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Fluctuation relations for diffusion that is thermally driven by a nonstationary bath

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In the context of the Markovian dynamical evolution in a nonstationary thermal bath, we construct a family of fluctuation relations for the entropy production that are not verified by the work performed on the system. We exhibit fluctuation relations, which are global versions either of the generalized fluctuation-dissipation theorem around a nonequilibrium diffusion or of the usual fluctuation-dissipation theorem for energy resulting from a pulse of temperature.

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I. INTRODUCTION

One important recent progress in nonequilibrium statistical physics is the discovery of various fluctuation relations which can be viewed as nonperturbative extensions of the usual fluctuation-dissipation theorem (FDT) [1,2]. Such relations pertain either to nonstationary transient situations [3,4] or to stationary regimes [5]. In particular, a family of fluctuation relations holds for the distribution of work performed on a system [4,6–8] which evolves in an equilibrium bath, and another family of relations holds for the entropy creation [7–10]. These developments had an important impact on the physics of nanosystems and biomolecules [11]. Here, we examine the question of the extension of such relations to an evolution in a nonequilibrium medium. More precisely, we consider systems placed in the thermal bath that is nonstationary, a situation which can be realized experimentally by adding or extracting heat, or by modulating the pressure. Let us remark that nonstationarity of the temperature is linked via Stokes law to nonstationarity of friction and that particles with time-dependent radii have nonconstant mass and friction. This last situation is an important problem in astrophysics for the formation of planets through dust aggregation [12,13]. There is another situation where the nonstationarity of friction can be realized with particles diffusing in ferroelectric fluids when external magnetic fields are controlling the intrinsic viscosity [14].

A famous example of a system that evolves in a nonstationary bath is the temperature ratchet model [15] of a Brownian motor. We can also consider the evolution of a system initially in equilibrium at high temperature T_i [i.e., in the state with the density $\rho_i = \exp(-H/T_i)$] put in contact with a nonstationary bath reaching a low temperature T_f. The function $T(t)$ is then called the cooling schedule. A particularly interesting case is the instantaneous quench of the initial system in a thermostat at temperature T_f, which corresponds to the cooling schedule with instantaneous initial change in temperature. The age (or waiting time) of the system is then the time elapsed since the quench. Finally, at the more formal level, such nonstationary evolution rules appear naturally from a stationary dynamics after system size or coarse-grained expansion [16]. Diffusion properties in a nonstationary medium have been recently studied in the case of unidimensional overdamped dynamics driven by multiplicative nonstationary noise [17,18], in the case of nonstationary colored bath [19–21], and in the case of local quasiequilibrium of slow relaxation systems [22,23].

The present paper consists of five sections. Section II sets the stage and notations for the model of nonequilibrium and nonlinear Langevin dynamics that we consider. In Sec. III, we recall the main result of [8] and we list different time inversions that permit us to obtain fluctuation relations for entropy production. We observe that in this context the work performed on the system no longer obeys the fluctuation relations but that there still exists a functional that upon averaging gives the free-energy change in a nonequilibrium protocol. In Sec. IV we explicitly construct fluctuation-dissipation relations that result from the Taylor expansion of the last fluctuation relation. In particular, we recover the generalized FDT [24] around a nonequilibrium diffusion and the usual FDT for the energy which results from a pulse of temperature [25]. We discuss also the physical meaning of the effective temperature of a harmonic oscillator in a nonstationary bath. Finally, Sec. V presents our conclusions.

II. THERMALLY DRIVEN NONLINEAR LANGEVIN DYNAMICS

The general dynamics that we consider is described by a nonautonomous diffusion process that we call Langevin dynamics (or nonlinear Brownian motion) in a d-dimensional (phase) space E. The process obeys the stochastic differential equation (SDE)

$$\dot{x}^i = -\Gamma_t^{ij}(x)\partial_j H_t(x) + \Pi_t^{ij}(x)\partial_j H_t(x) + G_t^i(x) + w_t^i(x) + \eta_t^i(x), \tag{1}$$

which should be interpreted in the Stratonovich convention. Here, $H_t(x)$ is the Hamiltonian of the system (the time indexation corresponds to an explicit time dependence), $\Gamma_t(x)$ is a family of non-negative matrices, $\Pi_t(x)$ is a family of antisymmetric matrices, $G_t(x)$ is an external force (or a shear), and $w_t(x)$ is an additional corrective term which will be explicit in Eq. (3) below and which comes from the x dependence of Γ_t and Π_t. Finally, $\eta_t(x)$ is a white (i.e., mimics a bath with very short time correlations or memory) Gaussian vector field with mean zero and covariance

$$\langle \eta^i_t(x)\,\eta^j_s(y)\rangle = 2D^{ij}_t(x,y)\,\delta(t-s) \quad \text{with } D_t(x,x) \equiv d_t(x) = \frac{1}{\beta_t}\Gamma_t(x). \tag{2}$$

The last equality, which contains the temperature of the bath $T_t = 1/\beta_t$, is called the Einstein relation. In the case when the bath is in equilibrium, this equation reflects the fact that the friction and the noise are two dual effects of the interaction with the bath. In this nonstationary setup, the Einstein relation is valid under the assumption of local thermal equilibrium, provided the thermalization of the immediate environment of the Brownian particle is much faster than the characteristic variation of temperature. Let us underline that in this context the term "nonlinear" concerns the nonhomogeneous properties of $\Gamma_t(x)$ and $\Pi_t(x)$ (which may characterize nonhomogeneous properties of the bath) and not the fact that Eq. (1) is nonlinear. Such nonlinear properties appear naturally in many situations: nonideal plasmas and gases [26,27], ultracold clusters of atoms, or molecules cooled by interaction with laser radiation active Brownian particles [28]. Finally, the additional corrective term in Eq. (1) is given by the expression

$$w^i_t(x) = \partial_{y^j} D^{ij}_t(x,y)\big|_{x=y} - \frac{1}{\beta_t}\partial_j \Pi^{ij}_t(x). \tag{3}$$

The presence of this term assures that in the case with stationary Hamiltonian and temperature (i.e., $H_t = H$, $\beta_t = \beta$) and without external force (i.e., $G = 0$), the Gibbs density $\exp(-\beta H)$ is an invariant density.[1] The presence of this term $w_t(x)$ in Eq. (1) can appear as a makeshift arrangement, but it was extensively studied in the literature of nonlinear Brownian motion [26,27]. Note that w_t vanishes in the case of linear Brownian motion where $D_t(x,y) = D_t$ and $\Pi_t(x) = \Pi_t$. We call the deterministic part of the second member of Eq. (1),

$$u^i_t(x) = -\Gamma^{ij}_t(x)\partial_j H_t(x) + \Pi^{ij}_t(x)\partial_j H_t(x) + G^i_t(x) + w^i_t(x), \tag{4}$$

the drift term. An elementary case of nonlinear Brownian motion is the Landau-Lifshitz-Bloch dynamics of a Brownian spin [29] in an effective magnetic field $B^{eff}_t(x) = -\nabla H_t$ (which can incorporate interaction with other spins). It follows the dynamics

$$\dot{x} = -x \times \nabla H_t + \lambda_t x \times (x \times \nabla H_t) + G_t(x) + x \times \zeta_t \quad \text{with } \langle \zeta^i_t \zeta^j_s\rangle = \frac{2\lambda_t}{\beta_t}\delta^{ij}\delta(t-s). \tag{5}$$

The first term on the right-hand side is the precession term, the second one is the damping term, and the third one is an external torque. The noise (and the damping term) accounts for the effect of the interaction with the microscopic degrees of freedom (phonons, conducting electrons, nuclear spins, etc.). This dynamics is a particular case of Eq. (1) with $\Gamma^{ij}_t(x) = \lambda_t(\delta^{ij}x^2 - x^i x^j)$ and $\Pi^{ij}_t(x) = \varepsilon^{ijk}x^k$ with ε^{ijk} as the totally antisymmetric tensor. In this example one sees the need for the term $\Pi^{ij}_t(x)\partial_j H_t(x)$ in Eq. (1) corresponding to the Hamiltonian vector field which also permits us to describe systems of nonoverdamped Brownian particles in the phase space with coordinates $\binom{q}{p}$, with nonstationary mass, in an external potential V_t, subjected to a nonconservative force f_t, and in a nonstationary bath giving rise to the noise and the nonhomogeneous nonlinear but isotropic drag $\gamma_t(q,p)$. The Stratonovich SDE that governs this model is then

$$\dot{q} = \frac{p}{m_t},$$

$$\dot{p} = -\frac{\gamma_t(q,p)}{m_t}p - \nabla V_t(q) + f_t(q) + \frac{\nabla_p \gamma_t(q,p)}{2\beta_t} + \sqrt{\frac{2\gamma_t}{\beta_t}}\eta, \quad \text{with } \langle \eta^i_t \eta^j_s\rangle = \delta^{ij}\delta(t-s), \tag{6}$$

which is again a subcase of Eq. (1) with

$$\Gamma_t = \begin{pmatrix} 0 & 0 \\ 0 & \gamma_t \end{pmatrix}, \quad \Pi = \begin{pmatrix} 0 & Id \\ -Id & 0 \end{pmatrix}, \quad H_t = \frac{p^2}{2m_t} + V_t(q), \quad G_t = \begin{pmatrix} 0 \\ f_t(q) \end{pmatrix}.$$

The Kramers case corresponds to the Stokes law of friction $\gamma_t(q,p) = \gamma_t(q)$. Another example of friction with $\gamma_t(q,p) = \gamma(p^2 - p_0^2)$ appears in the Rayleigh-Helmholtz theory of sound [30].

We start by collecting the elementary properties of diffusion processes that we shall need [31]. The Markovian generator L_t of the process x_t satisfying the SDE (1) is defined by the relation

$$\partial_t\langle f(x_t)\rangle = \langle (L_t f)(x_t)\rangle, \quad L_t = \hat{u}_t \cdot \nabla + \nabla \cdot \frac{\Gamma_t}{\beta_t}\nabla, \tag{7}$$

where the modified drift $\hat{u}_t$ is defined in terms of the drift (4),

$$\hat{u}^i_t(x) = u^i_t(x) - \partial_{y^j} D^{ij}_t(x,y)\big|_{x=y}. \tag{8}$$

The time evolution of the instantaneous probability density function of the process $\rho_t(x) = \langle \delta(x_t - x)\rangle$ is governed by the formal adjoint $L^\dagger_t$ of the generator L_t,

$$\partial_t \rho_t = L^\dagger_t \rho_t, \tag{9}$$

which can be rewritten as a continuity equation (respectively, a hydrodynamic advection equation) by defining the probability current $\tilde{j}_t$ (respectively, the mean local velocity $\tilde{v}_t$),

$$\partial_t \rho_t = -\nabla \cdot \tilde{j}_t = -\nabla \cdot (\rho_t \tilde{v}_t),$$

$$\times \tilde{j}^i_t \equiv (\hat{u}^i_t - \beta_t^{-1}\Gamma^{ij}_t \nabla_j)\rho_t, \quad \tilde{v}^i_t \equiv \frac{\tilde{j}^i_t}{\rho_t}. \tag{10}$$

As was explained in [24], it is convenient to use the freedom to add a divergenceless term in the definition of the probability current to obtain the modified current and the modified local velocity

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[1] In fact, the Gibbs density is then an equilibrium density: an invariant density with vanishing modified probability current [24].

$$j_t^i \equiv \widetilde{\dot{j}}_t^i + \beta_t^{-1}\nabla_j(\Pi^{ij}\rho_t), \quad v_t^i \equiv \frac{j_t^i}{\rho_t}, \tag{11}$$

which verify also the continuity equation (10) but vanish in the case of stationary Hamiltonian and temperature (i.e., $H_t = H$, $\beta_t = \beta$) for vanishing external force $G_t = 0$.

III. FLUCTUATION RELATIONS AND TIME INVERSION

In [8], various fluctuation relations were discussed for arbitrary diffusion processes. We recall here the main result in the context of systems with dynamics of type (1) (see also Sec. 3 of [32]). With the use of combined Girsanov and Feynman-Kac formulas, one obtains the detailed fluctuation relation (DFR)

$$\mu_0(dx)P_T(x;dy,dW)e^{-W} = \mu_0^r(dy^*)P_T^r(y^*;dx^*,d[-W]), \tag{12}$$

where

(1) $\mu_0(dx) = \varrho_0(x)dx$ is the initial distribution of the original forward process (1),

(2) $\mu_0^r(dx) = \varrho_0^r(x)dx$ is the initial distribution of the backward process obtained from the forward process by applying a time inversion (see below),

(3) $P_T(x;dy,dW)$ is the joint probability distribution of the time T and position x_T, of the forward process starting at time zero at x, and of a functional W_T (linked to the entropy production) of the same process on the interval $[0,T]$, (described later), and

(4) $P_T^r(x;dy,dW)$ is the similar joint probability distribution for the backward process.

The time inversion acts on time and space by an involution

$$(t,x) \mapsto (t^* = T - t, x^*). \tag{13}$$

Such an involution induces the action $x \mapsto \tilde{x}$ on trajectories by the formula $\tilde{x}_t = x^*_{T-t}$ and, further, the action on functionals of trajectories $F \mapsto \tilde{F}$, by setting $\tilde{F}[x] = F[\tilde{x}]$.

To recover various fluctuation relations discussed in the literature [4,6,9,33–35] into two parts, $u = u_+ + u_-$, with u_+ transforming as a vector field under the space-time involution (13) and u_- as a pseudo-vector-field,

$$u^{ri}_{T-t,\pm}(x^*) = \pm(\partial_k x^{*i})(x)u^k_{t,\pm}(x), \quad u^r = u^r_+ + u^r_-. \tag{14}$$

The random field $\eta_t(x)$ may be transformed with either rule. By definition, the backward process satisfies then the Stratonovich SDE

$$\dot{x} = u_t^r(x) + \eta_t^r(x). \tag{15}$$

Let us stress that, in general, this backward process differs from the naive time inversion $y_t = \tilde{x}_t$ of the forward process (called natural time inversion in [8]) which is the case where u_+ vanishes and which satisfies, for example, when $x^* = x$ the Stratonovich SDE

$$\dot{y} = -u_{t^*}(y) \pm \eta_{t^*}(y). \tag{16}$$

The requirement of a nonvanishing vector part u_+ in general can easily be seen by considering that a dissipative term [like $-\Gamma_t^{ij}(x)\partial_j H_t(x)$] in the forward process becomes antidissipative in process (16), while the backward process is still in contact with the bath. The functional W_T is given by the expression

$$W_T = \ln\varrho_0(x_0) - \ln[\varrho_0^r(x_T^*)\sigma(x_T)] + \int_0^T J_t dt, \tag{17}$$

where $\sigma(x) = |\det(\partial x^*/\partial x)|$ is the Jacobian of the spatial involution. The intensive functional J_t has the interpretation of the rate of entropy production in the environment and is given by the expression

$$J_t = \beta_t \hat{u}_{t,+}(x_t)\cdot\Gamma_t^{-1}(x_t)[\dot{x}_t - u_{t,-}(x_t)] - (\nabla\cdot u_{t,-})(x_t). \tag{18}$$

The time integral in Eq. (17) is taken in the Stratonovich sense. When $\mu_0^r(dx^*) = \mu_T(dx)$ then the boundary contribution $\{\ln\varrho_0(x_0) - \ln[\varrho_0^r(x_T^*)\sigma(x_T)]\}$ to W_T gives the change in the instantaneous entropy of the process. In this case, the functional W_T becomes equal to the overall entropy production. Moreover, with the interpretation of J_t as the entropy production in the environment, the first principles give us that the work $\mathcal{T}_T$ performed on the system can be expressed in terms of J_t,

$$\mathcal{T}_T = H_T(x_T) - H_0(x_0) + \int_0^T \frac{J_t}{\beta_t}dt. \tag{19}$$

We can underline that, in this setup, and contrary to the case of a stationary bath [8], the work $\mathcal{T}_T$ cannot be identified with the functional W_T for an appropriate choice of initial densities of the forward and backward processes. This means that the work does not verify the DFR.

The DFR (12) holds even if the measures μ_0 and μ_0^r are not normalized, or even not normalizable. When they are normalized, let us denote by $\langle - \rangle$ and by $\langle - \rangle^r$ the expectations of functionals of, respectively, the forward and the backward processes on the time interval $[0,T]$, with initial distributions μ_0 and μ_0^r. One of the immediate consequences of the DFR equation (12) is the (generalized) Jarzynski equality [4]

$$\langle e^{-W_T}\rangle = 1 \tag{20}$$

obtained by the integration of the both sides of Eq. (12). It implies the inequality $\langle W_T\rangle \geq 0$ that has the form of the second law of thermodynamics stating the positivity of the average entropy production. With a little more work [8], the DFR (12) may be cast into a form of the (generalized) Crooks relation [6]

$$\langle F\exp(-W_T)\rangle = \langle\tilde{F}\rangle^r. \tag{21}$$

We will now restrict ourselves to the class of time inversions (14) such that there exists a nonstationary density f_t such that

$$\hat{u}_{+,t} = \beta_t^{-1}\Gamma_t\nabla\ \ln f_t \quad \text{and then} \quad u_{-,t} = -\Gamma_t\nabla(H_t + \beta_t^{-1}\ln f_t)$$
$$+\Pi_t\nabla H_t - \frac{1}{\beta_t}\nabla\cdot\Pi_t^T + G_t. \tag{22}$$

After a straightforward calculation, the rate of entropy production in the environment may be expressed as

$$J_t = \dot{x}_t \cdot (\nabla \ln f_t)(x_t) + [(f_t)^{-1} L_t^{\dagger} f_t](x_t). \tag{23}$$

With the choice $\mu_0(dx) = f_0(x)dx$ and $\mu_0^r(dx^*) = f_T(x)dx$ the functional W_T takes then the simple form

$$W_T = \int_0^T [(f_t)^{-1} L_t^{\dagger} f_t - \partial_t \ln(f_t)](x_t)dt. \tag{24}$$

We can then remark that the class of time inversions (14) is the most general which allows the functional W_T to become a functional of just x_t and not of the derivative $\dot{x}_t$. We shall see that, due to this independence of $\dot{x}_t$, the fluctuation relations associated with this peculiar family of inversions are the natural generalizations of the FDT. We shall now describe particular cases in this family of time inversions.

A. Complete reversal

As the function f_t in Eq. (22) we take the instantaneous density function (i.e., $f_t = \rho_t$) of the forward process (1) distributed with initial condition f_0 [8]. Here, the functional (24) trivially vanishes $W_T = 0$ and the DFR (12) takes the form of the generalized detailed balance

$$\mu_0(dx) P_T(x;dy) = \mu_T(dy) P_T^r(y^*;dx^*). \tag{25}$$

One may show that $\rho_t^r(x) \equiv \rho_{t^*}(x^*)$ is the instantaneous density of the backward process and that the corresponding probability current satisfies the relation

$$\widetilde{j_t^{i,r}}(x) = -(\partial_k x^{*i})(x) \widetilde{j_{t^*}^{k}}(x^*). \tag{26}$$

This inversion is employed in many articles in probability theory [36–40]. It corresponds to the vanishing overall entropy production.

B. Current reversal

Another useful choice of time inversion, called the current reversal [8,35], is based on the choice $f_t = \pi_t$, where π_t satisfies $L_t^{\dagger} \pi_t = -\nabla \cdot \tilde{j}_t = 0$. In the case where $G_t = 0$, we have $\pi_t = \exp[-\beta_t(H_t - F_t)]$ with F_t as the free energy [i.e., $\exp(-\beta_t F_t) = \int \exp(-\beta_t H_t)$]. One can show [8] that $\pi_t^r(x) \equiv \pi_{t^*}(x^*)$ is the density for the backward process that corresponds to the conserved current $\nabla \cdot \widetilde{j_t^r} = 0$ and that Eq. (26) still holds. The functional (24) takes now the form

$$W_T^{ex} = -\int_0^T (\partial_t \ln \pi_t)(x_t)dt, \tag{27}$$

where the index "*ex*" stands for "excess" [7,8]. For the backward process, the functional $W_T^{ex,r}$ is given by the same expression with π_t replaced with π_t^r. The Jarzynski equality (20) for this case was first proven in one dimension in [41] and in the general case in [8,42,43].

C. Canonical inversion

A natural choice for systems (22) is to take f_t to be the Gibbs density $\exp[-\beta_t(H_t - F_t)]$, where F_t is the free energy [i.e., $\exp(-\beta_t F_t) = \int \exp(-\beta_t H_t)$] if the Gibbs density is normalizable and zero otherwise [8]. This corresponds to the choice $\hat{u}_{+,t} = -\Gamma_t \nabla H_t$ in Eq. (22). The functional (24) becomes

$$W_T^{ci} = -(\beta_T F_T - \beta_0 F_0) + \int_0^T [\partial_t(\beta_t H_t) + \beta_t G_t \cdot \nabla H_t - \nabla \cdot G_t] \times (x_t)dt, \tag{28}$$

where the index "*ci*" means "canonical inversion." The generalized Jarzynski equality (20) can be rewritten in the form

$$\left\langle \exp\left(-\int_0^T [\partial_t(\beta_t H_t) + \beta_t G_t \cdot \nabla H_t - \nabla \cdot G_t](x_t)dt \right) \right\rangle = \exp[-(\beta_T F_T - \beta_0 F_0)], \tag{29}$$

which permits us to extract the difference of free energy out of a nonequilibrium experiment in a nonstationary bath (but the connection with the work performed is lost). For example, for the Brownian particle (6), this functional takes the form

$$W_T^{ci} = -(\beta_T F_T - \beta_0 F_0) + \int_0^T \left[\dot{\beta}_t \left(\frac{p_t^2}{2m_t} + V_t(q_t) \right) + \beta_t \left(-\frac{p_t \dot{m}_t}{m_t^2} + (\partial_t V_t)(q_t) + (f_t \cdot \nabla V_t)(q_t) \right) \right] dt \tag{30}$$

which, as compared to the functional which appears in the usual Jarzynski equality [4,33], contains terms proportional to the variation of temperature $\partial_t \beta_t$ and of mass $\partial_t m_t$. From the Jarzynski equality (20), one can deduce the inequality which constrains the evolution in a nonstationary bath,

$$\langle W_T^{ci} \rangle \geq 0.$$

In the case with a stationary Hamiltonian (i.e., $H_t = H$) and without external force (i.e., $G_t = 0$) this constraint reads

$$\int_0^T (\partial_t \beta_t) \langle H(x_t) \rangle dt \geq (\beta_T F_T - \beta_i F_i).$$

Let us consider now the overdamped particle in a three-dimensional time-dependent harmonic potential. Such an example admits an analytical computation of the distribution of the functional W_T^{ci}. We consider a nonstationary bath (γ_t, β_t) and the harmonic potential $U_t(x) = (k_t/2)(x - a_t)^2$, where k_t is the stiffness coefficient and a_t is the instantaneous center of the potential. The particle is initially distributed with the Gibbs density $\rho_0(x) = \exp[-\beta_0(U_0(x) - F_0)] = (\frac{C}{2\pi})^{3/2} \exp[-\frac{C}{2}(x - a_0)^2]$. Further, we will restrict our study to the particular case, not necessarily physical, where the temperature of the bath and the stiffness coefficient are such that their product is stationary: $k_t \beta_t = C$. This setup generalizes the unidimensional stationary case ($k_t = k$, $\beta_t = \beta$, $\gamma_t = \gamma$, and $a_t = ut$) considered in [44,45]. The system satisfies the linear SDE

$$\dot{x} = -\frac{k_t}{\gamma_t}(x - a_t) + \eta_t \quad \text{with} \quad \langle \eta_s^i \eta_t^j \rangle = \frac{2}{\beta_t \gamma_t} \delta(t - s) \delta^{ij}. \tag{31}$$

The functional $W_T^{ci} = W_T^{ex}$ takes here the form

$$W_T^{ci} = -C\int_0^T \dot{a}_t \cdot (x_t - a_t).$$

The distribution of W_T^{ci} for a process with the initial Gaussian density ρ_0 is Gaussian due to the linearity of Eq. (31). A straightforward calculation gives the mean

$$\langle W_T^{ci}\rangle = C\int_0^T dt \int_0^t \dot{a}_t \cdot \dot{a}_s \exp\left(-\int_s^t \frac{k_u}{\gamma_u} du\right) ds \tag{32}$$

and the variance of this Gaussian (we assume that the integrals exist)

$$V_T \equiv \langle (W_T^{ci} - \langle W_T^{ci}\rangle)^2\rangle = 2C\int_0^T dt \int_0^t \dot{a}_t \cdot \dot{a}_s \times\exp\left(-\int_s^t \frac{k_u}{\gamma_u} du\right) ds. \tag{33}$$

The distribution of W_T^{ci} is then

$$P^T(W) = \frac{1}{\sqrt{2\pi V_T}} \exp\left(-\frac{(W - \langle W_T^{ci}\rangle)^2}{2V_T}\right), \tag{34}$$

and an elementary calculus shows that the Jarzynski equality (20) is equivalent to the fact that $V_T = 2\langle W_T^{ci}\rangle$, which is evident from the comparison of Eqs. (32) and (33).

D. Alternative inversion

We choose for the function f_t the mean instantaneous density ρ_t' of another Langevin dynamics (1) which possesses the same parameters Γ_t, Π_t, G_t but with another nonautonomous Hamiltonian H_t' and another bath temperature β_t'. We note L_t (respectively, L_t') the Markovian generators of the process with the Hamiltonian H_t and the bath temperature β_t (respectively, H_t' and β_t'). The functional (24) takes now the form

$$W_T = \int_0^T [(\rho_t')^{-1}(L_t - L_t')^\dagger \rho_t'](x_t) dt. \tag{35}$$

This alternative inversion will permit us to recover generalizations of the FDT around nonstationary nonequilibrium diffusions (see also [24,32,46]).

IV. GENERALIZATIONS OF THE FLUCTUATION-DISSIPATION THEOREM

As noted in [9,47], the fluctuation relations may be viewed as extensions to the nonperturbative regime of the Green-Kubo and Onsager relations for the nonequilibrium transport coefficients valid within the linear-response description of the vicinity of the equilibrium. Reference [8] contains a detailed argument showing that if in a stationary bath one perturbs an equilibrium system by introducing a weakly time-dependent Hamiltonian $H_t(x) = H(x) - g_{a,t} O^a(x)$ then the Jarzynski equality associated with Eq. (27) or Eq. (28) gives in the second order of the Taylor expansion in g the usual FDT. Reference [32] showed that similar correspondence still holds around nonequilibrium steady states for a stationary dynamics with an external force (i.e., $G \neq 0$). In this case, it is the Crooks relation (21) associated with the functional (27) that gives the modified fluctuation-dissipation theorem (MFDT) [32,48] after the first-order Taylor expansion in g. The second-order expansion of Jarzynski equality (20) associated with the functional (27) gives in such a situation only a special case of this theorem. We shall now investigate which type of fluctuation-dissipation identities may be deduced by Taylor expanding the fluctuation relation corresponding to the time inversion of Sec. III D.

A. FDT around nonstationary diffusions

We consider system (1) with the Hamiltonian $H_t(x) = H_t^0(x) - g_{a,t} O^a(x)$. Following Sec. III D, we choose f_t as the mean instantaneous density ρ_t^0 of the unperturbed system with $g = 0$. The functional (35) becomes

$$W_T = \int_0^T g_{a,s}[(\rho_s^0)^{-1} M_s^{a\dagger} \rho_s^0]_s ds \quad \text{with} \quad M_s^a = (\Gamma_s \nabla O^a - \Pi_s \nabla O^a) \cdot \nabla, \tag{36}$$

where the subscript "s" on $[(\rho_s^0)^{-1} M_s^{b\dagger} \rho_s^0]$ signals that the latter function should be taken at the point x_s. Let us now write a particular case of Crooks relation (21), where the average is in system (1) with the Hamiltonian H_t, associated with a single time functional $F[x] = O^a(x_t) \equiv O_t^a$ $(0 < t < T)$,

$$\langle O_t^a e^{-W_T}\rangle = \langle O_{T-t}^a\rangle^r. \tag{37}$$

We shall denote by $\langle\cdot\rangle_0$ the average of the process with the dynamics driven by H_t^0 and by L^0, $\widetilde{v^0}$ and v^0, respectively, its Markovian generator, its mean local velocity, and its modified mean local velocity. The first-order Taylor expansion

$$\exp(-W_T) = 1 + \int_0^T g_{b,s}[(\rho_s^0)^{-1} M_s^{b\dagger} \rho_s^0]_s ds + \mathcal{O}(g^2) \tag{38}$$

in Eq. (37) gives the relation

$$\langle O_t^a\rangle_0 + \int g_{b,s} \left.\frac{\delta}{\delta g_{b,s}}\right|_{g=0} \langle O_t^a\rangle ds - \int_0^T g_{b,s} \langle O_t^a (\rho_s^{0-1} M^{b\dagger} \rho_s^0)_s\rangle_0 ds + \mathcal{O}(g^2) = \langle O_{T-t}\rangle^r. \tag{39}$$

The right-hand side has a functional dependence only on $\{g_u, u > t\}$, so if we apply $(\delta/\delta g_{b,s})|_{g=0}$, for $0 < s \leq t$, to the last identity, we obtain

$$\left.\frac{\delta}{\delta g_{b,s}}\right|_{g=0} \langle O_t^a\rangle = \langle [(\rho_s^0)^{-1} M^{b\dagger} \rho_s^0]_s O_t^a\rangle_0. \tag{40}$$

A short calculation gives

$$\begin{aligned}(\rho_s^0)^{-1} M_s^{b\dagger} \rho_s^0 &= \beta_s \widetilde{v_s^0} \cdot \nabla O^b - \beta_s L_s^0 O_s^b + \Pi_s^{ij}(\partial_j O^b)\partial_i \ln(\rho_s^0) \\ &\quad + (\partial_i \Pi_s^{ij})\partial_j O^b = \beta_s(2\widetilde{v_s^0} \cdot \nabla - L_s^0) O^b \\ &\quad - \beta_s \widetilde{v_s^0} \cdot \nabla O^b + \Pi_s^{ij}(\partial_j O^b)\partial_i \ln(\rho_s) + (\partial_i \Pi_s^{ij})\partial_j O^b\end{aligned}$$

$$= \beta_s(2\widetilde{v_s^0} \cdot \nabla - L_s^0)O^b - \beta_s v_s^0 \cdot \nabla O^b. \tag{41}$$

Moreover, we have the sum rule (for $s \leq t$)

$$\partial_s \langle O_s^b O_t^a \rangle_0 = \langle [(2\widetilde{v_s^0}\, \nabla - L_s^0)O^b]_s O_t^a \rangle_0. \tag{42}$$

With Eqs. (40)–(42), we obtain the MFDT

$$\partial_s \langle O_s^b O_t^a \rangle_0 = \left.\frac{1}{\beta_s}\frac{\delta}{\delta g_{b,s}}\right|_{g=0} \langle O_t^a \rangle + \langle (v_s^0 \cdot \nabla O^b)_s O_t^a \rangle_0. \tag{43}$$

This is a generalization of the FDT around a nonequilibrium diffusion process in a stationary bath ($\beta_t = \beta$) of Refs. [24,32,46,49,50] and of FDT around nonstationary Langevin equation of Ref. [51]. It may be also proven as in [24] using the fact that the diffusion process becomes an equilibrium one in the Lagrangian frame of its modified mean local velocity v^0, verifying in that frame the usual FDT. The transformation of the latter back to the Eulerian (i.e., laboratory) frame leads to Eq. (43). Let us remark that, here, similarly as in the stationary case discussed in [32], the Jarzynski equality (20) for the functional (35) leads upon the second-order expansion in g to a particular case of the MFDT where the observable O_t^a is replaced with $A^a = (\rho_t^0)^{-1} M_t^{a\dagger} \rho_t^0$, which is a (time-dependent) functional of O^a.

The violation of the usual FDT can be parametrized by using Eq. (43) via the introduction of the so-called effective temperature [52,53] defined by

$$T^{eff}(s,t,O^a) \equiv \frac{\partial_s \langle O_s^a O_t^a \rangle_0}{\left.\frac{\delta}{\delta g_{a,s}}\right|_{g=0} \langle O_t^a \rangle} = \frac{1}{\beta_s} + \frac{\langle (v_s^0 \cdot \nabla O^a)_s O_t^a \rangle_0}{\left.\frac{\delta}{\delta g_{a,s}}\right|_{g=0} \langle O_t^a \rangle}. \tag{44}$$

We shall consider now the case where this effective temperature may be computed analytically in order to verify its physical consistency.

Unidimensional harmonic oscillator in a nonstationary bath. The SDE that governs this system is

$$\dot{x} = -\frac{k}{\gamma}x + \eta \quad \text{with } \langle \eta_t \eta_s \rangle = \frac{2T_t}{\gamma}\delta(t-s). \tag{45}$$

We take the cooling schedule T_t such that the bath passes from an initial temperature ($T_0 = T_i$) to a lower final temperature ($T_\tau = T_f < T_i$) during a time τ. The system is initially in equilibrium with the bath and its initial density is $\rho_0(x) = \exp[-(k/2T_i)x^2]/Z_i$. We consider two particular examples of cooling schedules:

(1) instantaneous quench

$$T_t = \begin{cases} T_i & \text{if} \quad t = 0 \\ T_f & \text{if} \quad t > 0, \end{cases} \tag{46}$$

(2) linear decrease in temperature

$$T_t = \begin{cases} T_i + \frac{t}{\tau}(T_f - T_i) & \text{if} \quad t \leq \tau \\ T_f & \text{if} \quad t \geq \tau. \end{cases} \tag{47}$$

Due to the linearity of Eq. (45), one can compute explicitly the response of the position to an external perturbation $V(x) = \frac{k}{2}x^2 \rightarrow V_t'(x) = V(x) - g_t x$ with $g_0 = 0$,

$$\left.\frac{\delta}{\delta g_s}\right|_{g=0} \langle x_t \rangle = \frac{1}{\gamma}\exp\left(-\frac{k}{\gamma}(t-s)\right). \tag{48}$$

It has a stationary form and is independent on the cooling schedule. In a similar way, we obtain also an explicit expression for the dynamical two-time correlation function of the position in the unperturbed system in the two cooling schedules. For $s \leq t$, we obtain for the instantaneous quench

$$\langle x_s x_t \rangle_0 = \frac{T_i - T_f}{k}\exp\left(-\frac{k}{\gamma}(s+t)\right) + \frac{T_f}{k}\exp\left(-\frac{k}{\gamma}(t-s)\right), \tag{49}$$

and for the linear decrease in temperature schedule

$$\langle x_s x_t \rangle_0 = \begin{cases} \left[\frac{T_i}{k} + \frac{T_f - T_i}{k\tau}\left(s - \frac{\gamma}{2k}\right)\right]\exp\left(-\frac{k}{\gamma}(t-s)\right) + \frac{\gamma(T_f - T_i)}{2k^2\tau}\exp\left(-\frac{k}{\gamma}(s+t)\right) & \text{if } s \leq \tau, \ s \leq t \\ \frac{T_f}{k}\exp\left(-\frac{k}{\gamma}(t-s)\right) - \frac{\gamma(T_f - T_i)}{2k^2\tau}\left[\exp\left(\frac{2k}{\gamma}\tau\right) - 1\right]\exp\left(-\frac{k}{\gamma}(s+t)\right) & \text{if } \tau \leq s \leq t. \end{cases} \tag{50}$$

We see in these two formulas that the characteristic time of convergence toward the Gibbs density $\exp[-(k/2T_f)x^2]/Z_f$ is $\frac{\gamma}{k}$ for the instantaneous quench and $\tau + \frac{\gamma}{k}$ for the linear decrease schedule. Note the relation

$$\langle x_s x_t \rangle_0 = \langle x_s^2 \rangle_0 \exp\left(-\frac{k}{\gamma}(t-s)\right) = \langle x_s^2 \rangle_0 \gamma \left.\frac{\delta}{\delta g_s}\right|_{g=0} \langle x_t \rangle \tag{51}$$

holding for both cooling schedules. It shows that at very large times (i.e., $t > s \gg \tau, \frac{\gamma}{k}$) the correlation functions (49) and (50) take a stationary form depending on $t-s$. The instantaneous mean density of the process is Gaussian at all times. It follows that the mean local velocity has the form

$$v_s^0(x) = \left(-\frac{k}{\gamma} + \frac{T_s}{\gamma \langle x_s^2 \rangle_0}\right)x \tag{52}$$

and the corrective term in the FDT (43) is

$$\langle (v_s^0 \cdot \nabla x)_s x_t \rangle_0 = \langle (v_s^0)_s x_t \rangle_0 = \left(-\frac{k}{\gamma} + \frac{T_s}{\gamma \langle x_s^2 \rangle_0} \right) \langle x_s x_t \rangle_0 . \tag{53}$$

Using relations (51) and (53), the FDT (43) may be rewritten in this case as the identity

$$\left(\partial_s + \frac{k}{\gamma} \right) \langle x_s x_t \rangle_0 = \left. 2T_s \frac{\delta}{\delta g_s} \right|_{g=0} \langle x_t \rangle, \tag{54}$$

which is easy to check directly.

The effective temperatures for the instantaneous quench T_Q^{eff} and for the linear decrease in temperature schedule T_{LD}^{eff} are

$$T_Q^{eff}(s,t,x) = T_f + (T_f - T_i)\exp\left(-\frac{2k}{\gamma} s \right) \quad \text{if } 0 < s \leq t, \tag{55}$$

$$T_{LD}^{eff}(s,t,x) = \begin{cases} T_i + \dfrac{1}{\tau}(T_f - T_i)\left\{ s + \dfrac{\gamma}{2k}\left[1 - \exp\left(-\dfrac{2k}{\gamma} s \right) \right] \right\} & \text{if } 0 < s \leq \tau, s \leq t \\ T_f + (T_f - T_i)\exp\left(-\dfrac{2k}{\gamma} s \right) \dfrac{\exp\left(\dfrac{2k}{\gamma}\tau \right) - 1}{\dfrac{2k}{\gamma}\tau} & \text{if } \tau \leq s \leq t. \end{cases} \tag{56}$$

The two effective temperatures have an expected behavior for large time s converging toward T_f. However, we may see in this system the problems with the physical interpretation of the effective temperature [52,53]. For example, $\lim_{s\to 0} T_Q^{eff} = 2T_f - T_i \neq T_i$ and this expression can be negative if $T_f < T_i/2$. The possibility to find negative effective temperature has been observed also in [32] and, for the kinetically constrained model, in [54]. Moreover, the effective temperature grows toward its limit T_f, which does not correspond to the physical intuition for the temperature of a cooled system. The investigation of the linear-decrease cooling schedule is instructive for the understanding of these two problems. For this schedule, $\lim_{s\to 0} T_{LD}^{eff} = T_i$ and the effective temperature decreases from this value until time τ when it reaches $T_{LD}^{eff}(\tau) = T_f + (T_f - T_i)\{[1 - \exp(-\frac{2k}{\gamma}\tau)] / \frac{2k}{\gamma}\tau\} < T_f$. So, the problem with the initial time limit of T_Q^{eff} was due to the instantaneous modeling of the quench. On the other hand, the second part of the evolution for the linear cooling schedule (i.e., for $s \geq \tau$) begins with an effective temperature below T_f that may be even negative. The last features are not really physically satisfying but the first one explains why the effective temperature T_Q^{eff} converges toward T_f by growing, the fact which is confirmed by Eq. (56). We present below in Fig. 1 a typical joint evolution for the linear cooling schedule of the bath temperature T (crosses) and the effective temperature T_{LD}^{eff} (solid lines) in the case where $T_i = 300$ K, $T_f = 200$ K, and $\frac{2k}{\gamma} = 1$ s^{-1} for $\tau = 0.1$ s (red), $\tau = 1$ s (blue), and $\tau = 10$ s (black). The singularity of the limit $\tau \to 0$ is evident on this graph.

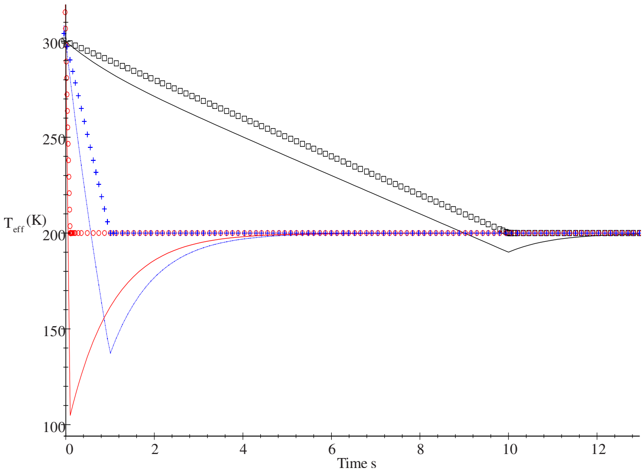

FIG. 1. (Color online) Red solid curve (extreme left at little time): T_{LD}^{eff} for $\tau = 0.1$ s. Red circles: T for $\tau = 0.1$ s. Blue dotted-dashed curve: T_{LD}^{eff} for $\tau = 1$ s. Blue crosses: T for $\tau = 1$ s. Black solid curve (upper in little time): T_{LD}^{eff} for $\tau = 10$ s. Black box (upper in little time): T for $\tau = 10$ s.

B. Response of a diffusion to a pulse of bath temperature

Let us consider the system whose dynamics is governed by Eq. (1) with the variable bath temperature $\beta_t^{-1} = (1 + g_t)\beta_0^{-1}$. We choose as the function f_t the mean instantaneous density ρ_t^0 of the similar system with the constant bath temperature β_0^{-1}. The functional (35) becomes

$$W_T = \int_0^T g_s [(\rho_s^0)^{-1} M_s^\dagger \rho_s^0]_s ds \quad \text{with } M_s = (\beta_0)^{-1}[(-\nabla_j \Pi_s^{ij} + \nabla_j \Gamma_s^{ij})\nabla_i + \Gamma_s^{ij}\nabla_i\nabla_j]. \tag{57}$$

With the same reasoning as in Sec. IV A, we find the link between the response to a pulse of temperature at time s and the dynamical correlation function in the system with stationary inverse temperature β_0,

$$\left.\frac{\delta}{\delta g_s}\right|_{g=0}\langle O_t\rangle = \langle[(\rho_s^0)^{-1}M^\dagger \rho_s^0]_s O_t(x_t)\rangle_0. \tag{58}$$

Here, there does not seem to exist a simplification of this relation in the spirit of Eq. (43) and we cannot say more except for the case when the system with the bath temperature β_0 is an equilibrium one.

Temperature pulse around equilibrium. In the case where the system with the bath temperature β_0 is in equilibrium (i.e., without external force $G_t=0$ and with a stationary Hamiltonian $H_t=H$ and the Gibbsian instantaneous density), the functional (28) takes the form

$$W_T^{ci} = W_T^{ex} = -(\beta_T F_T - \beta_0 F_0) + \int_0^T \dot{\beta}_t H(x_t)dt. \tag{59}$$

We want to prove that the Taylor expansion in the second order of the Jarzynski equality (20) associated with this functional gives the usual fluctuation-dissipation theorem for the energy [25]

$$\partial_s\langle H_s H_t\rangle_0 = \frac{1}{\beta_0}\left.\frac{\delta}{\delta g_s}\right|_{g=0}\langle H_t\rangle. \tag{60}$$

Equality (20) takes now the form

$$\left\langle \exp\left(-\int_0^T \dot{\beta}_t H(x_t)dt\right)\right\rangle = \frac{\int \exp[-\beta_T H(x)]dx}{\int \exp[-\beta_0 H(x)]dx}. \tag{61}$$

We develop the left member in second order in g_t or $h_t=g_t-g_t^2$ assuming that g_t vanishes for $t\leq 0$,

$$\begin{aligned}\left\langle \exp\left(-\int_0^T \dot{\beta}_t H(x_t)dt\right)\right\rangle &= \left\langle \exp\left(\beta_0\int_0^T \dot{h}_t H(x_t)dt\right) + \mathcal{O}(h^3)\right\rangle = \left\langle 1 + \beta_0\int_0^T \dot{h}_t H(x_t)dt + \frac{\beta_0^2}{2}\int_0^T dt\int_0^T \dot{h}_t\dot{h}_s H(x_t)H(x_s)ds\right.\\ &\left.\quad + \mathcal{O}(h^3)\right\rangle = 1 + \beta_0\int_0^T \dot{h}_t\langle H(x_t)\rangle_0 dt + \beta_0\int_0^T dt\int_0^t \dot{h}_t h_u \left.\frac{\delta}{\delta g_u}\right|_{g=0}\langle H_t\rangle du\\ &\quad + \frac{\beta_0^2}{2}\int_0^T dt\int_0^T \dot{h}_t\dot{h}_s\langle H(x_t)H(x_s)\rangle_0 ds + \mathcal{O}(h^3) = 1 + \beta_0\int_0^T \dot{h}_t\langle H(x_t)\rangle_0 dt + \beta_0\int_0^T dt\int_0^T \dot{h}_t\dot{h}_s\theta(t\\ &\quad - s)ds\int_s^t \left.\frac{\delta}{\delta g_u}\right|_{g=0}\langle H_t\rangle du + \frac{\beta_0^2}{2}\int_0^T dt\int_0^T \dot{h}_t\dot{h}_s\langle H(x_t)H(x_s)\rangle_0 ds + \mathcal{O}(h^3),\end{aligned}$$

where the last equality was obtained by expressing $h_u=\int_0^u \dot{h}_s ds$ in the second term and changing the order of integration over s and u. Expansion of the right member of Eq. (61) gives in turn

$$\begin{aligned}\frac{\int \exp[-\beta_T H(x)]dx}{\int \exp[-\beta_0 H(x)]dx} &= 1 + \beta_0 h_T\langle H\rangle_0 + \frac{1}{2}(\beta_0 h_T)^2\langle H^2\rangle_0 + \mathcal{O}(h^3)\\ &= 1 + \beta_0\int_0^T \dot{h}_t\langle H(x_t)\rangle_0 dt\\ &\quad + \frac{\beta_0^2}{2}\int_0^T dt\int_0^T \dot{h}_t\dot{h}_s\langle H(x_t)^2\rangle_0 ds.\end{aligned}$$

The comparison of the terms quadratic in $\dot{h}$ leads to the identity

$$\frac{1}{\beta_0}\int_s^t du\left.\frac{\delta}{\delta g_u}\right|_{g=0}\langle H_t\rangle = \langle H_t^2\rangle_0 - \langle H_s H_t\rangle_0 \tag{62}$$

for $s\leq t$, which gives relation (60) by the derivation with respect to s. Once again the Jarzynski equality appears as a global version of the FDT.

V. CONCLUSIONS

We have discussed fluctuation relations for diffusion processes (1) in a nonstationary thermal bath. Those included the fluctuation relations for the entropy production (12). The work performed on the system no longer verifies such fluctuation relations, but that there still exists relation (29) that permits us to extract the free-energy difference in a nonequilibrium experiment. We proved that the fluctuation relations involving the functional (35) are global versions of the MFDT (43) around a nonequilibrium diffusion extending the MFDT obtained before in [24,32] and of the usual FDT for energy (60) [25] resulting from a pulse of temperature. Along the way, in Sec. IV A we illustrated the extended MFDT on a simple example of a harmonic oscillator in a thermal bath with variable temperature and we investigated the physical meaning of the effective temperature introduced in [52,53] for such a system. One should underline that the interaction with a nonstationary bath is one among many ways to thermally drive a system. For example, the thermodiffusion effect (or Sorret effect) which appears in a bath with nonuniform temperature [i.e., $\beta(x)=\frac{1}{T(x)}$] has been explained in [55], for the unidimensional case, using a stationary nonequilibrium microscopic model of type (1) with $G=0$ but with the thermophoretic force $-\Gamma\frac{dT}{dx}$ added to the drift (4). In the same spirit, many years ago, Landauer [56] proposed a model with the wall temperature varying along a very narrow pipe filled with the Knudsen gas described by a stationary nonequilibrium microscopic model of type (1) but with the thermophoretic force $-\Gamma\frac{dT}{dx}$ and the chemical force $-T\frac{d\Gamma}{dx}$ added to the drift. Finally, another way to drive a system is to consider fluctuating coefficients in Eq. (1); for example, [13] considered a fluctuating mass and [57–59] considered a stochastic friction. It would be interesting to describe the fluctuation relations in those setups.

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