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Decomposition of force fluctuations far from equilibrium

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By studying a nonequilibrium Langevin system, we find that a simple condition determines the decomposition of the coarse-grained force into a dissipative force, an effective driving force and noise. From this condition, we derive a universal inequality, $D \geqslant \gamma\mu_{\rm d}^2 T$, relating the diffusion constant D, the differential mobility $\mu_{\rm d}$, the bare friction constant γ and the temperature T. Due to the general nature of the argument we present, we believe that our idea concerning this decomposition can be applied to a wide class of systems far from equilibrium.

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The nature of a force depends on the scale on which it is observed. For example, consider a force exerted by water molecules on a colloidal particle. Such a force can be described by mechanical laws on molecular time scales, while it is described as a dissipative (frictional) force and thermal noise on time scales of the order of 10^{-3} s. In an analogy to this example, for a wide range of systems, including biomechanical systems [1] and granular systems [2], it might be expected that a fluctuating force obtained through some coarse-graining procedure can be decomposed into a dissipative force and other components. In the regime near equilibrium, such a decomposition is uniquely determined by fluctuation-dissipation relations (FDRs). However, no rule is known that determines such a decomposition far from equilibrium [3]. We wish to discover a rule of this kind for a class of systems exhibiting fluctuating forces.

With the above-stated purpose, in the present paper, we study a Langevin equation describing the motion of a Brownian particle with a tilted periodic potential in a one-dimensional space. Although we study the simplest system realizing nonequilibrium steady states (NESSs), the arguments below can be applied to a wide class of Brownian motors [4–6]. The Langevin equation that we analyze is described by

$$\gamma \frac{dx}{dt} = f - \frac{dU(x)}{dx} + \xi(t). \tag{1}$$

Here, γ is a friction constant, $U(x)$ is a periodic potential of period ℓ, f is a constant external driving force, and ξ is Gaussian white noise satisfying

$$\langle \xi(t)\xi(t')\rangle = 2\gamma T \delta(t-t'), \tag{2}$$

where T is the temperature of the environment, and the Boltzmann constant is set to unity. We consider a description of large-scale motion, which is obtained by taking an average over a time interval δt that is chosen to be sufficiently longer than the characteristic time of the system. In this description, the finite time average of the force $-dU/dx$ acts on the particle as a fluctuating force. We conjecture that this fluctuating force can be decomposed into a dissipative force, an extra driving force, and random noise.

In order to investigate time-averaged quantities, including that of $-dU/dx$, we introduce the finite time average of an arbitrary quantity $Z(t)$

$$\bar{Z}_n \equiv \frac{1}{\delta t}\int_{t_n}^{t_{n+1}} dt Z(t), \tag{3}$$

where $t_n = n\delta t$, $n=0,1,2,\ldots$. Then, the finite time average of $-dU/dx$ that we consider is given by

$$-\overline{\frac{dU}{dx}}_n = -\frac{1}{\delta t}\int_{t_n}^{t_{n+1}} dt \left.\frac{dU}{dx}\right|_{x=x(t)}. \tag{4}$$

We hypothesize that this can be decomposed into a dissipative component $A(x_{n+1}-x_n)/\delta t$, where $x_n \equiv x(t_n)$, and a nondissipative component. That is, we assume the form

$$-\overline{\frac{dU}{dx}}_n = A\frac{x_{n+1}-x_n}{\delta t} + B_n, \tag{5}$$

where A is a constant and B_n is a fluctuating quantity whose statistical average $\langle B_n\rangle$ takes a nonzero value $f^{\rm p}$

$$B_n = f^{\rm p} + \delta B_n. \tag{6}$$

The quantities $f^{\rm p}$ and δB_n correspond to an extra driving force and random noise, respectively. Substituting (5) into an integrated form of (1), we obtain

$$(\gamma - A)\frac{x_{n+1}-x_n}{\delta t} = f + f^{\rm p} + \delta B_n + \bar{\xi}_n. \tag{7}$$

Then, with the definitions

$$\Gamma \equiv \gamma - A, \tag{8}$$

$$F \equiv f + f^{\rm p}, \tag{9}$$

$$\Xi_n \equiv \delta B_n + \bar{\xi}_n, \tag{10}$$

we express (7) as

$$\Gamma \frac{x_{n+1} - x_n}{\delta t} = F + \Xi_n, \tag{11}$$

where Ξ_n is expected to exhibit a Gaussian distribution for large δt. This equation is regarded as an effective model of (1).

The main claim of this Rapid Communication is that the simple condition

$$\lim_{\delta t \to \infty} \delta t \langle \delta B_n \bar{\xi}_n \rangle = 0 \tag{12}$$

uniquely determines the constant A in (5). Note that a correlation of time-averaged quantities is proportional to δt^{-1} in general and (12) indicates that the proportional constant for the case $\langle \delta B_n \bar{\xi}_n \rangle$ becomes zero. The condition (12) implies that $A(x_{n+1} - x_n)/\delta t$, which fluctuates in time, can be distinguished from B_n by the condition (12) [7]. After presenting the proof of this claim, we remark on three important topics related to it: a general inequality obtained as a direct application of (12), energetic considerations related to (12), and the relation between (12) and a time-reversal symmetry in the stochastic sense.

Preliminary consideration. Before presenting the proof of the main claim, we first consider how the parameters of the effective model (11) can be expressed in terms of the steady state velocity v_s and the diffusion constant D, defined as $v_s \equiv \lim_{t\to\infty} \langle (x(t) - x(0))/t \rangle$ and $D \equiv \lim_{t\to\infty} \langle (x(t) - x(0) - v_s t)^2 / 2t \rangle$. Because v_s and D are independent of the scale on which we describe the system, the same values of v_s and D should be obtained from (11). This implies the relations $F = \Gamma v_s$ and

$$\langle \Xi_n \Xi_m \rangle = 2 \delta_{nm} D \Gamma^2 (\delta t)^{-1}. \tag{13}$$

From these expressions, all the parameters of the effective model (11) are given in terms of v_s and D when Γ is determined.

In the equilibrium case ($f=0$), Γ should satisfy the relation $D\Gamma^2 = \Gamma T$, which is referred to as the FDR of the second kind [8]. From this, Γ is expressed as

$$\Gamma = \frac{T}{D}. \tag{14}$$

Furthermore, we can prove

$$D = \mu T, \tag{15}$$

where μ is the mobility, defined as $\mu = \lim_{f\to 0} v_{\rm s}(f)/f$. The two FDRs (14) and (15) lead to

$$\Gamma = \mu^{-1}. \tag{16}$$

However, for NESSs far from equilibrium, (15) is violated [9], and (14) does not hold in general. Therefore, for treatment of such systems, it is necessary to find a guiding principle to determine Γ.

In a previous work [9], we studied NESSs and proposed a natural method to determine Γ by considering the response of the particle to a slowly varying potential $V(x)$ in space. Below, we present a heuristic argument from which the result obtained there can be understood. For detailed presentation of the systematic perturbation method used to derive this result, see Ref. [9].

Because the gradient of the slowly varying potential $V(x)$ can be regarded as a modulation of the external force f, the large-scale motion in the modulated system is described by

$$\frac{x_{n+1} - x_n}{\delta t} \simeq v_{\rm s}\left(f - \frac{dV(x_n)}{dx_n} \right) + \frac{\Xi_n}{\Gamma} \simeq v_{\rm s}(f) - \mu_{\rm d} \frac{dV}{dx_n} + \frac{\Xi_n}{\Gamma}, \tag{17}$$

where $\mu_{\rm d}$ is the differential mobility defined by

$$\mu_{\rm d} \equiv \frac{dv_{\rm s}(f)}{df}. \tag{18}$$

Then, assuming that $-dV/dx_n$ is the force acting on the particle even in this effective description, from (17) we obtain the result

$$\Gamma = \mu_{\rm d}^{-1}. \tag{19}$$

Note that this expression represents an extension of (16) to the presently considered nonequilibrium case. From (8) and (19), we find that the constant A appearing in (5) should satisfy

$$\gamma - A = \mu_{\rm d}^{-1}. \tag{20}$$

Proof. We demonstrate that the decomposition condition (12) uniquely determines the constant A, yielding (20). Using (5) and (7), we rewrite the condition (12) as

$$\lim_{\delta t\to\infty} \delta t \left\langle \left(\overline{\frac{dU}{dx_n}} + A \frac{x_{n+1} - x_n}{\delta t} \right) \left(\gamma \frac{x_{n+1} - x_n}{\delta t} + \overline{\frac{dU}{dx_n}} \right) \right\rangle_{\rm c} = 0, \tag{21}$$

where $\langle \ \rangle_{\rm c}$ represents the cumulant. Through the definition

$$G \equiv - \lim_{\delta t\to\infty} \delta t \left\langle \overline{\frac{dU}{dx_n}} \frac{x_{n+1} - x_n}{\delta t} \right\rangle_{\rm c}, \tag{22}$$

it is easy to obtain

$$\gamma - A = \frac{2\gamma T}{2\gamma D - G}. \tag{23}$$

In order to connect (23) with the differential mobility $\mu_{\rm d}$ defined by (18), we express it in terms of D and $v_{\rm s}$. We start with the path integral representation

$$\langle x(\delta t) - x(0) \rangle = \int \mathcal{D}x [x(\delta t) - x(0)] \times e^{-(1/4\gamma T)\int_{-\infty}^{\delta t} dt (\gamma \dot{x} - f + (dU/dx))^2}. \tag{24}$$

Then, differentiating both sides with respect to f, we derive

$$\frac{d}{df} \langle x(\delta t) - x(0) \rangle = \frac{D}{T} \delta t - \frac{G}{2\gamma T} \delta t + O(\delta t^2) \tag{25}$$

for large δt. This leads to the relation

$$\mu_{\rm d} = \frac{2\gamma D - G}{2\gamma T}. \tag{26}$$

Comparing (23) and (26), we have arrived at (20).

Inequalities. As a simple application of the decomposition condition (12), we derive several useful inequalities. From the square of both sides of (10) and the condition (12), we obtain

$$2\Gamma^2 D = \delta t \langle (\delta B_n)^2 \rangle + 2\gamma T, \tag{27}$$

where we have used (13). This immediately leads to an inequality relating the intensity of the force noise in the original system and the quantity representing its effective value in the coarse-grained system

$$\Gamma^2 D \geqslant \gamma T. \tag{28}$$

Because $\Gamma = \mu_{\mathrm{d}}^{-1}$ [see (19)], (28) can be written as

$$D \geqslant \gamma \mu_{\mathrm{d}}^2 T. \tag{29}$$

We believe that this inequality holds in other Brownian motors [4–6] because the path integral expression is valid even for cases with a time-dependent potential and the expressions (21)–(26) given in the proof are the same for those models. The inequality (29) involves only directly measurable quantities and therefore can be tested experimentally.

In a related work, Sasaki conjectured that the inequality $D \geqslant \mu_{\mathrm{d}} T$ holds generally for Brownian motors [10]. If we define the effective temperature T_{eff} using the FDR violation factor [9], this conjecture is equivalent to the assertion that T_{eff} is not less than the temperature of the environment, T. Because it has been observed that $T_{\mathrm{eff}} > T$ in glassy systems [11] and driven many-body systems [12,13], the inequality $D \geqslant \mu_{\mathrm{d}} T$ does seem plausible. However, Sasaki reported that this inequality is violated for the model (1) with an appropriate choice of $U(x)$ [14]. This result leads us to believe that, in the present context, if there exists a generally valid inequality among measurable quantities, perhaps it involves the intensity of the force noise and its effective one, not the temperature and its effective one.

Energetics. In equilibrium systems, heat is distinguished from work according to the second law of thermodynamics. However, obviously, heat can be considered as a mechanical work done by a force at a microscopic scale. This tempts us to investigate how heat and work come to be expressed in different ways through a coarse-graining procedure. We treat this problem on the basis of the decomposition of the force $-\overline{dU/dx_n}$ given by (5).

In Langevin systems, the heat absorbed from a heat bath is interpreted as the work done by a force $-\gamma dx/dt + \xi$ exerted by the heat bath [15]. With this interpretation, the heat absorbed during an interval $t_n \leqslant t \leqslant t_{n+1}$ can be expressed as

$$q_n = -\int_{t_n}^{t_{n+1}} \left(\gamma \frac{dx}{dt} - \xi \right) \circ dx(t), \tag{30}$$

where the symbol $\circ$ indicates that the integral here is the stochastic Stieltjes integral in the Stratonovich sense [15]. Then, the energy balance equation for the model (1) is derived as

$$U(x_{n+1}) - U(x_n) = q_n + f(x_{n+1} - x_n). \tag{31}$$

Through similar considerations applied to the effective model (11), we define the heat (absorbed from an effective heat bath) as

$$Q_n = -\left(\Gamma \frac{x_{n+1} - x_n}{\delta t} - \Xi \right)(x_{n+1} - x_n). \tag{32}$$

With this, the energy balance equation for (11) is obtained as

$$Q_n + F(x_{n+1} - x_n) = 0. \tag{33}$$

The difference between the two quantities q_n and Q_n becomes obvious when their steady state averages are compared; we have $\langle q_n \rangle = -f v_s \delta t$ and $\langle Q_n \rangle = -F v_s \delta t$, while it is known that outside the linear response regime, in general $F \neq f$ (see Fig. 2 of Ref. [9]).

In order to understand the difference between q_n and Q_n, we consider a decomposition of the work done by the force $-dU/dx$ during a time interval $t_n \leqslant t \leqslant t_{n+1}$, which is written as

$$-\overline{\frac{dU}{dx}\frac{dx}{dt_n}}\delta t = Q_n^{\mathrm{p}} + W_n^{\mathrm{p}}. \tag{34}$$

Although we conjecture that Q_n^{p} and W_n^{p} correspond to "heat" and "work," respectively, no rule is known that distinguishes heat from work in this case. However, in the present system, it seems natural to assume

$$W_n^{\mathrm{p}} = f^{\mathrm{p}}(x_{n+1} - x_n), \tag{35}$$

because the extra driving force f^{p} was determined from the decomposition condition (12). With this assumption, we can derive the relation

$$Q_n = q_n + Q_n^{\mathrm{p}}, \tag{36}$$

which provides a clear interpretation of the difference between q_n and Q_n.

The argument above leads to the following question: is there a simple rule of energetics from which we can obtain the decomposition (34) along with (35) without the decomposition condition (12)? This question will be studied in the future. In addition to their role in such fundamental problems, energetic considerations applied to different time scales may be useful when we attempt to interpret the efficiency of motor proteins [16] within stochastic models [17].

Adjoint dynamics. As a final topic here, we consider the relation between the decomposition condition (12) and time-reversal symmetry. In order to represent this symmetry explicitly, we consider the path probability density P for a discrete time series $[x]_N = (x_0, x_1, \ldots, x_N)$ generated within the model under consideration. The time-reversed path probability density P^* is defined by $P^*([x]_N) = P([\tilde{x}]_N)$, where $[\tilde{x}]_N$ represents the time reversed trajectory of $[x]_N$, that is, $\tilde{x}_n = x_{N-n}$. When P^* is obtained from the frequency distribution of trajectories in a steady state for some stochastic dynamics, these dynamics are called adjoint dynamics.

When the constant A appearing in (5) is chosen correctly, (7) can be regarded as an effective Langevin model. In this case, we find that adjoint dynamics are described by

$$(\gamma - A)\frac{x_{n+1} - x_n}{\delta t} = - f - f^{\mathrm{p}} + \delta B_n + \bar{\xi}_n. \quad (37)$$

Then, note that the condition (12) is equivalent to

$$\langle(- \delta B_n + \bar{\xi}_n)^2\rangle = \langle(\delta B_n + \bar{\xi}_n)^2\rangle. \quad (38)$$

Because both δB_n and $\bar{\xi}_n$ exhibit Gaussian distributions for sufficiently large δt, the condition (38) allows us to replace $\delta B_n + \bar{\xi}_n$ in (37) by $-\delta B_n + \bar{\xi}_n$. Thus, the adjoint dynamics can be expressed as

$$(\gamma - A)\frac{x_{n+1} - x_n}{\delta t} = - f - B_n + \bar{\xi}_n. \quad (39)$$

Then, because (7) can be rewritten as

$$(\gamma - A)\frac{x_{n+1} - x_n}{\delta t} = f + B_n + \bar{\xi}_n, \quad (40)$$

we find that the decomposition of $-\overline{dU/dx_n}$ given by (5) is characterized by parity with respect to time reversal in the stochastic sense. More specifically, the dissipative force $A(x_{n+1} - x_n)/\delta t$ remains, while the other contribution, B_n, changes sign in the adjoint dynamics.

In a previous study related to adjoint dynamics, Bertini *et al.* succeeded in deriving the large deviation functional S of the density profile for a special model of the hydrodynamic equation $\partial_t \rho = \mathcal{D}(\rho)$, which is obtained as the continuum limit for a nonequilibrium lattice gas [18]. In their analysis, the equation describing the adjoint dynamics $\partial_t \rho = \mathcal{D}^*(\rho)$ was rigorously derived in the form

$$\mathcal{D}^*(\rho) = \frac{1}{2}\nabla\left(\chi(\rho)\nabla\frac{\delta S}{\delta \rho}\right) - \mathcal{A}(\rho) \quad (41)$$

for the case in which $\mathcal{D}(\rho)$ is given by

$$\mathcal{D}(\rho) = \frac{1}{2}\nabla\left(\chi(\rho)\nabla\frac{\delta S}{\delta \rho}\right) + \mathcal{A}(\rho), \quad (42)$$

where $\chi(\rho)$ is the current noise intensity. They called the relation $\mathcal{D}(\rho) + \mathcal{D}^*(\rho) = \nabla(\chi(\rho)\nabla(\delta S/\delta\rho))$ the fluctuation-dissipation relationship for NESSs far from equilibrium, because it reduces to one expression among the linear response relations for states near equilibrium, where the relation $\mathcal{D}(\rho) = \mathcal{D}^*(\rho)$ holds due to detailed balance. We remark that (41) and (42) are similar to (39) and (40) in the model we study.

Conclusion. We have found that the decomposition condition (12) leads to the relation $\Gamma = \mu_{\mathrm{d}}^{-1}$, which was obtained in our previous study [9]. The condition (12), which is related to time-reversal symmetry, yields the new inequality (29) and leads to an interesting question regarding the decomposition of work given in (34).

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