# **Energy representation for nonequilibrium Brownian-like systems: Steady states and fluctuation relations**

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Stochastic dynamics in the energy representation is used as a method to represent nonequilibrium Brownianlike systems. It is shown that the equation of motion for the energy of such systems can be taken in the form of the Langevin equation with multiplicative noise. Properties of the steady states are examined by solving the Fokker-Planck equation for the energy distribution functions. The generalized integral fluctuation theorem is deduced for the systems characterized by the shifted probability flux operator. From this theorem, a number of entropy and fluctuation relations such as the Evans-Searles fluctuation theorem, the Hatano-Sasa identity, and the Jarzynski's equality are derived.

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## **I. INTRODUCTION**

According to the basic principles of thermodynamics, when a macroscopic system is brought into contact with a thermostat (reservoir, heat bath), the system evolves in time approaching the equilibrium state in the course of relaxation. The state of equilibrium is well defined only under certain idealized conditions  $\lceil 1-3 \rceil$  $\lceil 1-3 \rceil$  $\lceil 1-3 \rceil$  so that, at equilibrium, the thermodynamical parameters of the system are adjusted to the values of the thermostat whereas all intrinsic flows have come to an end.

In most cases, however, systems are subject to nonequilibrium conditions and external constraints  $[3-9]$  $[3-9]$  $[3-9]$ . In open systems out of equilibrium, flows universally present and generally cannot cease to exist. So, it is difficult if not impossible to determine governing parameters that can be held constant.

Nevertheless, there are the stationary states that can be unambiguously defined for certain open systems  $[4]$  $[4]$  $[4]$ . Although such states are independent of time and thus might be regarded as "equilibrium" ones, the thermodynamical parameters of the system may significantly differ from those for the environment and, more generally, the stationary distribution functions cannot be described using the well-known equilibrium distributions.

Among all the nonequilibrium systems, the most studied and important case is represented by an ensemble of Brownian particles. In spite of the fact that Brownian motion has long been the subject of intense studies (a recent review on its history can be found in  $[10,11]$  $[10,11]$  $[10,11]$  $[10,11]$ ), it is still interesting to understand the behavior of Brownian particles as a model system driven far from equilibrium.

It should be emphasized that the theory of Brownian motion can be applied to the systems where the term "Brownian particle" does not mean a real particle. For example, it may point to some collective property of macroscopic systems such as the concentration of any component of a chemically reacting system  $[7]$  $[7]$  $[7]$ . For brevity, such systems will be referred to as the *Brownian systems*.

Though far-from-equilibrium Brownian systems are abundant in nature, there is no unified commonly accepted theoretical approach to determine possible states of such systems. Hence it is a fundamentally important task to develop a method to explore general properties of stationary states of open systems and to establish the conditions of their existence.

In equilibrium statistical mechanics, these states are known to be generally described in terms of the energy surfaces giving, for certain systems, microcanonical and canonical ensembles  $\lceil 1,3 \rceil$  $\lceil 1,3 \rceil$  $\lceil 1,3 \rceil$  $\lceil 1,3 \rceil$ . In a similar spirit, our considerations will be based on the *energy representation* where the states of the Brownian system are determined solely by their energies.

Note that similar representation has been previously used in energy controlled stochastic models such as the random energy model  $[12,13]$  $[12,13]$  $[12,13]$  $[12,13]$  and its generalizations  $[14]$  $[14]$  $[14]$ . The en-ergy master equation was also derived in Ref. [[15](#page-8-10)] as an low-temperature approximation describing energy fluctuations in the Bässler's phenomenological random-walk model for viscous liquids.

Typically, the interaction between the Brownian system such as a Brownian particle and the environment involves the process of direct energy interchange. During this process, the dissipation may take the energy away from the system leading to a loss of its energy (the positive friction). In the opposite case of negative friction, the transfer of energy from the thermostat results in the energy input.

In addition to the deterministic part of interaction, there are fluctuation effects of the environment that affect the system giving rise to rapid change of its state. Such changes may take place when the structure of the environment is complicated by the presence of additional systems or some of its characteristics can be directly influenced by the processes running in the Brownian system.

So, the above pattern suggests using stochastic dynamics to describe the behavior of Brownian systems interacting with surrounding media. Therefore, there is an ensemble of such systems characterized by the probability distribution

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function that, in particular, define stationary (steady) states formed under nonequilibrium conditions. Typically, in systems far from equilibrium, the distribution functions of the stationary states significantly differ from the well-known equilibrium distributions. For example, dissipative dynamics of the noise driven Hookean model for protein folding studied in Ref.  $[16]$  $[16]$  $[16]$  was found to be characterized by a distribution of energy states obeying a modified one-dimensional Ornstein-Uhlenbeck process.

In this paper, we suggest using the Langevin dynamics  $[17,18]$  $[17,18]$  $[17,18]$  $[17,18]$  in the energy representation to describe the class of systems called the Brownian systems. Properties of the stationary (steady) states of such systems will be of our primary concern. The layout of the paper is as follows.

In Sec. [II,](#page-1-0) we start with the purely dissipative dynamics of Brownian particles and show that, in the energy representation, it is governed by the Langevin equation with the multiplicative white noise. Then we generalize the results for the Brownian particles and formulate the Langevin Eq. ([7](#page-2-0)) for out-of-equilibrium Brownian-like systems.

In Sec. [III,](#page-2-1) the steady states of the Brownian systems are obtained as the stationary solutions of the Fokker-Planck equation and are characterized by the probability flux number,  $J_{st}$ . It is found that the steady-state distributions are additionally determined by the effective energy potential Eq.  $(12)$  $(12)$  $(12)$ ] and by the energy dependent diffusion coefficient. We also discuss some important examples of the steady states formed when the probability flux vanishes,  $J_{st}=0$ , and the distributions take the potential form.

In Sec. [IV A,](#page-4-0) we define the trajectory-dependent entropies whose ensemble averages can be associated with the entropy production rates of the system and of the environment. The steady-state values of the rates are evaluated. The rate of the medium entropy is found to depend heavily on the effective potential. Then, in Sec. [IV B,](#page-5-0) we derive the generalized integral fluctuation relation  $(47)$  $(47)$  $(47)$  for Brownian systems with the shifted probability flux operator characterized by the flux parameter and the probability flux number. It is shown that there are a number of the known fluctuation relations along with the fluctuation-dissipation theorem for the steady-state systems that immediately follow from the relation  $(47)$  $(47)$  $(47)$ .

We discuss our results and make some concluding remarks in Sec. [V.](#page-7-0)

## <span id="page-1-0"></span>**II. BROWNIAN PARTICLES AND ENERGY REPRESENTATION**

#### **A. Brownian particles**

The energy representation, where the states of the system are distinguished solely by their energies,  $\varepsilon$ , can be viewed as the very basic description of the nonequilibrium system behavior provided that other dynamical variables, for some reasons, are irrelevant and can be disregarded. In this section, the well-known case of purely dissipative dynamics of a Brownian particle  $[19]$  $[19]$  $[19]$  will be our initial concern. For this simple model system, we introduce the energy representation by deriving the Langevin equation for the energy of the particle.

So, we take, as the starting point, the following Langevin equation:

$$
\partial_t p_i = F_i + \eta_i,\tag{1}
$$

<span id="page-1-1"></span>where  $\mathbf{p} = (p_1, p_2, p_3)$  is the momentum of the particle;  $F_i =$  $-\frac{\partial E}{\partial p_i} \equiv -\partial_i E$  is the *i*th component of the force expressed in terms of the potential,  $E = E(p)$ . Note that the linear friction force  $\mathbf{F} = -\gamma \mathbf{p}$  represents the special case where the potential is proportional to the kinetic energy,  $\varepsilon = p^2/(2M)$  (*M* is the particle mass):  $E = \gamma p^2 / 2 = \gamma M \varepsilon$ , ( $\gamma$  is the friction coefficient).

For the white Gaussian noise with  $\langle \eta_i(t) \rangle = 0$  and  $\langle \eta_i(t) \eta_j(t') \rangle = 2\sigma^2 \delta_{ij} \delta(t-t')$ , where  $2\sigma^2$  is the intensity of the random force, Eq.  $(1)$  $(1)$  $(1)$  is known to give the Fokker-Planck (FP) equation

$$
\partial_t P(\mathbf{p}, t) = \partial_i [\sigma^2 \partial_i - F_i] P(\mathbf{p}, t)
$$
 (2)

<span id="page-1-2"></span>describing the stochastic dynamics as the evolution of the probability distribution function  $P(\mathbf{p}, t) = \langle \delta(\mathbf{p}(t) - \mathbf{p}) \rangle$ . The equilibrium distribution

$$
P_{\text{eq}}(\mathbf{p}) \propto \exp[-E(p)/\sigma^2] \tag{3}
$$

<span id="page-1-3"></span>then can be derived as the stationary solution to the FP Eq. ([2](#page-1-2)). For  $E = \gamma M \varepsilon$ , Eq. ([3](#page-1-3)) gives the well-known Boltzmann distribution provided the friction coefficient,  $\gamma$ , the mass,  $M$ , and the noise intensity,  $\sigma^2$ , are related to the inverse temperature,  $\beta = 1/(k_B T)$ , through the Einstein relation:  $\gamma M / \sigma^2 = \beta$ .

Our task now is to deduce the Langevin equation for the energy of the Brownian particle,  $\varepsilon$ . To this end we start from the FP Eq. ([4](#page-1-4)) written in the spherical coordinates  $(p, \theta, \phi)$ . Then, after averaging over angles and making the change of variables,  $p \rightarrow \varepsilon = p^2 / (2M)$ , we have

$$
\partial_t \widetilde{P}(\varepsilon, t) = \partial_{\varepsilon} \left[ \widetilde{\sigma}^2 \sqrt{\varepsilon} \partial_{\varepsilon} \sqrt{\varepsilon} + (\varepsilon \widetilde{E}' - \widetilde{\sigma}^2) \right] \widetilde{P}(\varepsilon, t), \tag{4}
$$

<span id="page-1-4"></span>where  $\tilde{\sigma}^2 = \sigma^2/(8M)$ ,  $\tilde{E} = E/(8M)$ ,  $\tilde{P}(\varepsilon, t)$  is the energy distribution function and prime stands for the derivative with respect to the energy,  $\varepsilon$ .

The steady-state distribution found as the stationary solution of the FP equation in the energy representation  $[Eq. (4)]$  $[Eq. (4)]$  $[Eq. (4)]$ is given by

$$
\widetilde{P}_{\text{eq}}(\varepsilon) \propto \sqrt{\varepsilon} \, \exp[-\,\widetilde{\sigma}^{-2}\widetilde{E}(\varepsilon)].\tag{5}
$$

<span id="page-1-5"></span>The difference between the distributions given in Eqs. ([3](#page-1-3)) and ([5](#page-1-5)) is due to the additional square root factor  $\sqrt{\epsilon}$  that accounts for the Jacobian of the transformation:  $p^2 dp$  $\propto \sqrt{\varepsilon}d\varepsilon$ .

For the Boltzmann equilibrium distribution with  $E/\sigma^2$  $=\vec{E}/\vec{\sigma}^2 = \beta \varepsilon$ , it can be readily seen that the function ([5](#page-1-5)) reaches its maximum value at  $\varepsilon = k_B T/2$  and the equilibrium mean value of the energy is  $\langle \varepsilon \rangle_{\text{eq}} = 3k_B T/2$ . This case represents the equilibrium conditions, when the Brownian system is at equilibrium and the environment plays the role of the thermostat.

Finally, from Eq. ([4](#page-1-4)), it can be inferred that the Langevin equation in the energy representation is given by

<span id="page-2-2"></span>This result shows that the stochastic equation governing the dynamics of the Brownian particle energy is characterized by the multiplicative noise.

Note that, though different representations of the Brownian motion are equivalent, the energy representation can be the preferential approach when it is necessary to take into account both loss and gain (dissipation and absorption) of the energy. The latter process implies that the Brownian particle is subjected to "negative" friction. We can now draw some generalizations from the results for the Brownian particle and treat the general case of Brownian systems.

#### **B. Langevin dynamics in energy representation**

Our basic assumption is that, similar to the above discussed Brownian particle, the energy dynamics of the Brownian system is governed by the Langevin equation with multiplicative noise of the general form

$$
\partial_t \varepsilon = -f(\varepsilon) + g(\varepsilon)\xi(t),\tag{7}
$$

<span id="page-2-0"></span>where  $\varepsilon$  is the energy,  $f(\varepsilon)$  is the function giving the *rate of*  $$ tion;  $\xi(t)$  represents Gaussian white noise. Mathematically, this equation can be regarded as the generalized version of Eq. ([6](#page-2-2)), where  $f(\varepsilon) = \varepsilon \tilde{E}' - \tilde{\sigma}^2$  and  $g(\varepsilon) = \sqrt{\varepsilon}$ .

The first term on the right hand side of dynamical Eq.  $(7)$  $(7)$  $(7)$ is due to the direct action of the environment on the system, The effect of direct external action described by the *exchange function,*  $f(\varepsilon)$ , is determined by the conditions under which the system is kept and by its physical characteristics.

But evolution of the system state is not determined solely by these factors. Each parameter of the system may undergo irregular variations caused either by fluctuation induced stochastic perturbations or by the complicated dynamical behavior of a nonlinear environment. These additional variations result in random migration of the system over various states.

Random influence of the environment is represented by the second term on the right hand side of Eq.  $(7)$  $(7)$  $(7)$ . This term accounts for the system-environment interaction induced by fluctuations in parameters of the system controlling conditions. It is taken in the form of multiplicative noise with the  $diffusion$  function,  $g(\varepsilon)$ , giving the energy dependent coupling strength.

Thus, for the Brownian system in contact with the environment, there are processes leading to gain and loss of the energy that underlie the Langevin dynamics in the energy representations. Our next problem is to identify the conditions for the system to be at equilibrium stationary states and to find the distribution function of the system out-ofequilibrium.

## **III. NONEQUILIBRIUM STEADY STATES**

<span id="page-2-1"></span>In the previous section we have found that, for the energy of the Brownian particle expressed in terms of the momenta,

the stochastic dynamics is governed by the Langevin Eq. ([6](#page-2-2)).

In more general nonequilibrium systems, it does not always happen that all necessary details on the dynamics of internal degrees of freedom such as the momenta are known. These nonequilibrium systems can be characterized by the energy dependent probability distribution function  $\rho(\varepsilon,t)$  $=\langle \delta[\varepsilon(t)-\varepsilon] \rangle$  describing the dynamical behavior that involves the processes of energy loss and gain induced by the environment.

Generally, the equation of motion for the energy change of out-of-equilibrium systems is complicated by nonlinearities present in both the internal dynamics of the system and in the system-environment coupling. For Brownian systems in the energy representation, this complexity can be described using the nonlinear Langevin equation with the multiplicative noise  $[Eq. (7)]$  $[Eq. (7)]$  $[Eq. (7)]$  that defines the nonlinear stochastic out-of-equilibrium dynamics.

In this section, we begin with the dynamics of the probability distribution function,  $\rho(\varepsilon, t)$ , and then examine the properties of the steady states. These are defined as the stationary solutions of the FP equation in the energy representation. This approach to the steady states closely resembles the widely accepted definition of the equilibrium distribution function which implies that, for a typical system, the distribution is determined solely by the energy as the only known integral of motion.

#### **A. Fokker-Planck dynamics**

Our first step is to deduce the FP equation that defines how the energy distribution  $\rho(\varepsilon, t)$  evolves in time. To this end, we adopt the symmetric Stratonovich convention and apply the standard procedure  $[19,20]$  $[19,20]$  $[19,20]$  $[19,20]$  to the Langevin Eq. ([7](#page-2-0)) giving the following result:

$$
\partial_t \rho(\varepsilon, t) = \partial_{\varepsilon} [\sigma^2 g(\varepsilon) \partial_{\varepsilon} g(\varepsilon) + f(\varepsilon)] \rho(\varepsilon, t). \tag{8}
$$

<span id="page-2-3"></span>Note that there are different interpretations of the Langevin Eq. ([7](#page-2-0)) that all boil down to providing the discretization rules employed to define the stochastic integral:  $\int_{t}^{t+\Delta t} g[\varepsilon(\tau)] \xi(\tau) d\tau$ . Mathematically rigorous considerations of stochastic differential equations are mostly based on the Itô calculus (the prepoint discretization rule), whereas the Stratonovich interpretation (the midpoint discretization rule) has simpler transformation properties under a change of variables and arises naturally when the delta-correlated noise is treated as the limiting case of a real noise with finite correlation time (colored noise). Since the drift term in the FP equation depends on the prescription for evaluating multiplicative noise (the results for the generalized Stratonovich prescription can be found in Ref.  $[21]$  $[21]$  $[21]$ ), the problem known as the Itô-Stratonovich dilemma arises  $[20,22,23]$  $[20,22,23]$  $[20,22,23]$  $[20,22,23]$  $[20,22,23]$ . Mathematically, the results for the Itô and Stratonovich stochastic differential equations are in one-to-one correspondence. But this correspondence is system dependent. So, additional information about the microscopic structure of the environment is required in order to decide which discretization rule is physically adequate (discussion of the dilemma in the context of "internal and external" noise is given in Chap. IX.5 of van Kampen's textbook [[23](#page-8-18)]).

The FP equation in the energy representation  $[Eq. (8)]$  $[Eq. (8)]$  $[Eq. (8)]$  can now be conveniently recast into the divergence form giving the conservation law

$$
\frac{\partial \rho(\varepsilon, t)}{\partial t} = \frac{\partial J(\varepsilon, \rho)}{\partial \varepsilon},\tag{9}
$$

<span id="page-3-1"></span>characterized by the probability flux *J*

$$
J = D(\varepsilon)\{V'(\varepsilon) + \partial_{\varepsilon}\}\rho \equiv \hat{\mathcal{J}}\rho \tag{10}
$$

<span id="page-3-2"></span>with the flux operator

$$
\hat{\mathcal{J}} = D(\varepsilon)\{V'(\varepsilon) + \partial_{\varepsilon}\},\tag{11}
$$

<span id="page-3-0"></span>where  $D(\varepsilon) = \sigma^2 g^2(\varepsilon)$  is the *diffusion coefficient* and *V* is the *effective energy potential* given by

$$
V(\varepsilon) = \ln|g(\varepsilon)| + \int_{\varepsilon_0}^{\varepsilon} f(\varepsilon') / D(\varepsilon') d\varepsilon'.
$$
 (12)

For the values of energy  $\varepsilon$  ranged between  $\varepsilon_{\min}$  and  $\varepsilon_{\max}$ ,  $\varepsilon \in [\varepsilon_{\min}, \varepsilon_{\max}]$ , the conservation law [Eq. ([9](#page-3-1))] combined with the normalization condition,  $\int_{\varepsilon_{\min}}^{\varepsilon_{\max}} \rho d\varepsilon = 1$ , gives the condition of conservation for the flow of probability:  $J|_{\varepsilon=\varepsilon_{\min}}$  $=J\big|_{\varepsilon=\varepsilon_{\max}}\equiv J_b.$ 

Temporal evolution of the probability distribution function  $\rho$  is governed by the evolution operator,  $\hat{\mathcal{U}}(t, t_0)$ , of the Fokker-Planck Eq. ([9](#page-3-1)). This operator can be found as the solution of the following initial value problem:

<span id="page-3-6"></span>
$$
-\partial_t \hat{\mathcal{U}}(t,t_0) = \hat{\mathcal{H}} \hat{\mathcal{U}}(t,t_0), \quad \hat{\mathcal{U}}(t_0,t_0) = \hat{\mathcal{I}}, \tag{13}
$$

$$
\hat{\mathcal{H}} = -\partial_{\varepsilon}\hat{\mathcal{J}}, \quad \hat{\mathcal{J}} = D[V' + \partial_{\varepsilon}], \tag{14}
$$

<span id="page-3-7"></span>where  $\hat{\mathcal{I}}$  is the identity operator;  $\hat{\mathcal{H}}$  is the Fokker-Planck operator that plays the role of the effective Hamiltonian related to the probability flux operator [Eq.  $(11)$  $(11)$  $(11)$ ].

Thus the probability distribution  $\rho(\varepsilon, t)$  evolves in time under the action of the evolution operator. This can be conveniently expressed using the quantum mechanical bracket notations as follows  $[19,24,25]$  $[19,24,25]$  $[19,24,25]$  $[19,24,25]$  $[19,24,25]$ :

$$
|\rho(t)\rangle = \hat{\mathcal{U}}(t,t_0)|\rho(t_0)\rangle, \quad \rho(\varepsilon,t) = \langle \varepsilon|\rho(t)\rangle. \tag{15}
$$

#### **B. Stationary distributions**

The general stationary solution of the FP Eq.  $(8)$  $(8)$  $(8)$ 

<span id="page-3-3"></span>
$$
\rho_{\rm st}(\varepsilon) = \exp[-V(\varepsilon)] \left\{ N_{\rm st} + J_{\rm st} \int_{\varepsilon_0}^{\varepsilon} \exp[V(\varepsilon')] / D(\varepsilon') d\varepsilon' \right\},\tag{16}
$$

where  $J_{\rm st} = J_b(\rho_{\rm st})$  is the stationary probability current and  $N_{\rm st}$ is the normalization constant, can be easily obtained by solving the first-order linear differential equation:  $\hat{J}\rho_{st} = J_{st}$ . Note that the bracket form of the equation for the steady states is

$$
\hat{\mathcal{J}}|\rho_{\rm st}\rangle = |J_{\rm st}\rangle,\tag{17}
$$

where  $\rho_{st}(\varepsilon) = \langle \varepsilon | \rho_{st} \rangle$  and  $\langle \varepsilon | J_{st} \rangle = J_{st}$ .

In the remaining part of the section we assume reflecting boundary conditions and restrict ourselves to the important special case where the stationary flow is absent,  $J_{st}=0$ . For instance, such boundary conditions apply to the case when the energy spectrum is unbounded from above,  $\varepsilon_{\text{max}} \rightarrow \infty$  and  $\varepsilon_{\min} \leq \varepsilon < \infty$ , and the steady-state distribution function rapidly decays to zero with the energy:  $\rho_{st} \rightarrow 0$  at  $\varepsilon \rightarrow \infty$ .

So, the steady-state distribution  $[Eq. (16)]$  $[Eq. (16)]$  $[Eq. (16)]$  takes the potential form

<span id="page-3-4"></span>
$$
\rho_{\rm st}(\varepsilon) = Z_{\rm st}^{-1} \exp\{-V(\varepsilon)\}, \quad Z_{\rm st} = \int_{\varepsilon_{\rm min}}^{\varepsilon_{\rm max}} \exp[-V(\varepsilon)] d\varepsilon. \tag{18}
$$

Note that, by analogy with equilibrium systems, the quantity  $F_{\text{st}}$ =−ln  $Z_{\text{st}}$  sometimes is referred to as the effective free energy  $\lceil 26 \rceil$  $\lceil 26 \rceil$  $\lceil 26 \rceil$ .

The shape of the distribution  $[Eq. (18)]$  $[Eq. (18)]$  $[Eq. (18)]$  is determined by the effective energy potential given in Eq.  $(12)$  $(12)$  $(12)$ . In particular, the distribution function reaches its extremal value at energies determined by the stationary points,  $\varepsilon = \varepsilon$ , of the potential *V*. These points can be found from the stationarity equation

$$
V'(\varepsilon_s) = \frac{1}{2D(\varepsilon_s)} [D'(\varepsilon_s) + 2f(\varepsilon_s)] = 0
$$
 (19)

that can be regarded as the condition of diffusion-drift balance,  $\sigma^2 g(\varepsilon_s) g'(\varepsilon_s) = -f(\varepsilon_s)$ , between the diffusion over states of the environment and the dissipation in the system. This balance condition gives the value of most probable steady-state energy,  $\varepsilon = \varepsilon_m$ , which corresponds to the minimum of the energy potential *V*.

In the vicinity of the most probable energy, the steadystate distribution can be approximated by the Gaussian function

$$
\rho_{\rm st}(\varepsilon) \approx \rho_G(\varepsilon) \propto \rho_{\rm st}(\varepsilon_m) \exp[-V''(\varepsilon_m)(\varepsilon - \varepsilon_m)^2/2], \tag{20}
$$

where  $V''(\varepsilon_m) = [D''(\varepsilon_m) + 2f'(\varepsilon_m)]/[2D(\varepsilon_m)]$  is the second derivative of the potential with respect to the energy.

There are a variety of typical cases representing newly formed steady states of nonequilibrium systems depending on the exchange and diffusion functions,  $f(\varepsilon)$  and  $g(\varepsilon)$ . Below we discuss some of the most important ones.

We begin with the noiseless case by assuming the singular limit of vanishing diffusion,  $g \rightarrow 0$ . Then temporal evolution of the energy distribution function initially prepared at  $\varepsilon$  $=\varepsilon_0$  with  $\rho(\varepsilon, 0) = \delta(\varepsilon - \varepsilon_0)$ , is as follows:

$$
\rho(\varepsilon, t) = \delta[\varepsilon - \varepsilon(\varepsilon_0, t)],\tag{21}
$$

<span id="page-3-8"></span><span id="page-3-5"></span>where  $\varepsilon(\varepsilon_0, t)$  is the solution of the initial value problem,

$$
\partial_t \varepsilon = -f(\varepsilon) \equiv -E'(\varepsilon), \quad \varepsilon(0) = \varepsilon_0. \tag{22}
$$

Suppose that there is a local minimum of the potential  $E(\varepsilon)$ located at  $\varepsilon = \varepsilon_s$ . Then the energy  $\varepsilon_s$  is the attracting stationary (equilibrium) point that defines the stationary distribution  $\rho_{st}(\varepsilon) = \delta(\varepsilon - \varepsilon_s)$ . This implies that, when the initial value of the energy,  $\varepsilon_0$ , falls within the corresponding basin of attraction, the distribution functions  $[Eq. (21)]$  $[Eq. (21)]$  $[Eq. (21)]$  evolve in time approaching the steady state:  $\rho(\varepsilon, t) \to \delta(\varepsilon - \varepsilon_s)$  at  $t \to \infty$ .

Interestingly, when the diffusion function is a nonzero constant,  $g(\varepsilon) = g_0 \neq 0$ , the steady-state distribution is  $\rho_{st}(\varepsilon)$  $=N \exp[-E(\varepsilon)/D_0]$ , where  $D_0 = \sigma^2 g_0^2$ , so that its maxima correspond to the equilibria of the potential *E*. By contrast to the noiseless case, at  $g_0 \neq 0$ , we can have the steady state even without equilibria. An important example is the canonical equilibrium Boltzmann-Gibbs (BG) distribution with  $E(\varepsilon)/D_0 = \beta \varepsilon$ .

Note that the steady-state function takes the form of "microcanonical distribution,"  $\rho_{st}(\varepsilon) = \delta(\varepsilon - \varepsilon_0)$ , parametrized by the energy value  $\varepsilon_0$ , only if the energy is conserved,  $f(\varepsilon)$  $= 0$ , and diffusion is absent,  $g(\varepsilon) = 0$ .

The limiting case with vanishing exchange function,  $f(\varepsilon)$ =0, describes the diffusion controlled systems. From Eq. ([18](#page-3-4)), we obtain the stationary distribution function  $\rho_{st}(\varepsilon)$  $= N|g(\varepsilon)|^{-1}$  expressed in terms of the diffusion function  $g(\varepsilon)$ . The BG distribution,  $\rho_{BG}(\varepsilon) = N \exp[-\beta \varepsilon]$ , can be realized as the steady-state distribution only if the diffusion coefficient depends exponentially on the energy:  $D(\varepsilon) \propto \exp[2\beta \varepsilon]$ , where  $\beta$  is the inverse temperature. The exponential dependence may emerge as the special feature of the interaction between the system and the environment.

The above considerations are also applicable to the systems of Brownian particles in a randomly inhomogeneous environment. In such environment, some characteristics such as the coupling constants and the friction coefficient may contain stochasticity induced contributions and thus become random variables. Examples include large particles in the inhomogeneous environment, impurity particles placed into the dusty plasma, as well as the systems whose kinetic properties depend nonlinearly on the velocity or the energy of particles.

As another simple example, we consider what happen when the second viscosity in mixtures and acoustic flow comes into play due to the dependence of the friction coefficient on the velocity  $[27]$  $[27]$  $[27]$ . In this case we can take the assumption that, in the Langevin Eq.  $(7)$  $(7)$  $(7)$  with the linear exchange function  $f(\varepsilon) = \gamma \varepsilon$ , the friction coefficient  $\gamma = \langle \gamma \rangle$  $+\xi(t)$  is a sum of its average value,  $\langle \gamma \rangle$ , and the noise term  $\xi$ that accounts for random variations of the friction coefficient in the environment that occur on the scales much less than those of the observed spatial variations of kinetic processes. From Eq. ([18](#page-3-4)) with  $f(\varepsilon) = \gamma \varepsilon$  and  $g(\varepsilon) = \varepsilon$ , we obtain the power law

$$
\rho_{\rm st}(\varepsilon) = N/\varepsilon^{\nu},\tag{23}
$$

<span id="page-4-1"></span>where  $\nu = 1 + \langle \gamma \rangle / \sigma^2$ , describing the dependence of the stationary distribution on the energy. Note that similar result can be found in Ref.  $[22]$  $[22]$  $[22]$  and the above power law distribution significantly differs from the well-known solution for the case where the coefficients of friction and diffusion are independent of the energy.

In the case of the "negative friction" with  $\langle \gamma \rangle < 0$ , the system absorbs the energy and, for the semi-indefinite energy interval  $[\varepsilon_{\min}, \infty)$ , the stationary solution ([23](#page-4-1)) does not represent the steady-state distribution as it fails to meet the normalization condition. So, in order to have a steady state, we need to introduce a mechanism that limits the energy absorption. For this purpose, we consider the system characterized

by the quadratic exchange function  $f(\varepsilon) = \langle \gamma \rangle \varepsilon + \gamma_2 \varepsilon^2$ , where the second order term with the positive coefficient  $\gamma_2 > 0$ bounds the energy absorption from above, so that  $f > 0$  at  $\epsilon > \max\{-\langle \gamma \rangle / \gamma_2, 0\}$ . At  $\gamma_2 > 0$ , the power law distribution  $[Eq. (23)]$  $[Eq. (23)]$  $[Eq. (23)]$  assumes the following modified form:

$$
\rho_{\rm st}(\varepsilon) = N \varepsilon^{-\nu} \exp\bigg[ -\frac{\gamma_2}{\sigma^2} \varepsilon \bigg]. \tag{24}
$$

<span id="page-4-2"></span>Interestingly, at  $\gamma_2 / \sigma^2 = \beta$  and  $\nu = (2-n)/2$  [ $\langle \gamma \rangle / \sigma^2 = -n/2$ ], the steady-state distribution  $[Eq. (24)]$  $[Eq. (24)]$  $[Eq. (24)]$  gives the well-known Maxwell distribution function of Brownian particles moving in the *n*-dimensional Euclidean space.

In conclusion, let us briefly comment on the so-called phenomenological Rayleigh model of active friction  $[28]$  $[28]$  $[28]$ . In this model, the friction coefficient expressed in terms of the velocity  $\gamma = -\gamma_0 + \gamma_2 v^2 \equiv \gamma_2 (v^2 - v_0^2) = \alpha (\varepsilon - \varepsilon_0)$ , where  $v_0^2$  $= \gamma_0 / \gamma_2$ , is negative at small velocities,  $v^2 < v_0^2$ , and, in the low energy region, the system absorbs the energy from the environment. By contrast, in the high energy region where  $\varepsilon > \varepsilon_0$ , the friction coefficient is positive and characterizes the exchange process accompanied by the loss of energy. In the case of the constant diffusion coefficient  $D_0 = \sigma^2 g_0^2$  with  $g = g<sub>0</sub>$ , the steady-state distribution is given by

$$
\rho_{\rm st}(\varepsilon) = N \exp\bigg[ -\frac{\alpha}{3D_0} (\varepsilon - \varepsilon_0)^2 (\varepsilon + \varepsilon_0/2) \bigg]. \tag{25}
$$

The only maximum of this distribution is located at  $\varepsilon = \varepsilon_0$ giving the most probable value of the energy that defines the unique steady-state distribution  $\rho_{st}(\varepsilon) = \delta(\varepsilon - \varepsilon_0)$  in the limit of low noise  $g_0 \rightarrow 0$ .

#### **IV. ENTROPY AND FLUCTUATION RELATIONS**

#### **A. Entropies and effective potential**

<span id="page-4-0"></span>In Sec. [III](#page-2-1) we have found that, when the stationary probability flux is zero,  $J_{st}=0$ , the steady-state distribution [Eq.  $(18)$  $(18)$  $(18)$ ] in the energy representation is completely determined by the effective energy potential  $[Eq. (12)]$  $[Eq. (12)]$  $[Eq. (12)]$ , whereas, in more general case with nonvanishing flux,  $J_{st} \neq 0$ , the stationary distribution  $[Eq. (16)]$  $[Eq. (16)]$  $[Eq. (16)]$  additionally depends on the diffusion coefficient *D*.

In order to further clarify the role of the effective potential we consider the trajectory-dependent entropy of the system  $[29,30]$  $[29,30]$  $[29,30]$  $[29,30]$ 

$$
s(t) = -\ln[\rho(\varepsilon, t)]_{\varepsilon = \varepsilon(t; \xi)},\tag{26}
$$

<span id="page-4-3"></span>defined for the trajectory  $\varepsilon(t;\xi)$  representing the noise de-pendent solution of the Langevin Eq. ([7](#page-2-0)).

Averaging the trajectory-dependent entropy  $[Eq. (26)]$  $[Eq. (26)]$  $[Eq. (26)]$ over noise gives the well-known result

$$
\langle s(t) \rangle = -\int \rho(\varepsilon, t) \ln[\rho(\varepsilon, t)] d\varepsilon \tag{27}
$$

that can be easily obtained using the general relation

$$
\langle \phi[\varepsilon(t;\xi)] \rangle = \int \langle \delta[\varepsilon(t;\xi) - \varepsilon] \rangle \phi(\varepsilon) d\varepsilon
$$

$$
= \int \rho(\varepsilon, t) \phi(\varepsilon) d\varepsilon \equiv \langle \phi | \rho(t) \rangle, \qquad (28)
$$

where  $\phi$  is a function of the energy and integrals are taken over the whole energy range. Another useful identity

<span id="page-5-1"></span>
$$
\langle \phi(\varepsilon(t;\xi))\dot{\varepsilon}(t;\xi) \rangle = \int \langle \delta(\varepsilon(t;\xi) - \varepsilon)\dot{\varepsilon}(t;\xi) \rangle \phi(\varepsilon) d\varepsilon
$$

$$
= -\int J(\varepsilon,\rho) \phi(\varepsilon) d\varepsilon \equiv -\langle \phi | \hat{J} | \rho \rangle \tag{29}
$$

is at the heart of the derivation of the FP Eq.  $(9)$  $(9)$  $(9)$ .

In the equation of motion for the entropy of the system  $[Eq. (26)]$  $[Eq. (26)]$  $[Eq. (26)]$ 

$$
\dot{s}(t) = -(\partial_t \rho + \rho' \dot{\varepsilon})/\rho = -\left[\frac{\partial_t \rho}{\rho} + \frac{J\dot{\varepsilon}}{D\rho}\right] + V' \dot{\varepsilon}
$$
(30)

we may single out the contribution due to change in entropy of the environment,  $s<sub>m</sub>$ , related to the rate of heat exchange in the medium

$$
\dot{s}_{\rm m}(t) = -V'\dot{\varepsilon}.
$$
\n(31)

<span id="page-5-5"></span>By using the identity  $[Eq. (29)]$  $[Eq. (29)]$  $[Eq. (29)]$  it is not difficult to evaluate its average

$$
\langle \dot{s}_{\rm m}(t) \rangle = \int J(\varepsilon, \rho) V'(\varepsilon) d\varepsilon. \tag{32}
$$

<span id="page-5-7"></span>Similarly, averaging the increase in the total entropy  $s_{\text{tot}} = s$  $+s<sub>m</sub>$  gives the expression for the total entropy production rate

$$
\langle \dot{s}_{\text{tot}}(t) \rangle = \langle \dot{s}(t) + \dot{s}_{\text{m}}(t) \rangle = \int \frac{J^2(\varepsilon, \rho)}{D(\varepsilon)\rho(\varepsilon, t)} d\varepsilon \ge 0, \quad (33)
$$

which clearly cannot be negative. Upon reaching the steady state characterized by the stationary distribution [Eq.  $(16)$  $(16)$  $(16)$ ], the production rates of the total and medium entropies take their steady-state values given by

<span id="page-5-2"></span>
$$
\langle \dot{s}_{\text{tot}} \rangle_{\text{st}} = \int \frac{J_{\text{st}}^2}{D(\varepsilon)\rho_{\text{st}}(\varepsilon)} d\varepsilon
$$
  
=  $J_{\text{st}} \ln \left[ 1 + J_{\text{st}}/N_{\text{st}} \int \exp\{V(\varepsilon)\} / D(\varepsilon) d\varepsilon \right],$  (34)

$$
\langle \dot{s}_{\rm m} \rangle_{\rm st} = J_{\rm st} \Delta V, \tag{35}
$$

<span id="page-5-3"></span>where  $\Delta V = V(\varepsilon_{\text{max}}) - V(\varepsilon_{\text{min}})$ . As evident from Eqs. ([34](#page-5-2)) and ([35](#page-5-3)), the entropy production rates both tend to zero only if the stationary flux vanishes,  $J_{st}=0$ .

In the path integral representation, Langevin dynamics governed by the equation of motion  $[Eq. (7)]$  $[Eq. (7)]$  $[Eq. (7)]$  is described by the generating functional of correlation functions, *GA*, written in the functional integral form  $[19,31]$  $[19,31]$  $[19,31]$  $[19,31]$ . Applying the standard procedure  $\left[32,33\right]$  $\left[32,33\right]$  $\left[32,33\right]$  $\left[32,33\right]$ , we deduce the generating functional

$$
G[A] = \int \mathcal{D}[\varepsilon] \prod_{t} g^{-1} \exp \left\{ -S_{\text{eff}}[\varepsilon] + \int_{0}^{t} A(\tau) \varepsilon(\tau) d\tau \right\}
$$
(36)

<span id="page-5-4"></span>expressed in terms of the effective action

$$
S_{\text{eff}}[\varepsilon] = \frac{1}{2} \int_0^t \left\{ \frac{(\dot{\varepsilon} + DV')^2}{2D} - f' \right\} d\tau \tag{37}
$$

that determines the weight,  $P[\varepsilon] \propto \exp(-S_{\text{eff}})$ , of a trajectory  $\varepsilon(\tau)$ . Equation ([37](#page-5-4)) agrees with the results for the effective action previously derived in Refs.  $[21,34,35]$  $[21,34,35]$  $[21,34,35]$  $[21,34,35]$  $[21,34,35]$ .

From the expression  $(37)$  $(37)$  $(37)$ , it is straightforward to evaluate the difference in the effective action for each forward path  $\varepsilon(\tau)$  and the corresponding reversed trajectory (backward path),  $\bar{\varepsilon}(\tau) = \varepsilon(t-\tau)$ ,

$$
S_{\text{eff}}[\varepsilon] - S_{\text{eff}}[\bar{\varepsilon}] = \int_0^t V' \dot{\varepsilon} d\tau = -\Delta s_{\text{m}}.
$$
 (38)

<span id="page-5-6"></span>This formula along the medium entropy defined by the relation  $(31)$  $(31)$  $(31)$  shows that this is the entropy generation in the medium,  $\Delta s_{\rm m}$ , which is solely responsible for the change in the weight under "time reversal:"  $P[\varepsilon]/P[\bar{\varepsilon}] = \exp(\Delta s_m)$ .

Note that, for suitably defined dissipation function, this result can be regarded as a version of the Evans-Searles fluctuation theorem  $\left[36\right]$  $\left[36\right]$  $\left[36\right]$ . It is also applicable to externally driven Brownian systems. In this case, there is a set of external control parameters,  $\lambda(\tau) = {\lambda_1(\tau), \ldots, \lambda_k(\tau)}$ , that vary in time from  $\lambda_0 = \lambda(0)$  to  $\lambda_t = \lambda(t)$  according to the prescribed (forward) protocol, whereas the reversed (backward) protocol is represented by the parameters  $\bar{\lambda}(\tau) = \lambda(t - \tau)$ . The energy potential,  $V = V[\varepsilon, \lambda(t)]$ , and the diffusion coefficient, *D*  $= D[\varepsilon, \lambda(t)]$ , are now a function of the time-dependent parameters, so that the FP and flux operators are both nonstationary.

Despite the evolution operator,  $\hat{U}(t, t_0)$ , is no longer given by the exponential solution  $\hat{\mathcal{U}}(t, t_0) = \exp[-(t - t_0)\hat{\mathcal{H}}]$  to the Cauchy problem  $[Eq. (13)]$  $[Eq. (13)]$  $[Eq. (13)]$  with the stationary FP Hamiltonian, the expression for the effective action  $[Eq. (37)]$  $[Eq. (37)]$  $[Eq. (37)]$  remains intact and its protocol dependent difference,  $S_{\text{eff}}[{\varepsilon},\lambda]-S_{\text{eff}}[{\bar\varepsilon},\bar{\lambda}]$ , is still given by the formula ([38](#page-5-6)), where  $V' \equiv V'(\varepsilon, \lambda)$ . This formula and the inequality ([34](#page-5-2)) justify our definition of the entropy of the environment.

#### **B. Generalized integral fluctuation relation**

<span id="page-5-0"></span>In general, there are a number of relations that can be derived by making the comparison between the trajectories and reversed "antitrajectories." Some of these—the so-called the fluctuation theorems—were recently reviewed in Refs. [[37](#page-8-32)[–39](#page-8-33)]. The fluctuation relations were tested experimentally in a variety of different systems such as colloidal particles manipulated by laser traps  $[40-46]$  $[40-46]$  $[40-46]$ , biomolecules pulled by atomic force microscopy's (AFM's) or optical tweezers  $[47,48]$  $[47,48]$  $[47,48]$  $[47,48]$ , AFM cantilever  $[49]$  $[49]$  $[49]$ , and an electric circuit with an imposed mean current  $[46]$  $[46]$  $[46]$ .

In this section, we apply the operator approach to deduce the integral fluctuation relation that can be regarded as the generalized version of the well-known Hatano-Sasa identity [50](#page-9-4). We also discuss how to recover other known results such as the Jarzynski's equality  $\lceil 51 \rceil$  $\lceil 51 \rceil$  $\lceil 51 \rceil$  and the relation for the total entropy obtained by Seifert in Ref. [[30](#page-8-25)]. Note that the fluctuation relations for stochastic systems originally studied in Refs.  $[24,36,52,53]$  $[24,36,52,53]$  $[24,36,52,53]$  $[24,36,52,53]$  $[24,36,52,53]$  $[24,36,52,53]$  were recently extended to the case of inhomogeneous stochastic processes  $[54–56]$  $[54–56]$  $[54–56]$  $[54–56]$ .

Our first step is to introduce a family of the modified FP operators

$$
\hat{\mathcal{H}}_p = -\partial_{\varepsilon} \hat{\mathcal{J}}_p, \quad \hat{\mathcal{J}}_p = \hat{\mathcal{J}} - p/\rho_{\rm st}, \tag{39}
$$

<span id="page-6-1"></span>where *p* is a real number which might be called the *flux parameter*. From Eq. ([39](#page-6-1)) it can be seen that the operator [Eq. ([14](#page-3-7))],  $\hat{\mathcal{H}} = \hat{\mathcal{H}}_0$ , transforms into  $\hat{\mathcal{H}}_p$  with the shifted flux operator  $\hat{\mathcal{J}}_p$  when the energy derivative of the potential *V'* is replaced with  $V' - p/(D\rho_{st})$ . So, the energy potential,  $V_p$ , for the deformed FP operator  $\hat{\mathcal{H}}_p$  is given by

$$
V_p = V - \frac{p}{J_{\rm st}} \ln \left[ 1 + J_{\rm st}/N_{\rm st} \int_{\varepsilon_0}^{\varepsilon} \exp\{V(\varepsilon')\} / D(\varepsilon') d\varepsilon' \right],\tag{40}
$$

where the potential  $V$  is defined in Eq.  $(12)$  $(12)$  $(12)$ .

<span id="page-6-2"></span>It is straightforward to check the validity of the algebraic identity

$$
\hat{\mathcal{H}}_p^{\dagger} = \hat{\mathcal{J}}_p^{\dagger} \partial_{\varepsilon} = e^{\Psi_{\text{st}}} \hat{\mathcal{H}}_q e^{-\Psi_{\text{st}}}, \quad q = 2J_{\text{st}} - p, \tag{41}
$$

$$
\Psi_{st} = -\ln \rho_{st},\tag{42}
$$

where the superscript  $\dagger$  stands for Hermitian conjugation, linking the Hermitian conjugate of the deformed FP operator

[Eq. ([39](#page-6-1))],  $\hat{\mathcal{H}}_p^{\dagger}$ , at  $p = J_{st} + \Delta J$  and the operator  $\hat{\mathcal{H}}_q$  with the flux parameter  $q = 2J_{st} - p = J_{st} - \Delta J$  through the steady-state potential  $\Psi_{st}$ , of the stationary distribution,  $\rho_{st}$ , characterized by the flux number (stationary probability flux)  $J_{st}$ .

Another important point is that the evolution operator  $\hat{\mathcal{U}}_p(t,0;\bar{\lambda})$  of the deformed Hamiltonian ([39](#page-6-1)) computed at the reversed protocol  $\overline{\lambda}$  preserves the normalization condition of a probability distribution function  $\rho_f$ : $\langle 1 | \rho_f \rangle = \langle \rho_f | 1 \rangle$  $=f \rho_f(\varepsilon) d\varepsilon = 1$ . Our method is to combine the normalization preserving condition

$$
\langle 1 | \hat{\mathcal{U}}_p(t,0;\bar{\lambda}) | \rho_{\rm f} \rangle = \langle \rho_{\rm f} | \hat{\mathcal{U}}_p^{\dagger}(t,0;\bar{\lambda}) | 1 \rangle = \langle \rho_{\rm f} | 1 \rangle = 1 \qquad (43)
$$

with the identity  $[Eq. (41)]$  $[Eq. (41)]$  $[Eq. (41)]$ . To this end, we slice the time interval  $[0,t]$  into a large number  $N+1$  of small pieces of the thickness  $\Delta \tau = t/(N+1)$  and approximate the evolution operator  $\hat{\mathcal{U}}_p(\tau_i, \tau_{i-1}; \overline{\lambda})$ , where  $\tau_i = \tau_{i-1} + \Delta \tau$  and  $\tau_0 = 0$ , by the operator exponent  $\exp[-\Delta \tau \hat{\mathcal{H}}_p(\overline{\lambda}_i)],$  where  $\overline{\lambda}_i = \overline{\lambda}(\tau_i) = \lambda_{N+1-i}$ . Then, by using the identity  $[Eq. (41)]$  $[Eq. (41)]$  $[Eq. (41)]$ , the discretized evolution operator

$$
\hat{\mathcal{U}}_p^{\dagger}(t,0;\bar{\lambda}) \approx [e^{-\Delta \tau \hat{\mathcal{H}}_p(\lambda_0)} e^{-\Delta \tau \hat{\mathcal{H}}_p(\lambda_1)} \cdots e^{-\Delta \tau \hat{\mathcal{H}}_p(\lambda_N)}]^{\dagger}.
$$
 (44)

can be recast into the following operator product

$$
\hat{\mathcal{U}}_{p}^{\dagger}(t,0;\bar{\lambda}) \approx \rho_N^{-1} e^{-\Delta \tau \hat{\mathcal{H}}_q(\lambda_N)} e^{-\Delta \Psi_N} \cdots e^{-\Delta \tau \hat{\mathcal{H}}_q(\lambda_1)} e^{-\Delta \Psi_1} e^{-\Delta \tau \hat{\mathcal{H}}_q(\lambda_0)} \rho_0,
$$
\n(45)

where  $\rho_i = \rho_{st}(\lambda_i)$  and exp[ $-\Delta \Psi_i$ ] =  $\rho_i / \rho_{i-1}$ .

Assuming that the initial probability distribution is  $\rho_{\text{in}}$ , we derive the equality

$$
\langle 1|e^{-(\Psi_{\rm f}-\Psi_{\rm N})}e^{-\Delta\tau\hat{\mathcal{H}}_q(\lambda_{\rm N})}e^{-\Delta\Psi_{\rm N}}\cdots e^{-\Delta\tau\hat{\mathcal{H}}_q(\lambda_1)}e^{-\Delta\Psi_{1}}e^{-\Delta\tau\hat{\mathcal{H}}_q(\lambda_0)}e^{\Psi_{\rm in}-\Psi_{0}}|\rho_{\rm in}\rangle = 1,
$$
\n(46)

where  $\Psi_{\text{in,f}}$ =−ln  $\rho_{\text{in,f}}$ , which, in the limit of large slice number,  $N \rightarrow \infty$ , gives our key result in the form of the identity

<span id="page-6-0"></span>
$$
\left\langle \exp\left\{-\int_0^t \frac{\partial \Psi_{st}}{\partial \lambda_\alpha} \dot{\lambda}_\alpha d\tau + \Delta \Psi_{in} - \Delta \Psi_f \right\} \right\rangle_q = 1, \quad (47)
$$

where  $\Delta \Psi_{in} = \Psi_{in} - \Psi_{st}(\lambda_0) = -\ln[\rho_{in}/\rho_{st}(\lambda_0)]$  and  $\Delta \Psi_f = \Psi_f$  $-\Psi_{st}(\lambda_t) = -\ln[\rho_f/\rho_{st}(\lambda_t)]$  and the index *q* indicates using the deformed FP operator  $\mathcal{H}_a$ .

The relation  $(47)$  $(47)$  $(47)$  involves the flux parameter,  $q$ , and the two probability distributions,  $\rho_f$  and  $\rho_{in}$ . In what follows we concentrate on the important case of nondeformed effective potential, where the flux parameter *q* is zero and  $p=2J_{st}$ . On substituting  $\rho_{\text{in}} = \rho_{\text{st}}(\lambda_0)$  and  $\rho_f = \rho_{\text{st}}(\lambda_t)$  into the identity [Eq. ([47](#page-6-0))], we recover the result obtained by Hatano and Sasa in Ref.  $[50]$  $[50]$  $[50]$ .

$$
\left\langle \exp\left\{-\int_0^t \frac{\partial \Psi_{st}}{\partial \lambda_\alpha} \dot{\lambda}_\alpha d\tau \right\} \right\rangle = 1, \tag{48}
$$

<span id="page-6-3"></span>which is just the special case of the relation  $(47)$  $(47)$  $(47)$  with  $q=0$ and  $\Delta \Psi_{\text{in.f}} = 0$ .

When the stationary flux vanishes and  $J_{st}=0$ , the steady-state distribution is given in Eq. ([18](#page-3-4)) with  $\Psi_{st} = V - F_{st}$ , so that the Hatano-Sasa formula  $(48)$  $(48)$  $(48)$  can be rewritten as the Jarzynski's equality  $|51|$  $|51|$  $|51|$ 

<span id="page-6-4"></span>
$$
\left\langle \exp\left\{-\int_0^t \frac{\partial V}{\partial \lambda_\alpha} \dot{\lambda}_\alpha d\tau \right\} \right\rangle = \exp\{-\Delta F_{\rm st}\},\qquad(49)
$$

where  $\Delta F_{\rm st} = F_{\rm st}(\lambda_t) - F_{\rm st}(\lambda_0)$ . Since the integral

$$
\int_{0}^{t} \frac{\partial V}{\partial \lambda_{\alpha}} \dot{\lambda}_{\alpha} d\tau = \Delta V - \int_{0}^{t} V' \dot{\varepsilon} d\tau = \Delta V + \Delta s_{\rm m}
$$
 (50)

can be expressed in terms of the entropy of the medium Eq.  $(31)$  $(31)$  $(31)$ , at  $J_{st}$ =0, the relation  $(47)$  $(47)$  $(47)$  takes the following form:

$$
\langle \exp\{-\Delta s_{\rm m} - (\Psi_{\rm f} - \Psi_{\rm in})\}\rangle = 1\tag{51}
$$

that reduces to the integral fluctuation theorem for the total change in entropy  $\left[30,57,58\right]$  $\left[30,57,58\right]$  $\left[30,57,58\right]$  $\left[30,57,58\right]$  $\left[30,57,58\right]$ 

$$
\langle \exp\{-\Delta s_{\text{tot}}\} \rangle = 1 \tag{52}
$$

<span id="page-7-1"></span>when  $|\rho_f\rangle = \hat{\mathcal{U}}(t,0)|\rho_{in}\rangle$  and  $\Delta s = \Psi_f - \Psi_{in}$ . From Eq. ([52](#page-7-1)) combined with the Jensen's inequality  $\langle \exp(x) \rangle \geq \exp(x)$ , the averaged change in the total entropy cannot be negative, so that, in agreement with Eq.  $(33)$  $(33)$  $(33)$  and the second law of thermodynamics,  $\langle \Delta s_{\text{tot}} \rangle \ge 0$ .

So, we have found that the well-known results such as the Hatano-Sasa identity [Eq. ([48](#page-6-3))], the Jarzynski's equality [Eq.  $(49)$  $(49)$  $(49)$ ] and the fluctuation theorem for the total entropy [Eq. ([52](#page-7-1))] immediately follow from our generalized fluctuation relation  $(47)$  $(47)$  $(47)$  derived for nonzero flux parameters.

Our concluding remark concerns the generalized fluctuation-exchange theorem for the steady-state systems formulated in a very recent paper  $[59]$  $[59]$  $[59]$ . This theorem is essentially a direct consequence of the Hatano-Sasa identity  $[Eq. (48)]$  $[Eq. (48)]$  $[Eq. (48)]$  applied to the limiting case of small perturbations, where  $\lambda(t) = \lambda_0 + \delta \lambda(t)$ . More specifically, it asserts that the response functions

$$
\frac{\partial \langle \Psi_{\alpha}(t) \rangle}{\partial \lambda_{\beta}(\tau)} = R_{\alpha\beta}(t - \tau)
$$
\n(53)

<span id="page-7-2"></span>of the averages

$$
\langle \Psi_{\alpha}(t) \rangle = \langle 1 | \Psi_{\alpha} \hat{\mathcal{U}}(t,0) | \rho_0 \rangle, \quad \Psi_{\alpha} \equiv \left. \frac{\partial \Psi_{\text{st}}}{\partial \lambda_{\alpha}} \right|_{\lambda = \lambda_0},
$$

$$
\rho_0 \equiv \rho_{\rm st}|_{\lambda = \lambda_0} \tag{54}
$$

meet the fluctuation-exchange relation

$$
R_{\alpha\beta}(t-\tau) = \partial_t C_{\alpha\beta}(t-\tau),\tag{55}
$$

<span id="page-7-3"></span>where  $C_{\alpha\beta}(t-\tau)$  is the correlation function given by

$$
C_{\alpha\beta}(t-\tau) = \langle \Psi_{\alpha}(t) \Psi_{\beta}(\tau) \rangle_0
$$
  
=  $\langle 1 | \Psi_{\alpha} \exp[-(t-\tau) \hat{\mathcal{H}}(\lambda_0)] \Psi_{\beta} | \rho_0 \rangle.$  (56)

From the above considerations, the formulas  $(53)$  $(53)$  $(53)$ – $(56)$  $(56)$  $(56)$  written down explicitly in the bracket notations are applicable to the Brownian systems in the energy representation. We shall extend on the subject elsewhere.

### **V. DISCUSSION AND CONCLUSIONS**

<span id="page-7-0"></span>In this paper, we have demonstrated that the method that combines the energy representation with the Langevin dynamics approach may provide an effective tool to explore the steady states of certain nonequilibrium Brownian systems.

These states can be analytically studied as the stationary solutions of the corresponding FP equation expressed in terms of the exchange and diffusion functions,  $f(\varepsilon)$  and  $g(\varepsilon)$ .

The function of energy exchange rate, *f*, defined as the deterministic part of the Langevin Eq. ([7](#page-2-0)) describes the process of direct energy interchange between the system and the environment, whereas the multiplicative white noise characterized by the diffusion function *g* represent the fluctuation induced effects. The latter may depend on the energy of the system and hence the diffusion function is generally not a constant.

We have shown that, in the limit of low noise with *g*  $\rightarrow$ 0, the steady states are determined by equilibria of the effective potential,  $\tilde{E}$ , defined in Eq. ([22](#page-3-8)). At  $g = g_0 \neq 0$ , these equilibria correspond to the maxima of the steady state (stationary) distribution,  $\rho_{st}$  Eq. ([18](#page-3-4)).

In the well-known case of linear exchange function, *f*  $=\gamma(\varepsilon-\varepsilon_0)$ , the system losses its energy via the processes of dissipation when the friction coefficient is positive  $\gamma > 0$ . Then the energy  $\varepsilon_0$  is the only equilibrium point of the harmonic potential,  $\tilde{E} = \gamma (\varepsilon - \varepsilon_0)^2 / 2$ , and, at energy independent diffusion function  $g = g_0$ , the steady-state distribution is of the Gaussian form:  $\rho_{st} = N \exp[-\gamma(\varepsilon - \varepsilon_0)^2/(2D_0)].$ 

In the opposite case of negative friction coefficient with  $\gamma$ <0, the system absorbs the energy from the environment and there are no steady states without mechanisms limiting the gain of energy. Interestingly, we have found that, when the friction coefficient depends on the energy and contains the random contribution coming from the environment stochasticity, the steady-state distribution may take the form of the standard Maxwell equilibrium distribution for Brownian particles [the conditions can be found below Eq.  $(24)$  $(24)$  $(24)$ ].

So, using our approach it is relatively easy to conclude on stationary distribution functions of nonequilibrium Brownian systems for different mechanisms of energy interchange complicated by the stochastic nonlinear coupling with the environment. In particular, it is not difficult to recover the results on statistics of energy states obtained in Ref.  $[16]$  $[16]$  $[16]$  for a generic model of a random polypeptide chain that reproduces the energy probability distribution of real proteins over a very large range of energies. It is also pertinent to note that the systems such as dust particles in plasma  $\lceil 60 \rceil$  $\lceil 60 \rceil$  $\lceil 60 \rceil$  and Brownian particles with the randomly inhomogeneous friction coefficient present important cases for which theoretical predictions can be experimentally verified, but this task requires additional analysis and experimental data.

As first steps in this direction, we have introduced the trajectory-dependent entropy in the energy representation so as to define both the entropy of the Brownian system and the entropy production in the environment. The latter, similar to the steady-state distributions at  $J_{st}=0$ , was found to be determined by the effective energy potential  $[Eq. (12)].$  $[Eq. (12)].$  $[Eq. (12)].$ 

As far as the fluctuation relations are concerned, our key finding is the generalized integral fluctuation theorem [Eq. ([47](#page-6-0))] describing nonequilibrium Brownian systems with the shifted effective potential characterized by the flux parameter,  $q$ , and the flux number,  $J_{st}$  (the stationary value of the probability current). It turned out that the fluctuation theorem for the total entropy change  $[Eq. (52)]$  $[Eq. (52)]$  $[Eq. (52)]$  can be deduced from

this theorem. The Hatano-Sasa identity  $[Eq. (48)]$  $[Eq. (48)]$  $[Eq. (48)]$  and the Jarzynski's equality  $[Eq. (49)]$  $[Eq. (49)]$  $[Eq. (49)]$  can equally be derived as the special cases of our fluctuation relation  $(47)$  $(47)$  $(47)$ .

At  $p = J<sub>st</sub> = 0$ , the algebraic identity [Eq. ([41](#page-6-2))] reduces to the detailed balance relation

$$
\hat{\mathcal{H}}^{\dagger} = e^{\Psi_{\rm st}} \hat{\mathcal{H}} e^{-\Psi_{\rm st}} \tag{57}
$$

for the steady-state distribution of the potential form Eq.  $(18)$  $(18)$  $(18)$ ]. In the opposite case characterized by nonzero flux number,  $J_{st} \neq 0$ , and nonreflecting boundary conditions, the identity  $[Eq. (41)]$  $[Eq. (41)]$  $[Eq. (41)]$  describes the systems where detailed balance is violated. Alternatively, in multidimensional systems, explicit violation of detailed balance can be caused by nonconservative forces  $\lceil 61 \rceil$  $\lceil 61 \rceil$  $\lceil 61 \rceil$ .

From the integral theorem  $[Eq. (47)]$  $[Eq. (47)]$  $[Eq. (47)]$ , and, following the line of reasoning presented in Ref.  $[62]$  $[62]$  $[62]$ , it can be concluded that the energy representation serves as a coarse-grained description of stochastic systems where the energy can be viewed as the reduced variable such that information on its trajectories is enough to reproduce the statistics of the entropy production.

Our concluding remark concerns the detailed theorem that underlies the integral fluctuation relation  $(47)$  $(47)$  $(47)$ . According to the general results of the very recent paper  $\lceil 63 \rceil$  $\lceil 63 \rceil$  $\lceil 63 \rceil$ , when a variable obeys an integral fluctuation theorem, it automatically obeys a detailed theorem. So, the relation  $(47)$  $(47)$  $(47)$  additionally indicates that, in the energy representation, there are detailed fluctuation theorems describing Brownian-like systems in the absence of detailed balance. These theorems and related analysis are beyond the scope of this paper and will be discussed elsewhere.

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