Phi + \mathcal{W}[\mathbf{q}]} = e^{-\mathcal{Q}[\mathbf{q}]}, \end{aligned} \quad (9)$$

where $\Delta\Phi = \Phi[\mathbf{q}(t)] - \Phi[\mathbf{q}(0)]$ is the energy difference between the final and initial time. We interpret $\mathcal{W}[\mathbf{q}]$ as the work production done by the NEQ force \mathbf{g} along the path $\mathbf{q}(\tau)$,

$$\mathcal{W}[\mathbf{q}] = \int_0^t d\tau \left(\dot{\mathbf{q}}^T \cdot \mathbf{g} + \frac{\partial\Phi}{\partial\tau} \right). \quad (10)$$

The second term appears only when Φ has an explicit time dependence, which is the case of the Jarzynski type work [12].

Accordingly we define $\mathcal{Q} \equiv \Delta\Phi - \mathcal{W}$ and interpret it as the heat transferred into the system from the reservoir along the path. We can rewrite

$$\mathcal{Q}[\mathbf{q}] = \int_0^t d\tau (-\dot{\mathbf{q}}^T \cdot \mathbf{D}^{-1} \cdot \dot{\mathbf{q}} + \dot{\mathbf{q}}^T \cdot \boldsymbol{\zeta}) \quad (11)$$

where $\boldsymbol{\zeta} = \mathbf{D}^{-1} \cdot (\dot{\mathbf{q}} - \mathbf{f}) = \mathbf{D}^{-1} \cdot \boldsymbol{\xi}$. This definition of heat can be understood in terms of the corresponding Brownian dynamics. The diffusion dynamics can be regarded as the Brownian dynamics in the overdamped limit (or zero inertia limit). In this case, the corresponding Brownian dynamics is given by

$$m\ddot{\mathbf{q}} = -\gamma \cdot \dot{\mathbf{q}} + \mathbf{D}^{-1} \cdot \mathbf{f} + \boldsymbol{\zeta}, \quad (12)$$

where the friction matrix $\gamma = \mathbf{D}^{-1}$ and the diffusion matrix for the noise $\boldsymbol{\zeta} = \mathbf{D}^{-1}\boldsymbol{\xi}$ becomes $\mathbf{D}_\zeta = \mathbf{D}^{-1}\mathbf{D}\mathbf{D}^{-1} = \mathbf{D}^{-1}$. Therefore the generalized Einstein relation is satisfied ($\mathbf{D}_\zeta = k_B T \gamma$ with our presetting of $k_B T = 1$). As noticed, $\mathbf{D}^{-1} \cdot \mathbf{f}$ plays the role of force. From the point of view of the Brownian dynamics, $\boldsymbol{\zeta} = \mathbf{D}^{-1} \cdot \boldsymbol{\xi}$ is a random diffusive force exerted by noise. Then \mathcal{Q} is the sum of dissipative work due to the random collisions and the diffusive work due to noise, known as the Onsager heat. Therefore the interpretation of \mathcal{Q} as the heat production is reasonable.

Up to now, we introduced the appropriate definitions of generalized energy, work, and heat for an arbitrary diffusion dynamics without conventional thermodynamic quantities. Note that the work and the heat production depend on the path, i.e., functionals of the path, while the energy is a state function and independent of the path. One may restore the temperature by scaling the diffusion matrix as $\mathbf{D} \rightarrow (k_B T)\mathbf{D}$, then Φ_{st} scales as $\Phi_{st}/(k_B T)$. $k_B T$ can parametrize the overall strength of noise and can be interpreted as an effective temperature, if necessary. There are other temperaturelike parameters, the eigenvalues of \mathbf{D} , which may be different each other. The presence of multi and heterogeneous temperatures might also cause NEQ, though not always.

Equation (9) is the fundamental equation from which various fluctuation theorems can be derived. It replaces the detailed balance relation for equilibrium, so it is referred to as the detailed fluctuation relation or the generalized detailed balance relation for NEQ. If we choose an initial PDF $\propto e^{-\Phi(\mathbf{q})}$ [Boltzmann distribution with the energy function $\Phi(\mathbf{q})$], we can easily show the Crooks fluctuation theorem [6]:

$$\langle \mathcal{A}[\mathbf{q}] \rangle_F = \langle \hat{\mathcal{A}}[\bar{\mathbf{q}}] e^{\hat{\mathcal{W}}[\bar{\mathbf{q}}]} \rangle_R e^{-\Delta\mathcal{F}}, \quad (13)$$

where $\mathcal{A}[\mathbf{q}]$ is any functional of the path $\mathbf{q}(\tau)$ and $\hat{\mathcal{A}}[\bar{\mathbf{q}}] \equiv \mathcal{A}[\mathbf{q}]$. The free energy \mathcal{F} is defined as $e^{-\mathcal{F}} \equiv \int dq e^{-\Phi}$ and Eq. (10) guarantees that $\hat{\mathcal{W}}[\bar{\mathbf{q}}] = -\mathcal{W}[\bar{\mathbf{q}}]$. The subscripts F and R denote the averages along the forward $\mathbf{q}(\tau)$ and the reverse $\bar{\mathbf{q}}(\tau)$ path, respectively. The free energy difference

$\Delta\mathcal{F} = \mathcal{F}(t) - \mathcal{F}(0)$ does not vanish only when Φ has an explicit time dependence ($\frac{\partial\Phi}{\partial t} \neq 0$). In this paper, we assume no explicit time dependence in \mathbf{f} and \mathbf{D} , so we always find $\Delta\mathcal{F} = 0$.

III. TRANSIENT STATE FAR FROM STEADY STATE FOR LINEAR DIFFUSION DYNAMICS

From now on, we focus on the case of a linear drift force,

$$\mathbf{f} = -\mathbf{F} \cdot \mathbf{q}, \quad (14)$$

which can be analytically tractable. The force matrix \mathbf{F} is constant of state \mathbf{q} and time t . The exact steady state probability distribution $P_{st} \propto e^{-\Phi_{st}}$ was found by Kwon *et al.* [24] with

$$\Phi_{st} = \frac{1}{2} \mathbf{q}^T \cdot \mathbf{U} \cdot \mathbf{q}, \quad (15)$$

where the symmetric matrix $\mathbf{U} = \mathbf{U}^T$ is given by

$$\mathbf{U} = (\mathbf{D} + \mathbf{Q})^{-1} \mathbf{F} \quad (16)$$

with the antisymmetric matrix $\mathbf{Q} = -\mathbf{Q}^T$ satisfying

$$\mathbf{F}\mathbf{Q} + \mathbf{Q}\mathbf{F}^T = \mathbf{F}\mathbf{D} - \mathbf{D}\mathbf{F}^T. \quad (17)$$

The equilibrium detailed balance condition, Eq. (6), yields $\mathbf{Q} = \mathbf{0}$ and $\mathbf{U} = \mathbf{D}^{-1}\mathbf{F}$, which satisfies Eq. (4) as expected. Thus the existence of nonzero \mathbf{Q} implies the breaking of the detailed balance and results in the nonzero steady state current as $\mathbf{j}_{st} = -(\mathbf{Q} \cdot \nabla \Phi_{st}) P_{st}$. The general solution for \mathbf{Q} can be found in a series form by using the Jordan transformation for asymmetric \mathbf{F} or in an integral form in the frequency space [24,26].

The time-dependent PDF solution for the Fokker-Planck equation of Eq. (2) can be formally found from the path integral, using Eq. (8), as

$$P(\mathbf{q}, t; \mathbf{l}) = \int d\mathbf{q}(0) P[\mathbf{q}(0)] \int D[\mathbf{q}] e^{-\int_0^t d\tau (L(\mathbf{q}, \dot{\mathbf{q}}) - \mathbf{l}^T \cdot \dot{\mathbf{q}})}, \quad (18)$$

where $P[\mathbf{q}(0)]$ is the initial PDF and the Lagrangian L reads as

$$L(\mathbf{q}, \dot{\mathbf{q}}) = \frac{1}{4} (\dot{\mathbf{q}} + \mathbf{F} \cdot \mathbf{q})^T \cdot \mathbf{D}^{-1} \cdot (\dot{\mathbf{q}} + \mathbf{F} \cdot \mathbf{q}). \quad (19)$$

$\int D[\mathbf{q}] \dots$ denotes the integration over all paths reaching a fixed final state $\mathbf{q}(t)$ at time t , starting from an initial state $\mathbf{q}(0)$, with proper normalizations. The source field $\mathbf{l}(\tau)$ is introduced for later use to generate time correlation functions and moments of \mathbf{q} .

We calculate explicitly the time-dependent PDF $P(\mathbf{q}, t) = P(\mathbf{q}, t; \mathbf{l} = \mathbf{0})$ with the initial Gaussian PDF of $P[\mathbf{q}(0)] \propto e^{-(1/2)\mathbf{q}^T(0)\mathbf{A}(0)\mathbf{q}(0)}$ with a symmetric $\mathbf{A}(0)$. The detailed calculation steps are given in Appendix A. From Eq. (A27), we get

$$P(\mathbf{q}, t) = |\det[2\pi\mathbf{A}^{-1}(t)]|^{-1/2} e^{-(1/2)\mathbf{q}^T \cdot \mathbf{A}(t) \cdot \mathbf{q}}, \quad (20)$$

where the symmetric matrix $\mathbf{A}^{-1}(t)$ is given from Eq. (A23) as

$$\mathbf{A}^{-1}(t) = \mathbf{U}^{-1} + e^{-\mathbf{F}t} [\mathbf{A}^{-1}(0) - \mathbf{U}^{-1}] e^{-\mathbf{F}^T t}. \quad (21)$$

As $\lim_{t \rightarrow \infty} \mathbf{A}^{-1}(t) = \mathbf{U}^{-1}$ for positive definite \mathbf{F} , the steady-state solution of Eq. (15) is recovered.

The differential equation for \mathbf{A}^{-1} can be derived from the recursion relation in Eq. (A18) as [30]

$$\frac{d\mathbf{A}^{-1}}{dt} = 2\mathbf{D} - \mathbf{F}\mathbf{A}^{-1} - \mathbf{A}^{-1}\mathbf{F}^T. \quad (22)$$

One can easily show that $\mathbf{A}^{-1}(t)$ in Eq. (21) is the solution of this differential equation.

Now we define the generating functional for time-correlation functions and cumulants of \mathbf{q} as

$$Z[\mathbf{l}(\tau)] = \int d\mathbf{q} P[\mathbf{q}, t; \mathbf{l}(\tau)] = e^{(1/2) \int d\tau \int d\tau' \mathbf{l}^T(\tau) \Gamma(\tau, \tau') \mathbf{l}(\tau')} \quad (23)$$

In Appendix B, we find from Eqs. (B6) and (B7)

$$\Gamma(\tau, \tau') = \begin{cases} e^{-(\tau-\tau')\mathbf{F}} \mathbf{A}^{-1}(\tau'), & \tau > \tau', \\ \mathbf{A}^{-1}(\tau) e^{-(\tau'-\tau)\mathbf{F}^T}, & \tau' > \tau, \end{cases} \quad (24)$$

where $\Gamma(\tau, \tau') = \Gamma^T(\tau', \tau)$. Then we can compute the time average of any functional \mathcal{A} of path $\mathbf{q}(\tau)$:

$$\langle \mathcal{A}[\mathbf{q}(\tau)] \rangle = \mathcal{A} \left[\frac{\delta}{\delta \mathbf{l}(\tau)} \right] Z[\mathbf{l}] \Big|_{\mathbf{l} \rightarrow \mathbf{0}}, \quad (25)$$

with $Z[\mathbf{0}] = 1$. For example, we get the two-time correlation function as

$$\begin{aligned} \langle q_\alpha(\tau) q_\beta(\tau') \rangle &= \frac{\delta}{\delta l_\alpha(\tau)} \frac{\delta}{\delta l_\beta(\tau')} Z[\mathbf{l}] \Big|_{\mathbf{l} \rightarrow \mathbf{0}} \\ &= \Gamma_{\alpha\beta}(\tau, \tau') = \Gamma_{\beta\alpha}(\tau', \tau). \end{aligned} \quad (26)$$

IV. NONEQUILIBRIUM WORK PRODUCTION

The work production by the NEQ force \mathbf{g} along the path $\mathbf{q}(\tau)$ is given by Eq. (10) with $\frac{\partial \Phi}{\partial \tau} = 0$. As discussed in Sec. II, we have some arbitrariness in choosing the energy functional $\Phi(\mathbf{q})$, thus also the NEQ force $\mathbf{g}(\mathbf{q})$ in Eq. (7).

In the case of the linear drift force with $\mathbf{D}^{-1} \cdot \mathbf{f} = -\mathbf{D}^{-1}\mathbf{F} \cdot \mathbf{q}$, Eq. (7) becomes

$$(\mathbf{D}^{-1}\mathbf{F})_{\alpha\beta} = \nabla_\beta \nabla_\alpha \Phi - \nabla_\beta g_\alpha. \quad (27)$$

If $\mathbf{D}^{-1}\mathbf{F}$ is symmetric, the detailed balance condition, Eq. (6), is satisfied and one may choose $\Phi = \frac{1}{2} \mathbf{q}^T \cdot (\mathbf{D}^{-1}\mathbf{F}) \cdot \mathbf{q}$ with $\mathbf{g} = \mathbf{0}$. So we get no NEQ work production with this choice of the energy function, as expected.

When $\mathbf{D}^{-1}\mathbf{F}$ is not symmetric, we must have a nonzero NEQ force \mathbf{g} . The energy function $\Phi(\mathbf{q})$ can be written in general as

$$\Phi(\mathbf{q}) = \frac{1}{2} \mathbf{q}^T \cdot \mathbf{G}_s \cdot \mathbf{q}, \quad (28)$$

with a symmetric matrix $\mathbf{G}_s = \mathbf{G}_s^T$, which is a part of $\mathbf{D}^{-1}\mathbf{F}$. Then we can divide $\mathbf{D}^{-1}\mathbf{F}$ into the symmetric part \mathbf{G}_s and the remainder \mathbf{G}_a :

$$\mathbf{D}^{-1}\mathbf{F} = \mathbf{G}_s + \mathbf{G}_a, \quad (29)$$

and the NEQ driving force is given as

$$\mathbf{g}(\mathbf{q}) = -\mathbf{G}_a \cdot \mathbf{q}. \quad (30)$$

There is no unique way to determine \mathbf{G}_s or \mathbf{G}_a out of $\mathbf{D}^{-1}\mathbf{F}$ [31]. One natural possible choice is to enforce \mathbf{G}_a antisymmetric ($\mathbf{G}_a = -\mathbf{G}_a^T$), such as

$$\begin{aligned} \mathbf{G}_s &= \frac{1}{2} (\mathbf{D}^{-1}\mathbf{F} + \mathbf{F}^T \mathbf{D}^{-1}) \equiv \bar{\mathbf{G}}_s, \\ \mathbf{G}_a &= \frac{1}{2} (\mathbf{D}^{-1}\mathbf{F} - \mathbf{F}^T \mathbf{D}^{-1}) \equiv \bar{\mathbf{G}}_a, \end{aligned} \quad (31)$$

which will be called the *antisymmetric* (AS) choice.

Another interesting choice, called as the *steady-state* (SS) choice, is

$$\mathbf{G}_s = \mathbf{U}, \quad \mathbf{G}_a = \mathbf{D}^{-1}\mathbf{Q}\mathbf{U}. \quad (32)$$

If we take an initial Boltzmann distribution with this energy function, the system stays in the NESS from the beginning.

In general, one can choose

$$\begin{aligned} \mathbf{G}_s &= \bar{\mathbf{G}}_s + \delta\mathbf{G}_s, \\ \mathbf{G}_a &= \bar{\mathbf{G}}_a - \delta\mathbf{G}_s, \end{aligned} \quad (33)$$

with an arbitrary symmetric matrix $\delta\mathbf{G}_s$. From Eq. (10), the NEQ work production during time t is given by

$$\mathcal{W}[\mathbf{q}] = - \int_0^t d\tau \dot{\mathbf{q}}^T \cdot \mathbf{G}_a \cdot \mathbf{q} = \bar{\mathcal{W}} + \delta\mathcal{W}, \quad (34)$$

where $\bar{\mathcal{W}}$ is the NEQ work production in the AS choice,

$$\bar{\mathcal{W}}[\mathbf{q}] = - \int_0^t d\tau \dot{\mathbf{q}}^T \cdot \bar{\mathbf{G}}_a \cdot \mathbf{q}. \quad (35)$$

The contribution $\delta\mathcal{W}$ from the additive symmetric matrix $\delta\mathbf{G}_s$ is given by

$$\begin{aligned} \delta\mathcal{W} &= \int_0^t d\tau \dot{\mathbf{q}}^T \cdot \delta\mathbf{G}_s \cdot \mathbf{q} \\ &= \frac{1}{2} \mathbf{q}^T(t) \cdot \delta\mathbf{G}_s \cdot \mathbf{q}(t) - \frac{1}{2} \mathbf{q}^T(0) \cdot \delta\mathbf{G}_s \cdot \mathbf{q}(0), \end{aligned} \quad (36)$$

which comes only from boundaries and also exactly compensates the additional energy term due to $\delta\mathbf{G}_s$ in the energy function Φ .

It is important to note that the heat $\mathcal{Q} = \Delta\Phi - \mathcal{W}$ is independent of the choice of $\delta\mathbf{G}_s$, in contrast to the NEQ work \mathcal{W} . In the long-time limit, $\delta\mathcal{W}$ becomes negligible as the NEQ work usually increases incessantly in time. Thus the main contribution to the NEQ work production in the steady state comes from the purely antisymmetric part $\bar{\mathcal{W}}$.

We now consider the generating function $\mathcal{G}(\lambda)$ for the PDF of the NEQ work production $P(\mathcal{W})$ as

$$\begin{aligned} \mathcal{G}(\lambda) &= \langle e^{-\lambda\mathcal{W}} \rangle \\ &= \int d\mathbf{q}(t) d\mathbf{q}(0) P[\mathbf{q}(0)] \int D[\mathbf{q}] e^{-\int_0^t d\tau L(\mathbf{q}, \dot{\mathbf{q}}) - \lambda\mathcal{W}[\mathbf{q}]}, \\ &= \int d\mathcal{W} P(\mathcal{W}) e^{-\lambda\mathcal{W}}, \end{aligned} \quad (37)$$

with the initial equilibrium Boltzmann distribution $P[\mathbf{q}(0)] \propto e^{-(1/2)\mathbf{q}^T(0) \cdot \mathbf{G}_s \cdot \mathbf{q}(0)}$. The PDF of the work production $P(\mathcal{W})$ can be obtained formally by

$$P(\mathcal{W}) = \int \frac{d\lambda}{2\pi} e^{i\lambda\mathcal{W} + \ln \mathcal{G}(i\lambda)}. \quad (38)$$

We calculate $\mathcal{G}(\lambda)$ in a similar way in which the path integral is computed in Appendix A for the time-dependent PDF, $P(\mathbf{q}, t)$. As \mathcal{W} is quadratic in \mathbf{q} , the integral in Eq. (37) is basically the same as the integral in Eq. (18) except for the final integral over $\mathbf{q}(t)$ with the *modified* Lagrangian as

$$L = \frac{1}{4}(\dot{\mathbf{q}} + \tilde{\mathbf{F}} \cdot \mathbf{q})^T \cdot \mathbf{D}^{-1} \cdot (\dot{\mathbf{q}} + \tilde{\mathbf{F}} \cdot \mathbf{q}) + \frac{1}{2}\mathbf{q}^T \cdot \Lambda \cdot \mathbf{q}, \quad (39)$$

where

$$\tilde{\mathbf{F}} = \mathbf{F} - 2\lambda\mathbf{D}\tilde{\mathbf{G}}_a, \quad (40)$$

$$\begin{aligned} \Lambda &= \frac{1}{2}(\mathbf{F}^T\mathbf{D}^{-1}\mathbf{F} - \tilde{\mathbf{F}}^T\mathbf{D}^{-1}\tilde{\mathbf{F}}) \\ &= \lambda(\mathbf{F}^T\tilde{\mathbf{G}}_a - \tilde{\mathbf{G}}_a\mathbf{F} + 2\lambda\tilde{\mathbf{G}}_a\mathbf{D}\tilde{\mathbf{G}}_a). \end{aligned} \quad (41)$$

The contribution from $e^{-\lambda\delta\mathcal{W}}$ only modifies the initial and final distribution according to Eq. (36).

Before going further, we briefly comment on the discrete-time representation of the path integral and the work \mathcal{W} . In Appendix A, we perform the path integral in the discrete-time representation. The choice of the $\mathbf{q}(\tau)$ value between the discrete time interval Δt does not affect the PDF at final time t in the limit of $\Delta t \rightarrow 0$. However, it is well known that one should choose the midpoint \mathbf{q} value for the definition of the work for the correct description [33]. So $\tilde{\mathcal{W}}$ in Eq. (35) should be written as

$$\begin{aligned} \tilde{\mathcal{W}} &= -\frac{1}{2} \sum_{i=1}^N (\mathbf{q}_i - \mathbf{q}_{i-1})^T \cdot \tilde{\mathbf{G}}_a \cdot (\mathbf{q}_i + \mathbf{q}_{i-1}) \\ &= -\sum_{i=1}^N (\mathbf{q}_i - \mathbf{q}_{i-1})^T \cdot \tilde{\mathbf{G}}_a \cdot \mathbf{q}_{i-1}, \end{aligned} \quad (42)$$

where $\mathbf{q}_i = \mathbf{q}(t_i)$ for $i = 0, \dots, N$ with $\Delta t = t/N$. Note that it is first expressed in the midpoint representation but becomes identical to the so-called prepoint representation due to the antisymmetry of $\tilde{\mathbf{G}}_a$. In Appendix A, all calculations are done in the prepoint representation for convenience.

First, we perform the path integral of Eq. (37) without the final integral over $\mathbf{q}(t)$. The integration procedure is basically identical to the case for the time-dependent PDF calculation in Appendix A except for the different initial condition and the modified Lagrangian. Let $\tilde{\mathbf{A}}(t; \lambda)$ be the modified kernel for $\mathbf{A}(t)$ of Eq. (20). In the discrete-time representation, the recursion relation Eq. (A18) in Appendix A is modified as

$$\tilde{\mathbf{A}}_i^{-1} = 2\Delta t\mathbf{D} + \tilde{\mathbf{V}}(\tilde{\mathbf{A}}_{i-1} + \Delta t\Lambda)^{-1}\tilde{\mathbf{V}}^T, \quad (43)$$

with $\tilde{\mathbf{V}} = 1 - \Delta t\tilde{\mathbf{F}}$. Taking $\Delta t \rightarrow 0$ limit, the differential equation for $\tilde{\mathbf{A}}^{-1}$ can be derived,

$$\frac{d\tilde{\mathbf{A}}^{-1}}{dt} = 2\mathbf{D} - \tilde{\mathbf{F}}\tilde{\mathbf{A}}^{-1} - \tilde{\mathbf{A}}^{-1}\tilde{\mathbf{F}}^T - \tilde{\mathbf{A}}^{-1}\Lambda\tilde{\mathbf{A}}^{-1}. \quad (44)$$

In contrast to Eq. (22), this is a nonlinear differential matrix equation, which cannot be solved analytically in general. Using $d\tilde{\mathbf{A}}^{-1}/dt = -\tilde{\mathbf{A}}^{-1}(d\tilde{\mathbf{A}}/dt)\tilde{\mathbf{A}}^{-1}$, we can rewrite this equation as

$$\frac{d\tilde{\mathbf{A}}}{dt} = -2\tilde{\mathbf{A}}\mathbf{D}\tilde{\mathbf{A}} + \tilde{\mathbf{A}}\tilde{\mathbf{F}} + \tilde{\mathbf{F}}^T\tilde{\mathbf{A}} + \Lambda. \quad (45)$$

The initial condition is given as $\tilde{\mathbf{A}}(0; \lambda) = \tilde{\mathbf{G}}_s + (1 - \lambda)\delta\mathbf{G}_s$, where the λ -dependent term comes from $\delta\mathcal{W}$. We solve this equation numerically for a specific case in Sec. VI.

We obtain $\mathcal{G}(\lambda)$ after integrating over the final $\mathbf{q}(t) = \mathbf{q}_N$ in Eq. (37), leading to

$$\mathcal{G}(\lambda) = \left| \frac{\det(\tilde{\mathbf{A}}_N + \lambda\delta\mathbf{G}_s)}{\det(\tilde{\mathbf{G}}_s + \delta\mathbf{G}_s)} \right|^{-1/2} \prod_{i=0}^{N-1} \left| \frac{\det(\tilde{\mathbf{A}}_i + \Delta t\Lambda)}{\det(\tilde{\mathbf{A}}_{i+1})} \right|^{-1/2}. \quad (46)$$

Using $\tilde{\mathbf{A}}_{i+1} = \tilde{\mathbf{A}}_i + (\Delta t)d\tilde{\mathbf{A}}_i/dt$ for the denominator, we finally get

$$\begin{aligned} \ln \mathcal{G}(\lambda) &= -\frac{1}{2} \int_0^t d\tau \text{Tr} \left(\Lambda - \frac{d\tilde{\mathbf{A}}(\tau; \lambda)}{d\tau} \right) \tilde{\mathbf{A}}^{-1}(\tau; \lambda) \\ &\quad - \frac{1}{2} \ln \left(\frac{\det[\tilde{\mathbf{A}}(t; \lambda) + \lambda\delta\mathbf{G}_s]}{\det(\tilde{\mathbf{G}}_s + \delta\mathbf{G}_s)} \right) \\ &= -\int_0^t d\tau \text{Tr}(\tilde{\mathbf{A}}\mathbf{D} - \tilde{\mathbf{F}}) \\ &\quad - \frac{1}{2} \ln \left(\frac{\det[\tilde{\mathbf{A}}(t; \lambda) + \lambda\delta\mathbf{G}_s]}{\det(\tilde{\mathbf{G}}_s + \delta\mathbf{G}_s)} \right). \end{aligned} \quad (47)$$

It is not possible to perform the integral of Eq. (38) to find $P(\mathcal{W})$ in a closed form. However, the explicit form of $\mathcal{G}(\lambda)$ reveals many interesting properties of $P(\mathcal{W})$. For example, as $\ln \mathcal{G}$ is not quadratic in λ , $P(\mathcal{W})$ is not Gaussian in general. Due to the logarithmic boundary term, the divergence $\mathcal{G}(\lambda)$ may appear in λ , which determines the asymptotic behavior of a non-Gaussian tail of $P(\mathcal{W})$ for large $|\mathcal{W}|$. This will be investigated in more detail in Sec. VI.

The fluctuation theorem yields

$$\mathcal{G}(\lambda) = \mathcal{G}(1 - \lambda) \quad (48)$$

by substituting \mathcal{A} with $e^{-\lambda\mathcal{W}}$ in Eq. (13). The work $\mathcal{W}[\bar{\mathbf{q}}]$ in the reverse path should be the same as $-\mathcal{W}[\mathbf{q}]$ in the forward path, and the forward and reverse paths are identical with the same initial conditions with the same energy function. It does not seem easy to prove Eq. (48) for general λ , directly from Eq. (47).

However, we can prove $\mathcal{G}(1) = \langle e^{-\mathcal{W}} \rangle = 1$ easily, which corresponds to the Jarzynski equality for a time-dependent potential. For $\lambda = 1$, $\tilde{\mathbf{A}}(0) = \tilde{\mathbf{G}}_s$, $\tilde{\mathbf{F}} = \mathbf{D}\tilde{\mathbf{G}}_s - \mathbf{D}\tilde{\mathbf{G}}_a$, and $\Lambda = \tilde{\mathbf{G}}_s\mathbf{D}\tilde{\mathbf{G}}_a - \tilde{\mathbf{G}}_a\mathbf{D}\tilde{\mathbf{G}}_s$. We can show that the initial state is the fixed point of Eq. (44) or Eq. (45), i.e., $\tilde{\mathbf{A}}(t) = \tilde{\mathbf{G}}_s$. Then the logarithmic part vanishes in Eq. (47). We can also see $\text{Tr}(\tilde{\mathbf{A}}\mathbf{D} - \tilde{\mathbf{F}}) = \text{Tr}\mathbf{D}\tilde{\mathbf{G}}_a = 0$. Hence $\mathcal{G}(1) = 1$.

V. CUMULANTS OF NEQ WORK PRODUCTION

In this section, we calculate the cumulants of the work production by using the two-time correlation function $\Gamma(\tau, \tau')$ in Eqs. (24) and (26). For simplicity, we take the AS choice where $\mathbf{G}_a = \tilde{\mathbf{G}}_a$, $\delta\mathbf{G}_s = 0$, $\delta\mathcal{W} = 0$, and $\mathcal{W} = \tilde{\mathcal{W}}$. However, in the long-time limit, all results are choice independent.

First, consider the first cumulant of \mathcal{W} in the discrete-time representation, Eq. (42), as

$$\begin{aligned}
 \langle \mathcal{W} \rangle &= - \sum_{i=1}^N \langle \mathbf{q}_i^T \cdot \bar{\mathbf{G}}_a \cdot \mathbf{q}_{i-1} \rangle \\
 &= \sum_{i=1}^N \text{Tr} \Gamma_{i,i-1} \bar{\mathbf{G}}_a \\
 &= \sum_{i=1}^N \text{Tr} (1 - \Delta t \mathbf{F}) \mathbf{A}_{i-1}^{-1} \bar{\mathbf{G}}_a \\
 &= - \sum_{i=1}^N (\Delta t) \text{Tr} \mathbf{F} \mathbf{A}_{i-1}^{-1} \bar{\mathbf{G}}_a \\
 &\rightarrow - \int_0^t d\tau \text{Tr} \mathbf{A}^{-1}(\tau) \bar{\mathbf{G}}_a \mathbf{F}, \quad (49)
 \end{aligned}$$

where we use $\mathbf{q}_{i-1}^T \cdot \bar{\mathbf{G}}_a \cdot \mathbf{q}_{i-1} = 0$, $\Gamma_{i,i-1} = (1 - \Delta t \mathbf{F}) \mathbf{A}_{i-1}^{-1}$, and Eq. (26).

In the long-time limit, we can replace \mathbf{A}^{-1} by \mathbf{U}^{-1} and find

$$\begin{aligned}
 \langle \mathcal{W} \rangle &\rightarrow -t \text{Tr} \mathbf{U}^{-1} \bar{\mathbf{G}}_a \mathbf{F} \\
 &= -t \text{Tr} \mathbf{Q} \bar{\mathbf{G}}_a \\
 &= t \text{Tr} \mathbf{Q} \mathbf{F}^T \mathbf{D}^{-1}, \quad (50)
 \end{aligned}$$

which should be choice independent. Since the energy difference $\langle \Delta \Phi \rangle$ is finite, $\langle \mathcal{W} \rangle \simeq -\langle \mathcal{Q} \rangle$ measures the entropy production piled up in the reservoir with the mean rate

$$\langle \sigma \rangle = t^{-1} \langle \mathcal{W} \rangle = \text{Tr} \mathbf{Q} \mathbf{F}^T \mathbf{D}^{-1}. \quad (51)$$

It is expected to be positive from the fluctuation theorem, which implies that the second law of thermodynamics should hold for general NEQ phenomena with no thermodynamic origin.

The second cumulant of \mathcal{W} can also be found as

$$\begin{aligned}
 \langle \mathcal{W}^2 \rangle_c &= \langle \mathcal{W}^2 \rangle - \langle \mathcal{W} \rangle^2 \\
 &= \sum_{i,j} \langle (\mathbf{q}_i^T \cdot \bar{\mathbf{G}}_a \cdot \mathbf{q}_{i-1}) (\mathbf{q}_j^T \cdot \bar{\mathbf{G}}_a \cdot \mathbf{q}_{j-1}) \rangle \\
 &\quad - \left(\sum_i \langle \mathbf{q}_i^T \cdot \bar{\mathbf{G}}_a \cdot \mathbf{q}_{i-1} \rangle \right)^2 \\
 &= \sum_{i,j} \text{Tr} (-\Gamma_{ij} \bar{\mathbf{G}}_a \Gamma_{j-1,i-1} \bar{\mathbf{G}}_a + \Gamma_{ij-1} \bar{\mathbf{G}}_a \Gamma_{ji-1} \bar{\mathbf{G}}_a), \quad (52)
 \end{aligned}$$

where the summation $\sum'_{i,j} = 2 \sum_{i>j} + \sum_{i=j}$ due to the symmetry between i and j . Using Eq. (24), we can write

$$\begin{aligned}
 \langle \mathcal{W}^2 \rangle_c &= \sum_i \left[2 \sum_{j=1}^{i-1} \text{Tr} \{ -e^{-\mathbf{F}(t_i-t_j)} \mathbf{A}_j^{-1} \bar{\mathbf{G}}_a \mathbf{A}_{j-1}^{-1} e^{-\mathbf{F}^T(t_{i-1}-t_{j-1})} \bar{\mathbf{G}}_a \right. \\
 &\quad + e^{-\mathbf{F}(t_i-t_{j-1})} \mathbf{A}_{j-1}^{-1} \bar{\mathbf{G}}_a \mathbf{A}_j^{-1} e^{-\mathbf{F}^T(t_{i-1}-t_j)} \bar{\mathbf{G}}_a \} \\
 &\quad + \text{Tr} (-\mathbf{A}_i^{-1} \bar{\mathbf{G}}_a \mathbf{A}_{i-1}^{-1} \bar{\mathbf{G}}_a + \\
 &\quad \left. + e^{-\mathbf{F}\Delta t} \mathbf{A}_{i-1}^{-1} \bar{\mathbf{G}}_a e^{-\mathbf{F}\Delta t} \mathbf{A}_{i-1}^{-1} \bar{\mathbf{G}}_a) \right]. \quad (53)
 \end{aligned}$$

In $\Delta t \rightarrow 0$ limit, we get

$$\begin{aligned}
 \langle \mathcal{W}^2 \rangle_c &= -2 \text{Tr} \int_0^t d\tau e^{-\mathbf{F}^T \tau} (\mathbf{F}^T \bar{\mathbf{G}}_a - \bar{\mathbf{G}}_a \mathbf{F}) \mathbf{F} e^{-\mathbf{F} \tau} \\
 &\quad \times \int_0^\tau d\tau' e^{\mathbf{F} \tau'} \mathbf{A}^{-1}(\tau') \bar{\mathbf{G}}_a \mathbf{A}^{-1}(\tau') e^{\mathbf{F}^T \tau'} \\
 &\quad - 2 \text{Tr} \int_0^t d\tau e^{-\mathbf{F}^T \tau} (\mathbf{F}^T \bar{\mathbf{G}}_a - \bar{\mathbf{G}}_a \mathbf{F}) e^{-\mathbf{F} \tau} \\
 &\quad \times \int_0^\tau d\tau' e^{\mathbf{F} \tau'} \dot{\mathbf{A}}^{-1}(\tau') \bar{\mathbf{G}}_a \mathbf{A}^{-1}(\tau') e^{\mathbf{F}^T \tau'} \\
 &\quad - 2 \text{Tr} \int_0^t d\tau \bar{\mathbf{G}}_a \mathbf{F} \mathbf{A}^{-1}(\tau) \bar{\mathbf{G}}_a \mathbf{A}^{-1}(\tau) \\
 &\quad - \frac{1}{2} \text{Tr} [\bar{\mathbf{G}}_a \mathbf{A}^{-1}(t) \bar{\mathbf{G}}_a \mathbf{A}^{-1}(t) - \bar{\mathbf{G}}_a \mathbf{A}^{-1}(0) \bar{\mathbf{G}}_a \mathbf{A}^{-1}(0)], \quad (54)
 \end{aligned}$$

where $\dot{\mathbf{A}}^{-1}(\tau)$ is given by Eq. (22).

Using the identity of Eq. (A21) found in Appendix A, one can perform the above integral in principle. In this paper, rather than reporting the exact time dependence of Eq. (54), we compute the long-time behavior by keeping only the most dominant contributions,

$$\begin{aligned}
 \langle \mathcal{W}^2 \rangle_c &\rightarrow -t \text{Tr} (\mathbf{C} + \mathbf{E}) (\mathbf{F}^T \bar{\mathbf{G}}_a - \bar{\mathbf{G}}_a \mathbf{F}) \\
 &\quad - 2t \text{Tr} \bar{\mathbf{G}}_a \mathbf{F} \mathbf{U}^{-1} \bar{\mathbf{G}}_a \mathbf{U}^{-1} \\
 &= 2t \text{Tr} [\bar{\mathbf{G}}_a \mathbf{F} (\mathbf{E} - \mathbf{C})], \quad (55)
 \end{aligned}$$

where the matrix \mathbf{C} is antisymmetric, defined by

$$\mathbf{C} = \mathbf{U}^{-1} \bar{\mathbf{G}}_a \mathbf{U}^{-1}, \quad (56)$$

and the matrix \mathbf{E} is symmetric, determined by

$$\mathbf{F} \mathbf{E} + \mathbf{E} \mathbf{F}^T = \mathbf{F} \mathbf{C} - \mathbf{C} \mathbf{F}^T. \quad (57)$$

Note that $\langle (t^{-1} \mathcal{W})^2 \rangle_c \sim t^{-1}$. It implies that the PDF of the entropy production rate, $\sigma = t^{-1} \mathcal{W}$, shows a sharp distribution with the mean value $\langle \sigma \rangle$ found in Eq. (51) and the variance of order t^{-1} . Assuming it as Gaussian, $P(\sigma) \sim e^{-(\sigma - \langle \sigma \rangle)^2 / 2 \langle \sigma^2 \rangle_c}$, we obtain

$$\frac{P(\sigma)}{P(-\sigma)} = e^{ts\sigma}, \quad s = \frac{2\langle \sigma \rangle}{t \langle \sigma^2 \rangle_c}. \quad (58)$$

From Eq. (55), $\langle \sigma^2 \rangle_c \sim t^{-1}$. It qualitatively agrees with the fluctuation theorem for the entropy production. However, the fluctuation theorem predicts $s = 1$, while it seems not equal to 1 if estimated by assuming the Gaussian distribution. This implies that the PDF for the work and entropy production is in general non-Gaussian and non-Gaussian tails make a significant contribution to the exact theorem.

More information on $P(\mathcal{W})$ might come from higher cumulants in \mathcal{W} . In principle, it can be done systematically by using Eq. (25), but it is too complicated to proceed further calculation in detail.

VI. EXAMPLE: DIFFUSION IN TWO DIMENSIONS

Now, we take an example of a two dimensional diffusive motion, $\mathbf{q} = (x, y)^T$, for more explicit calculations. Consider

$$\mathbf{F} = \begin{pmatrix} k_1 & \kappa_1 \\ \kappa_2 & k_2 \end{pmatrix}, \quad \mathbf{D} = \begin{pmatrix} \alpha & \epsilon \\ \epsilon & \gamma \end{pmatrix}. \quad (59)$$

By the orthogonal coordinate transformation, \mathbf{D} can be diagonalized and the calculation goes simpler. However, we keep the present form of \mathbf{D} in order to examine the effect of noise correlations. If $\mathbf{F}\mathbf{D} - \mathbf{D}\mathbf{F}^T \neq \mathbf{0}$, we have a nonzero antisymmetric matrix \mathbf{Q} , a measure for NEQ, which can be obtained easily from Eq. (17),

$$\mathbf{Q} = \begin{pmatrix} 0 & \hat{q} \\ -\hat{q} & 0 \end{pmatrix}, \quad \hat{q} = \frac{\epsilon(k_1 - k_2) + \gamma\kappa_1 - \alpha\kappa_2}{k_1 + k_2}. \quad (60)$$

The system goes to equilibrium for $\hat{q} = 0$. The conventional Gibbs-Boltzmann (GB) type equilibrium is a trivial case where the force is conservative ($\kappa_1 = \kappa_2$), and the noises are identical and independent ($\alpha = \gamma$, $\epsilon = 0$). The equilibrium PDF is given as $e^{-\alpha^{-1}\mathcal{E}(x,y)}$, and $\mathcal{E}(x,y) = \frac{1}{2}(k_1x^2 + 2\kappa_1xy + k_2y^2)$. There are also nontrivial equilibria possible even for a nonconservative force ($\kappa_1 \neq \kappa_2$) and nonidentical or correlated noises ($\alpha \neq \gamma$, $\epsilon \neq 0$) as long as $\hat{q} = 0$. In this case, the equilibrium PDF is given as $e^{-(1/2)\mathbf{q}^T\mathbf{D}^{-1}\mathbf{F}\mathbf{q}}$. There seems to be no fundamental difference among many equilibria from the viewpoint of our paper in the sense that they all preserve the detailed balance. Now we consider the NESS for $\hat{q} \neq 0$. From Eq. (51), the entropy production rate in the long-time limit is given as

$$\langle \sigma \rangle = t^{-1} \langle \mathcal{W} \rangle = \frac{\text{Tr}\mathbf{F}}{\det \mathbf{D}} \hat{q}^2, \quad (61)$$

where $\det \mathbf{D} = \alpha\gamma - \epsilon^2$ and $\text{Tr}\mathbf{F} = k_1 + k_2$.

The second cumulant of the work can be obtained from Eq. (55). The matrices \mathbf{C} and \mathbf{E} are found from Eqs. (56) and (57) as

$$\mathbf{C} = \begin{pmatrix} 0 & c \\ -c & 0 \end{pmatrix}, \quad \mathbf{E} = \frac{c}{\text{Tr}\mathbf{F}} \begin{pmatrix} -2\kappa_1 & k_1 - k_2 \\ k_1 - k_2 & 2\kappa_2 \end{pmatrix}, \quad (62)$$

with $c = \frac{\det(\mathbf{D}+\mathbf{Q})\text{Tr}\mathbf{F}}{2\det\mathbf{D}\det\mathbf{F}}\hat{q}$. Then, $\langle \sigma^2 \rangle$ is found as

$$\begin{aligned} \langle \sigma^2 \rangle_c &= t^{-2} \langle \mathcal{W}^2 \rangle_c \\ &= 2t^{-1} \frac{(\det \mathbf{D} + \hat{q}^2)\text{Tr}\mathbf{F}}{(\det \mathbf{D})^2} \hat{q}^2. \end{aligned} \quad (63)$$

For the stability of the NESS, we assumed at the beginning that the matrices \mathbf{D} and \mathbf{F} are positive definite. Thus $\det \mathbf{D} > 0$ and $\text{Tr}\mathbf{F} > 0$, which guarantee the positivity of $\langle \mathcal{W} \rangle_c$ and $\langle \mathcal{W}^2 \rangle_c$. From Eq. (58), it can be shown that $s = \det \mathbf{D} / (\det \mathbf{D} + \hat{q}^2) < 1$, which indicates that $P(\sigma)$ is more distributed than the Gaussian distribution for nonzero \hat{q} . We observe that all higher-order cumulants are also of order t , i.e., $\langle \mathcal{W}^n \rangle_c \sim t$, so $\langle \sigma^n \rangle_c \sim t^{-(n-1)}$.

We have performed a numerical analysis to confirm our analytic results and gain more insights on the work distribution function $P(\mathcal{W})$. Here we present numerical data taken at $(k_1, k_2) = (4, 1)$, $(\kappa_1, \kappa_2) = (2, 1)$, $(\alpha, \gamma) = (1, 1)$, and $\epsilon = \sin \theta$ with $\theta = 0.1$. This system has a NESS with a nonzero value

of \hat{q} . We expect that the results do not depend on a specific choice of parameter values as far as \hat{q} is nonzero.

For \mathbf{G}_s and \mathbf{G}_a , we adopt the AS choice given in Eq. (31) for convenience. Hence the system is assumed to have an initial probability distribution,

$$P[\mathbf{q}(0)] = |\det[2\pi\mathbf{A}^{-1}(0)]|^{-1/2} e^{-(1/2)\mathbf{q}^T(0)\mathbf{A}(0)\mathbf{q}(0)}$$

with $\mathbf{A}(0) = \tilde{\mathbf{G}}_s$ and the work for a path $\mathbf{q}(\tau)$ is obtained using Eq. (35).

The generating function $\mathcal{G}(\lambda)$ defined in Eq. (37) can be estimated by direct numerical integrations of the Langevin equation in Eq. (1). One starts from an initial state $\mathbf{q}(0)$ drawn from the initial distribution $P[\mathbf{q}(0)]$. The Langevin equation is then integrated with discretized time intervals

$$\mathbf{q}(\tau + \Delta t) = \mathbf{q}(\tau) - \mathbf{F} \cdot \mathbf{q}(\tau)\Delta t + \Delta \mathbf{W}(\tau),$$

where $\Delta \mathbf{W}(\tau) = (\Delta W_x(\tau), \Delta W_y(\tau))^T$ are correlated random variables constructed as

$$\begin{pmatrix} \Delta W_x(\tau) \\ \Delta W_y(\tau) \end{pmatrix} = \sqrt{2\Delta t} \begin{pmatrix} 1 & 0 \\ \sin \theta & \cos \theta \end{pmatrix} \begin{pmatrix} \eta_x(\tau) \\ \eta_y(\tau) \end{pmatrix}.$$

Here, $\eta_{x,y}(\tau)$ are independent and identically distributed Gaussian random variables with zero mean and unit variance. One can check easily that such random variables $\Delta \mathbf{W}(\tau)$ satisfy the required correlation property $\langle \Delta \mathbf{W}(\tau)\Delta \mathbf{W}(\tau')^T \rangle = 2(\Delta t)\mathbf{D}\delta(\tau - \tau')$.

The work production is estimated as

$$\mathcal{W}(\tau + \Delta t) = \mathcal{W}(\tau) - [\mathbf{q}(\tau + \Delta t) - \mathbf{q}(\tau)]^T \cdot \tilde{\mathbf{G}}_a \cdot \mathbf{q}(\tau).$$

Repeating N_S independent simulations, one obtains a numerical estimate

$$\mathcal{G}_N(t; \lambda) = \frac{1}{N_S} \sum_{n=1}^{N_S} e^{-\lambda \mathcal{W}_n(t)}. \quad (64)$$

We can also utilize our analytic expression for \mathcal{G} in Eq. (47) in order to get a more precise numerical estimate. We first solve the NLDE for $\tilde{\mathbf{A}}(t; \lambda)$ in Eq. (45) with the initial condition $\tilde{\mathbf{A}}(0) = \tilde{\mathbf{G}}_s$. The solution is obtained numerically in discretized times using the recursion relation $\tilde{\mathbf{A}}(\tau + \Delta t) = \tilde{\mathbf{A}}(\tau) + \Delta t \frac{d\tilde{\mathbf{A}}(\tau)}{d\tau}$. The generating function \mathcal{G} can be evaluated easily using the numerical solution $\tilde{\mathbf{A}}$. The generating function evaluated numerically using the analytic expression will be denoted as \mathcal{G}_A .

In Fig. 1, we compare the results $\mathcal{G}_N(t; \lambda)$ and $\mathcal{G}_A(t; \lambda)$ at $t = 0.5$ and 2.0 obtained with $\Delta t = 0.0001$ and $N_S \leq 10^7$. The two methods yield almost identical results for small values of $|\lambda|$. Both data confirm the Jarzynski equality $\mathcal{G}(\lambda = 1) = 1$ and the Crooks fluctuation theorem $\mathcal{G}(\lambda) = \mathcal{G}(1 - \lambda)$. However, there is a noticeable discrepancy at larger values of $|\lambda|$. Even the Crooks fluctuation theorem seems to be violated in the \mathcal{G}_N data. One might suspect a finite Δt as a source of systematic errors. We have also taken data with $\Delta t = 0.01$, 0.001 , and 0.0001 and found no significant difference, which means that $\Delta t = 0.0001$ is already small enough. In fact, the discrepancy is due to limited sampling in obtaining \mathcal{G}_N . When $|\lambda|$ is large, $\mathcal{G}(\lambda)$ is dominated by rare events with large $|\mathcal{W}|$. If one compares \mathcal{G}_N obtained from $N_S = 10^5$, 10^6 , and 10^7 samples, there are strong fluctuations at large values of $|\lambda|$. This means that the

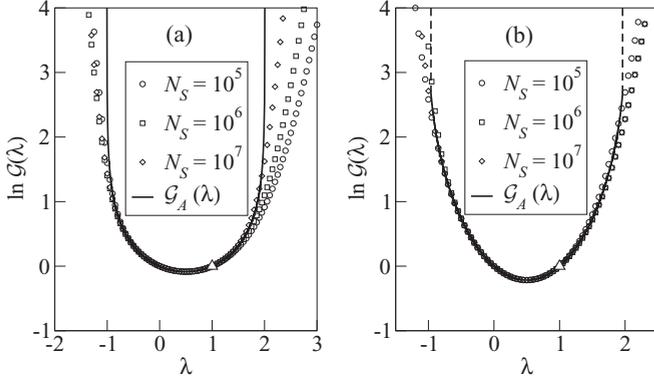


FIG. 1. Comparison of $\mathcal{G}_N(\lambda)$ and $\mathcal{G}_A(\lambda)$ at $t = 0.5$ in (a) and $t = 2.0$ in (b). Symbols represent \mathcal{G}_N and lines represent \mathcal{G}_A . Location of the symbol Δ represents the Jarzynski equality $\mathcal{G}(1) = 1$. In (a), \mathcal{G}_A diverges continuously as λ approaches a threshold. On the other hand, in (b), \mathcal{G}_A remains finite up to a threshold and diverges discontinuously beyond it.

tail property of the work distribution function $P(\mathcal{W})$ cannot be accessed from numerical simulations even with 10^7 samples. On the contrary, the analytic formalism allows us to study the work distribution function in detail without any statistics problem.

Figure 1 shows that $\mathcal{G}(t; \lambda)$ becomes singular at $\lambda = \lambda_0$ and $1 - \lambda_0$ with a t -dependent threshold $\lambda_0 = \lambda_0(t) > 1$. The singular behavior is evident in Fig. 2(a), where we plot $\ln \mathcal{G}(t; \lambda)$ as a function of t at several values of $\lambda \geq 1/2$. It suffices to consider $\lambda \geq 1/2$ because of the symmetry $\mathcal{G}(\lambda) = \mathcal{G}(1 - \lambda)$. When $\lambda < \lambda_c \simeq 1.962(1)$, $\mathcal{G}(t; \lambda)$ remains finite for all t . On the other hand, it diverges at $t = t_c \simeq 0.86(1)$ at $\lambda = \lambda_c$, and diverges at $t < t_c$ when $\lambda > \lambda_c$. From these plots, we conclude that $\mathcal{G}(t; \lambda)$, being viewed as a function of λ , diverges at t -dependent thresholds $\lambda = \lambda_0(t)$ and $\lambda = 1 - \lambda_0(t)$. The threshold, numerically determined, is drawn in Fig. 2(b). Figure 2 also allows us to conclude that $\mathcal{G}(t; \lambda)$ diverges continuously as $\lambda \rightarrow \lambda_0(t)$ [see the solid line in Fig. 2(b)] when $t \leq t_c$. On the other hand, when $t > t_c$, $\mathcal{G}(t; \lambda)$ displays a discontinuous jump to infinity at $\lambda = \lambda_c$ [see the dashed line in Fig. 2(b)]. Interestingly, Fig. 2(b) resembles a phase diagram of a system having a tricritical point where

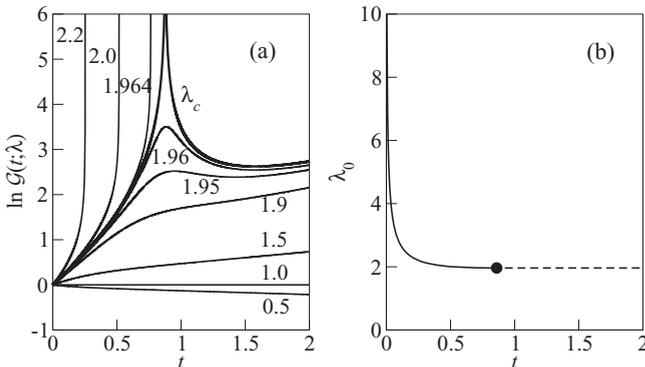


FIG. 2. (a) Plot of $\ln \mathcal{G}(t; \lambda)$ against t at several values of λ . (b) Threshold curve $\lambda_0(t)$ above which $\mathcal{G}(t; \lambda)$ is infinite. The symbol represents a point $(t_c, \lambda_c) \simeq (0.86, 1.962)$.

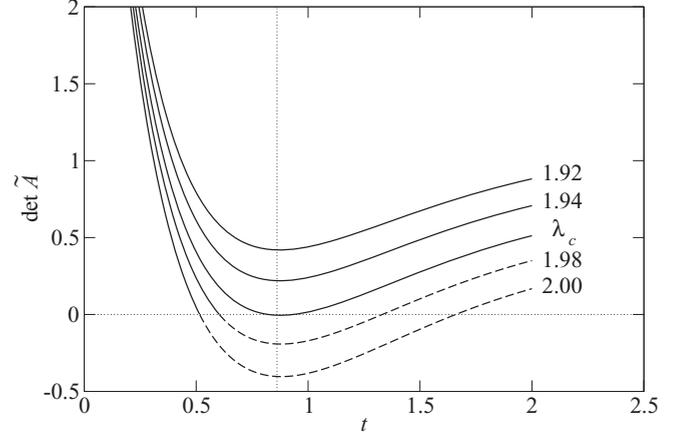


FIG. 3. Time evolution of $\det \tilde{\mathbf{A}}(t; \lambda)$ near $t = t_c$ and $\lambda = \lambda_c$. When $\lambda < \lambda_c \simeq 1.962$, the determinant seems to be always positive for any t . In contrast, it becomes negative at finite t for $\lambda > \lambda_c$ and the time evolution afterward (the dashed line) is meaningless. When $\lambda = \lambda_c$, it is tangential to the x axis (the horizontal dotted line) at $t = t_c$ (the vertical dotted line).

a continuous phase transition line turns into a discontinuous phase transition line.

Origin and nature of the divergence are understood from the analytic expression for $\ln \mathcal{G}$ in Eq. (47). Due to the logarithmic boundary term, $\mathcal{G}(t; \lambda)$ is well defined only when $\det[\tilde{\mathbf{A}}(t'; \lambda) + \lambda \delta \mathbf{G}_s]$ is positive for all $t' < t$. In contrast, there is no singularity in the bulk term for any t . From the numerical solution of the NLDE, Eq. (45), we observed that $\det[\tilde{\mathbf{A}}(t; \lambda)]$ ($\delta \mathbf{G}_s = \mathbf{0}$ with the AS choice) behaves as

$$\det[\tilde{\mathbf{A}}(t; \lambda)] \simeq a(\lambda_c - \lambda) + b(t - t_c)^2 \quad (65)$$

near $\lambda = \lambda_c$ and $t = t_c$ with positive constants a and b ; see Fig. 3. This behavior explains the singularity in \mathcal{G} .

When $t \leq t_c$, the determinant becomes zero at $\lambda = \lambda_0$ with

$$\lambda_0 \simeq \lambda_c + \frac{b}{a}(t - t_c)^2. \quad (66)$$

Consequently, \mathcal{G} diverges *continuously* as

$$\mathcal{G}(t; \lambda) \sim (\lambda_0 - \lambda)^{-1/2} \quad (67)$$

as λ approaches λ_0 from below.

The determinant is always positive when $\lambda < \lambda_c$, while it becomes negative at $t < t_c$ when $\lambda > \lambda_c$. Hence when $t > t_c$, $\mathcal{G}(t; \lambda)$ remains finite up to $\lambda < \lambda_c$ and diverges *discontinuously* at $\lambda = \lambda_c$. As λ approaches λ_c from below, it behaves regularly as

$$\mathcal{G}(t; \lambda) \sim h + a(\lambda_c - \lambda)^1 \quad (68)$$

with the t -dependent constant h .

The singularities in $\mathcal{G}(\lambda) = \int d\mathcal{W} P(\mathcal{W}) e^{-\lambda \mathcal{W}}$ at $\lambda = \lambda_0$ and $1 - \lambda_0$ indicate that the PDF $P(\mathcal{W})$ has exponential tails,

$$P(\mathcal{W}) \sim \begin{cases} \mathcal{W}^{-r} e^{-\mathcal{W}/\mathcal{W}_+} & \text{for } \mathcal{W} \rightarrow \infty, \\ (-\mathcal{W})^{-r} e^{-\mathcal{W}/\mathcal{W}_-} & \text{for } \mathcal{W} \rightarrow -\infty, \end{cases} \quad (69)$$

with characteristic works \mathcal{W}_+ (> 0) and \mathcal{W}_- (< 0), and possible power-law corrections with exponent r . The power-law

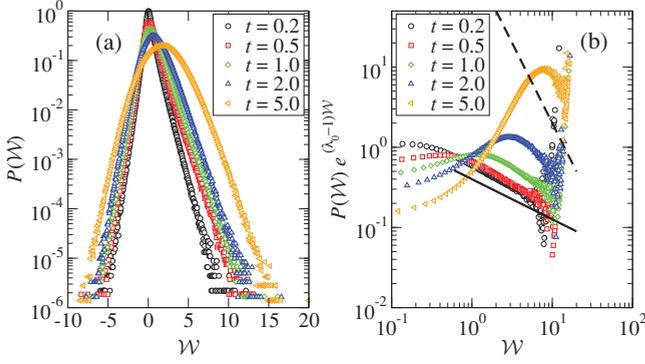


FIG. 4. (Color online) (a) Plot of $P(W)$ versus W at several values of t . (b) Plot of $P(W)e^{(\lambda_0-1)W}$ versus W for $W > 0$. The solid and dashed lines are guides to eyes with slope $-1/2$ and -2 , respectively.

prefactor is necessary in order to account for the way \mathcal{G} becomes singular at $\lambda = \lambda_0$. From the Crooks fluctuation theorem, $P(W) = e^{W\lambda} P(-W)$, the exponent r should be the same for both tails, and it suffices to consider one of the tails.

It is easy to check that the negative tail yields $\mathcal{G}(\lambda) \sim (1/|\mathcal{W}_-| - \lambda)^{r-1}$. Comparing it with Eqs. (67) and (68), we find that the characteristic work is given by $\mathcal{W}_- = -1/\lambda_0$ and that the exponent is given by

$$r = \begin{cases} \frac{1}{2} & \text{for } t \leq t_c, \\ 2 & \text{for } t > t_c. \end{cases} \quad (70)$$

The positive tail has $\mathcal{W}_+ = 1/(\lambda_0 - 1)$ and the same exponent r .

Numerical data are consistent with the tail property in Eq. (69). The PDF $P(W)$ was obtained from numerical simulations of $N_S = 10^7$ samples. Figure 4(a) presents the plot of $P(W)$ against W at several values of t in the semilog scale. As expected, $P(W)$ becomes more distributed and the mean work production $\langle W \rangle$ increases with time t . Moreover, the exponential tails are clearly seen in all plots with characteristic works [slopes of plots in Fig. 4(a)] saturating with time t .

We also present the log-log plot of $P(W)e^{(\lambda_0-1)W}$ against W for $W > 0$ in Fig. 4(b). We use the threshold value $\lambda_0(t)$ obtained from the singularities in \mathcal{G}_A , which are shown in Fig. 2(b). One can see that the $P(W)e^{(\lambda_0-1)W}$ has a power-law tail as predicted in Eq. (69). When $t = 0.2$ and $0.5 (< t_c)$, the power-law tail is manifest and the exponent is in good agreement with the analytic prediction $r = 1/2$. When $t = 2.0$ and $5.0 (> t_c)$, the power law sets in at larger values of W and the exponent value is drifting with increasing W . In this case, we need many more samples ($N_S \gg 10^7$) to get good statistics for rare events at large W , in order to extract reliable quantitative information on the power-law tail. Nevertheless, one can see that the exponent value becomes close to $r = 2$ for large t . At $t = 1.0$, the crossover effect dominates the numerical data, since t is too close to $t_c \simeq 0.86$.

It is not surprising to see the exponential tail with a power-law prefactor with the exponent $r = 1/2$. This exponential tail has also been observed in other NEQ systems [34–37]. When the PDF is given as $P(\mathbf{q}) \sim e^{-\Phi(\mathbf{q})}$ with the energylike function $\Phi(\mathbf{q})$ quadratic in \mathbf{q} as in our case, the PDF for any quantity

\mathcal{A} also quadratic in \mathbf{q} should become $P(\mathcal{A}) \sim \mathcal{A}^{-1/2} e^{-\mathcal{A}/\mathcal{A}_c}$ with the characteristic value of \mathcal{A}_c . As the work \mathcal{W} is simply quadratic in \mathbf{q} and proportional to t , at least for a short time t , i.e., $\mathcal{W} \sim tq^2$ as in our case, our finding in Eqs. (69) and (70) can be understood with \mathcal{A}_c increasing in time t .

However, there is a sharp dynamic phase transition at $t = t_c$ beyond which \mathcal{A}_c is constant of time and the power-law exponent changes from $1/2$ to 2 . This implies that the characteristic positive work production $\mathcal{W}_+ = 1/[\lambda_0(t) - 1]$ increases monotonically in time for $t < t_c$, but saturates to a finite value $1/(\lambda_c - 1)$ at $t = t_c$. For $t > t_c$, the characteristic work production remains unchanged and only the power-law prefactor adjusts the PDF accordingly. We do not have an intuitive understanding for the dynamic phase transition at this moment, except that the transition time t_c may be related to the intrinsic relaxation time of the system. Furthermore, t_c depends on the choice of the NEQ force, i.e., $\delta\mathcal{G}_s$. It calls for a further study to understand this phase transition, which is currently under investigation.

VII. SUMMARY AND DISCUSSION

We have studied NEQ fluctuations for high-dimensional diffusion dynamics driven by a linear drift force. The drift force is not derivable from a scalar potential function and the noises are not identical white noises in components with possible correlations. In general, these NEQ features generate a circulating probability current at the steady state, with the drift force inward to the origin (the force matrix \mathbf{F} is positive definite). It is interesting to notice that the equilibrium can be restored with a specific combination of these two NEQ features by satisfying the detailed balance condition ($\mathbf{D}\mathbf{F} = \mathbf{F}^T\mathbf{D}$).

Recent experiments on an optical trap report that particles can be confined in a field-induced potential well, approximated by an asymmetric harmonic potential. It may be possible to apply our study to such experiments. However, there are some technical issues to be resolved in experiments. In particular, in order to break the detailed balance, the noises should be applied in such a way that their principal axes do not coincide with those of the potential well [25].

In this paper, we started with the Langevin equation with an arbitrary drift force and arbitrary additive noises in high dimensions without any thermodynamic origin. The generalized thermodynamic quantities like energy, work, and heat are defined, with which the Crooks fluctuation theorems are reproduced. For the case of the linear drift force, we derive the exact evolution function of the PDF analytically. More importantly, we analyzed the work PDF, $P(W)$ through the generating function method, which showed an interesting dynamic phase transition in the exponential tail shape of $P(W)$. As the tail property is governed by rare events, it is crucial to develop an analytic theory to show this subtle dynamic phase transition, which can be hardly identified by numerical simulations only. Even though we expect that this phase transition is one of the generic features found in the NEQ systems described by Langevin equations, it would be a big challenge to understand analytically how and when this can arise.

Finally, in the case of nonlinear forces, it has been found that there is an additional current which shifts the probability

maximum from the fixed point at the origin [26]. It may have the same origin as the noise-driven directed current in extended systems [27,28]. The ingredients for those currents are (a) nonlinearity of the inward drift force and high-dimensional noises for the confined system, (b) asymmetric (ratchet-type) potential and an additional noise for the extended system. The NEQ fluctuation theory for this nonlinear diffusion dynamics will be another challenging topic.

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APPENDIX A: TIME-DEPENDENT PDF

We keep the source field for later use to compute the generating functional. The initial probability distribution is chosen as

$$P[\mathbf{q}(0)] = \mathcal{N}_0 e^{-(1/2)\mathbf{q}^T(0)\cdot\mathbf{A}(0)\cdot\mathbf{q}(0)}, \quad (\text{A1})$$

with $\mathcal{N}_0 = |\det[2\pi\mathbf{A}^{-1}(0)]|^{-1/2}$ and $\mathbf{A}^T(0) = \mathbf{A}(0)$. We consider the sequence of discrete times, $0 = t_0 < t_1 < t_2 < \dots < t_N = t$ with time interval $\Delta t = t/N$ in the $N \rightarrow \infty$ limit. Denoting $\mathbf{q}_i \equiv \mathbf{q}(t_i)$ with $\mathbf{q}_0 = \mathbf{q}(0)$ and $\mathbf{q}_N = \mathbf{q}(t) = \mathbf{q}$, and $\mathbf{A}_0 = \mathbf{A}(0)$, we write $P(\mathbf{q}, t; \mathbf{l})$ in the prepoint representation [set $\mathbf{q}(\tau) = \mathbf{q}_{i-1}$ for $t_{i-1} \leq \tau \leq t_i$] as

$$\begin{aligned} P(\mathbf{q}, t; \mathbf{l}) &= \mathcal{N}_0 \int \prod_{i=0}^{N-1} \frac{d\mathbf{q}_i}{|\det(4\pi\Delta t\mathbf{D})|^{1/2}} \\ &\times e^{-(1/4\Delta t)\sum_{i=1}^N (\mathbf{q}_i - \mathbf{q}_{i-1} + \Delta t\mathbf{F}\cdot\mathbf{q}_{i-1})^T \cdot \mathbf{D}^{-1} \cdot (\mathbf{q}_i - \mathbf{q}_{i-1} + \Delta t\mathbf{F}\cdot\mathbf{q}_{i-1})} \\ &\times e^{\Delta t \sum_{i=0}^{N-1} \mathbf{l}_i^T \cdot \mathbf{q}_i - (1/2)\mathbf{q}_0^T \cdot \mathbf{A}_0 \cdot \mathbf{q}_0} \\ &= \mathcal{N}_0 \int \prod_{i=0}^{N-1} \frac{d\mathbf{q}_i}{|\det(4\pi\Delta t\mathbf{D})|^{1/2}} \left[\prod_{i=1}^N T_{i,i-1} \right] e^{-(1/2)\mathbf{q}_0^T \cdot \mathbf{A}_0 \cdot \mathbf{q}_0}. \end{aligned} \quad (\text{A2})$$

The transfer matrix $T_{i,i-1}$ is given as

$$\begin{aligned} \ln T_{i,i-1}(\mathbf{l}_{i-1}) &= -\frac{1}{4\Delta t} [(\mathbf{q}_i - \mathbf{V}\mathbf{q}_{i-1})^T \cdot \mathbf{D}^{-1} \cdot (\mathbf{q}_i - \mathbf{V}\mathbf{q}_{i-1}) \\ &+ (\Delta t)\mathbf{l}_{i-1}^T \cdot \mathbf{q}_{i-1}], \end{aligned} \quad (\text{A3})$$

where

$$\mathbf{V} = 1 - \Delta t\mathbf{F}. \quad (\text{A4})$$

Integrating over \mathbf{q}_0 , we obtain

$$\begin{aligned} I_0 &= \int \frac{d\mathbf{q}_0}{|\det(4\pi\Delta t\mathbf{D})|^{1/2}} T_{1,0} e^{-(1/2)\mathbf{q}_0^T \cdot \mathbf{A}_0 \cdot \mathbf{q}_0} \\ &= |\det(\mathbf{D}\mathbf{B}_0)|^{-1/2} e^{-(1/2)\mathbf{q}_1^T \cdot \mathbf{A}_1 \cdot \mathbf{q}_1} \\ &\times e^{(\Delta t)^3 \mathbf{l}_0^T \cdot \mathbf{B}_0^{-1} \cdot \mathbf{l}_0 + (\Delta t)\mathbf{l}_0^T \cdot \mathbf{B}_0^{-1} \mathbf{V}^T \mathbf{D}^{-1} \cdot \mathbf{q}_1}, \end{aligned} \quad (\text{A5})$$

where

$$\mathbf{A}_1 = \frac{1}{2\Delta t} (\mathbf{D}^{-1} - \mathbf{D}^{-1}\mathbf{V}\mathbf{B}_0^{-1}\mathbf{V}^T\mathbf{D}^{-1}) \quad (\text{A6})$$

$$\mathbf{B}_0 = \mathbf{V}^T\mathbf{D}^{-1}\mathbf{V} + 2\Delta t\mathbf{A}_0, \quad (\text{A7})$$

where \mathbf{A}_1 and \mathbf{B}_0 are both symmetric. Then we can find

$$\begin{aligned} T_{2,1}(\mathbf{l}_1)I_0 &= T_{2,1}(\tilde{\mathbf{l}}_1) e^{-(1/2)\mathbf{q}_1^T \cdot \mathbf{A}_1 \cdot \mathbf{q}_1} \\ &\times |\det(\mathbf{D}\mathbf{B}_0)|^{-1/2} e^{(\Delta t)^3 \mathbf{l}_0^T \cdot \mathbf{B}_0^{-1} \cdot \mathbf{l}_0}, \end{aligned} \quad (\text{A8})$$

where $\tilde{\mathbf{l}}_1 = \mathbf{l}_1 + \mathbf{D}^{-1}\mathbf{V}\mathbf{B}_0^{-1} \cdot \mathbf{l}_0$. Notice that the integration of $T_{1,0}$ over \mathbf{q}_0 results in a change $\tilde{\mathbf{l}}_1$ in the source field of $T_{2,1}$. It happens iteratively for subsequent integrations over the rest of \mathbf{q}_i . We show it explicitly for the next step,

$$\begin{aligned} I_1 &= \int \frac{d\mathbf{q}_1}{|\det(4\pi\Delta t\mathbf{D})|^{1/2}} T_{2,1}(\tilde{\mathbf{l}}_1) e^{-(1/2)\mathbf{q}_1^T \cdot \mathbf{A}_1 \cdot \mathbf{q}_1} \\ &= |\det(\mathbf{D}\mathbf{B}_1)|^{-1/2} e^{-(1/2)\mathbf{q}_2^T \cdot \mathbf{A}_2 \cdot \mathbf{q}_2} \\ &\times e^{(\Delta t)^3 \mathbf{l}_1^T \cdot \mathbf{B}_1^{-1} \cdot \tilde{\mathbf{l}}_1 + (\Delta t)\mathbf{l}_1^T \cdot \mathbf{B}_1^{-1} \mathbf{V}^T \mathbf{D}^{-1} \cdot \mathbf{q}_1}, \end{aligned} \quad (\text{A9})$$

where

$$\mathbf{A}_2 = \frac{1}{2\Delta t} (\mathbf{D}^{-1} - \mathbf{D}^{-1}\mathbf{V}\mathbf{B}_1^{-1}\mathbf{V}^T\mathbf{D}^{-1}) \quad (\text{A10})$$

$$\mathbf{B}_1 = \mathbf{V}^T\mathbf{D}^{-1}\mathbf{V} + 2\Delta t\mathbf{A}_1. \quad (\text{A11})$$

Then we get

$$\begin{aligned} T_{3,2}(\mathbf{l}_2)I_1 &= T_{3,2}(\tilde{\mathbf{l}}_2) e^{-(1/2)\mathbf{q}_2^T \cdot \mathbf{A}_2 \cdot \mathbf{q}_2} \\ &\times |\det(\mathbf{D}\mathbf{B}_1)|^{-1/2} e^{(\Delta t)^3 \mathbf{l}_1^T \cdot \mathbf{B}_1^{-1} \cdot \tilde{\mathbf{l}}_1}, \end{aligned} \quad (\text{A12})$$

where $\tilde{\mathbf{l}}_2 = \mathbf{l}_2 + \mathbf{D}^{-1}\mathbf{V}\mathbf{B}_1^{-1} \cdot \tilde{\mathbf{l}}_1$. We repeat the integrations over all \mathbf{q}_i up to $i = N - 1$. Finally, we reach the result

$$\begin{aligned} P(\mathbf{q}_N, t_N; \mathbf{l}) &= |\det(2\pi\mathbf{A}_0^{-1})|^{-1/2} \prod_{i=0}^{N-1} |\det(\mathbf{D}\mathbf{B}_i)|^{-1/2} \\ &\times e^{-(1/2)\mathbf{q}_N^T \cdot \mathbf{A}_N \cdot \mathbf{q}_N + (\Delta t)\mathbf{l}_N^T \cdot \mathbf{q}_N + (\Delta t)^3 \sum_{i=0}^{N-1} \mathbf{l}_i^T \cdot \mathbf{B}_i^{-1} \cdot \tilde{\mathbf{l}}_i}. \end{aligned} \quad (\text{A13})$$

We have the recursion relations

$$\mathbf{A}_i = \frac{1}{2\Delta t} (\mathbf{D}^{-1} - \mathbf{D}^{-1}\mathbf{V}\mathbf{B}_{i-1}^{-1}\mathbf{V}^T\mathbf{D}^{-1}) \quad (\text{A14})$$

$$\mathbf{B}_{i-1} = \mathbf{V}^T\mathbf{D}^{-1}\mathbf{V} + 2\Delta t\mathbf{A}_{i-1} \quad (\text{A15})$$

$$\tilde{\mathbf{l}}_i = \mathbf{l}_i + \mathbf{D}^{-1}\mathbf{V}\mathbf{B}_{i-1}^{-1} \cdot \tilde{\mathbf{l}}_{i-1}, \quad (\text{A16})$$

for $i = 1, \dots, N$ with $\tilde{\mathbf{l}}_0 = \mathbf{l}_0$.

A_i determines the intermediate probability distribution function at time t_i . We can find a simple recursion relation for A_i^{-1} . First we observe the following relation:

$$\begin{aligned} A_i &= \frac{1}{2\Delta t} \left\{ D^{-1} - (D + 2\Delta t D(V^T)^{-1} A_{i-1} V^{-1} D)^{-1} \right\} \\ &= (V^T)^{-1} A_{i-1} V^{-1} D (D + 2\Delta t D(V^T)^{-1} A_{i-1} V^{-1} D)^{-1} \\ &= (2\Delta t D + V A_{i-1}^{-1} V^T)^{-1}. \end{aligned} \quad (\text{A17})$$

Therefore we get

$$A_i^{-1} = 2\Delta t D + V A_{i-1}^{-1} V^T, \quad (\text{A18})$$

which leads to

$$\begin{aligned} A_i^{-1} &= 2\Delta t D + 2\Delta t V D V^T + V^2 A_{i-2}^{-1} (V^T)^2 \\ &= 2\Delta t \sum_{j=0}^{i-1} V^j D (V^T)^j + V^i A_0^{-1} (V^T)^i. \end{aligned} \quad (\text{A19})$$

In the continuum limit, we have

$$A^{-1}(t) = 2 \int_0^t d\tau e^{-F\tau} D e^{-F\tau} + e^{-Ft} A^{-1}(0) e^{-Ft}. \quad (\text{A20})$$

We use the integral identity as

$$\begin{aligned} &\int_0^t d\tau e^{-H\tau} C e^{-H\tau} \\ &= \frac{1}{2} H^{-1} [(C + E) - e^{-Ht} (C + E) e^{-Ht}], \end{aligned} \quad (\text{A21})$$

for arbitrary matrices C and H where E is determined from the equation

$$HE + EH^T = HC - CH^T. \quad (\text{A22})$$

Note that E is antisymmetric (symmetric) if C is symmetric (antisymmetric).

Here, with $C = D$ and $H = F$, the antisymmetric matrix E becomes equal to Q from Eq. (17). Then $H^{-1}(C + E) = F^{-1}(D + Q) = U^{-1}$ from Eq. (16). Therefore we have

$$A^{-1}(t) = U^{-1} + e^{-Ft} [A^{-1}(0) - U^{-1}] e^{-Ft}. \quad (\text{A23})$$

Using Eq. (A18), the recursion relation for B_{i-1} , Eq. (A15), becomes

$$\begin{aligned} B_{i-1} &= A_{i-1} V^{-1} (V A_{i-1}^{-1} V^T + 2\Delta t D) D^{-1} V \\ &= A_{i-1} V^{-1} A_i^{-1} D^{-1} V. \end{aligned} \quad (\text{A24})$$

Therefore we get

$$\det(DB_{i-1}) = \frac{\det A_{i-1}}{\det A_i}, \quad (\text{A25})$$

which simplifies the normalization factor in Eq. (A13) as

$$\begin{aligned} &|\det(2\pi A_0^{-1})|^{-1/2} \prod_{i=0}^{N-1} |\det(DB_i)|^{-1/2} \\ &= \left| \det(2\pi A_0^{-1}) \frac{\det A_0}{\det A_1} \frac{\det A_1}{\det A_2} \cdots \frac{\det A_{N-1}}{\det A_N} \right|^{-1/2} \\ &= |\det(2\pi A_N^{-1})|^{-1/2}. \end{aligned} \quad (\text{A26})$$

It is the correct normalization factor for the final time-dependent probability distribution, $P(\mathbf{q}, t) = P(\mathbf{q}, t; \mathbf{l} = 0)$:

$$P(\mathbf{q}, t) = |\det[2\pi A^{-1}(t)]|^{-1/2} e^{-(1/2)\mathbf{q}^T A(t) \mathbf{q}}. \quad (\text{A27})$$

APPENDIX B: GENERATING FUNCTIONAL

The generating functional $Z[\mathbf{l}] = \int d\mathbf{q} P(\mathbf{q}, t; \mathbf{l})$ is obtained by integrating out Eq. (A13) over \mathbf{q}_N :

$$\begin{aligned} Z[\mathbf{l}] &= e^{(\Delta t)^3 \sum_{i=0}^N \bar{l}_i^T \cdot B_i^{-1} \cdot \bar{l}_i} \\ &\equiv e^{(1/2)(\Delta t)^2 \sum_{i,j} \bar{l}_i^T \cdot \Gamma_{ij} \cdot \bar{l}_j}, \end{aligned} \quad (\text{B1})$$

where $B_N \equiv 2(\Delta t)A_N$. Investigating the recursion relation for \bar{l}_i , Eq. (A16), we first show that for $i > j$

$$\Gamma_{ii} = 2\Delta t B_i^{-1} + C_i^T \Gamma_{i+1, i+1} C_i \quad (\text{B2})$$

$$\Gamma_{ij} = \Gamma_{ii} C_{i-1} C_{i-2} \cdots C_j, \quad (\text{B3})$$

where

$$C_i = D^{-1} V B_i^{-1} = A_{i+1} V A_i^{-1}. \quad (\text{B4})$$

Using $\Gamma_{NN} = A_N^{-1}$ and Eq. (A24),

$$\begin{aligned} \Gamma_{N-1, N-1} &= 2\Delta t V^{-1} D A_N V A_{N-1}^{-1} + C_{N-1}^T A_N^{-1} C_{N-1} \\ &= 2\Delta t V^{-1} D A_N V A_{N-1}^{-1} + A_{N-1}^{-1} V^T A_N V A_{N-1}^{-1} \\ &= V^{-1} (2\Delta t D + V A_{N-1}^{-1} V^T) A_N V A_{N-1}^{-1} \\ &= A_{N-1}^{-1}. \end{aligned} \quad (\text{B5})$$

We can prove by induction that

$$\Gamma_{ii} = A_i^{-1}. \quad (\text{B6})$$

By noting

$$C_{i-1} C_{i-2} \cdots C_j = A_i V^{i-j} A_j^{-1},$$

we find

$$\Gamma_{ij} = V^{i-j} A_j^{-1}. \quad (\text{B7})$$

Note that for $j > i$, $\Gamma_{ij} = \Gamma_{ji}^T$. In the continuum limit, we finally get Eq. (24).

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