Fluctuation Relations for Diffusions Thermally Driven by a Non-Stationary Bath

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Abstract
In the context of the dynamical evolution in a non-stationary 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 non-equilibrium diffusion or of the usual Fluctuation-Dissipation Theorem for energy resulting from a pulse of temperature.

1 Introduction

One important recent progress in non-equilibrium statistical physics is the discovery of various fluctuation relations which can be viewed as non-perturbative extensions of the usual Fluctuation-Dissipation Theorem (FDT) [29, 36]. Such relations pertain either to non-stationary transient situations [14, 25] or to stationary regimes [16]. In particular, a family of fluctuation relations holds for the distribution of work performed on a system [25, 10, 45, 5] which evolves in an equilibrium bath. This developments had an important impact on the physics of nanosystems and biomolecules [43]. Here, we examine the question of the extension of such relations to an evolution in a non-equilibrium medium. More precisely, we consider systems placed in the thermal bath that is non-stationary, a situation which can be realized experimentally by adding or extracting heat, or by modulating the pressure. Let us remark that non-stationarity of the temperature is linked via Stokes law to non-stationarity of friction and that particles with time-dependent radii have non-constant mass and friction. This last situation is an important problem in astrophysics for the formation of planets through dust aggregation [4, 1]. There is another situation where the non-stationarity of friction can be realized with particles diffusing in ferroelectric fluids when external magnetic fields are controlling the intrinsic viscosity [22].

A famous example of a system which evolves in a non-stationary bath is the Temperature Ratchet Model [11] of a Brownian motor. We can also consider the evolution of a system initially in equilibrium at high temperature (i.e. in the state with the probability density $p_i = \exp(-\frac{H}{T_i})$) put in contact with a non stationary 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 correspond to the cooling schedule with instantaneous initial change of temperature. The age (or waiting time) of the system is then the time elapsed since the quench. Finally, at the more formal level, such non-stationary evolution rules appear naturally from a stationary dynamics after system size or coarse grained expansion [19]. Diffusion properties in a non-stationary medium have been recently studied in the case of unidimensional overdamped dynamics driven by multiplicative non stationary noise [15, 51].

The present paper consists of five sections. Sect. 2 sets the stage and notations for the model of non-equilibrium and non-linear Langevin dynamics that we consider. In Sect. 3, we recall the
In fact, the Gibbs density is then an equilibrium density: an invariant density with vanishing modified probability current \[1\].

The presence of this term assures that in the case with stationary Hamiltonian and temperature \(T\) the characteristic variation of temperature. Let us underline that in this context the term non-linear "concerns the non-homogeneous properties of \(\Gamma_t(x)\) and \(\Pi_t(x)\) (which may characterize non-homogeneous properties of the bath) and not the fact that equation \([1]\) is non-linear. Such non-linear properties appear naturally in many situations: non-ideal plasmas and gases \([26, 27]\), ultracold clusters of atoms or molecules cooled by interaction with laser radiation, active Brownian particles \([13]\). Finally, the additional corrective term in \([1]\) is given by the expression

\[
w_t^i(x) = \partial_y D_t^{ij}(x, y)|_{x=y} - \frac{1}{\beta_t} \partial_j \Pi_t^{ij}(x).
\]

The presence of this term assu ... an external force \(i.e. G = 0\), the Gibbs density \(\text{exp}(-\beta H)\) is an invariant density. The presence of this term \(w_t(x)\) in equation \([1]\) can appear as a makeshift arrangement, but it was extensively studied in the literature of non-linear 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 the second equation of eq. \([1]\):

\[
u_t(x) = -\Gamma_t^{ij}(x) \partial_j H_t(x) + \Pi_t^{ij}(x) \partial_j H_t(x) + G_t^i(x) + w_t^i(x)
\]
the drift term. An elementary case of non-linear Brownian motion is the Landau-Lifshitz-Bloch dynamics of a Brownian spin \[\mathbf{B}\] in an effective magnetic field \[\mathbf{B}_t^{ij}(x) = -\nabla H_t\] (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} \quad \langle \zeta_t \zeta'_s \rangle = \frac{2\lambda_t}{\beta_t} \delta(t - s). \quad (5)

The first term on the right hand side is the precession term, the second one is the damping term and the third one 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...). This dynamics is a particular case of (1) with \(\Gamma_t^{ij}(x) = \lambda_t (\delta^{ij} x^2 - x^i x^j)\) and \(\Pi_t^{ij}(x) = \varepsilon^{ijk} x^k\) with \(\varepsilon^{ijk}\) the totally antisymmetric tensor. In this example one sees the need for the term \(\Pi_t^{ij}(x)\partial_j H_t(x)\) in eq. (1) corresponding to the Hamiltonian vector field which also permits to describe systems of non- overdamped Brownian particles in the phase-space with coordinates \((x, p)\), non-stationary mass, in an external potential \(V_t\), subjected to a non-conservative force \(f_t\) and in a non-stationary bath giving rise to the noise and the non-homogeneous, non-linear but isotropic drag \(\gamma_t(q, p)\). The Stratonovich SDE which governs this model is then

\[
\begin{align*}
\dot{q} &= \frac{p}{m_t} - \gamma_t(q, p) - \nabla V_t(q) + f_t(q) + \frac{\nabla \gamma_t(q, p)}{2\beta_t} + \sqrt{\frac{2\gamma_t}{m_t}} \eta(t) \\
\dot{p} &= -\frac{\gamma_t(q, p)}{m_t} p - \nabla V_t(q) + f_t(q) + \frac{\nabla \gamma_t(q, p)}{2\beta_t} + \sqrt{\frac{2\gamma_t}{m_t}} \eta(t) \quad \text{with} \quad \langle \eta(t) \eta'(s) \rangle = \delta(t - s). \quad (6)
\end{align*}
\]

which is again a sub-case of (1) with \(\Gamma_t = (0 \quad 0 \quad 0)\), \(\Pi_t = (0 \quad 0 \quad 0)\), \(H_t = f_t^2 + V_t(q)\) and \(G_t = (0 \quad 0)\). 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 \[\text{(10)}\].

We start by collecting the elementary properties of diffusion processes that we shall need \[\text{(17)}\]. The Markovian generator \(L_t\) of the process \(x_t\) satisfying the SDE (1) is defined by the relation

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

where the modified drift \(\hat{u}_t\) is defined in term of the drift (4):

\[\hat{u}_t(x) = u_t(x) - \partial_j D_t^{ij}(x, y)|_{y=x}. \quad (8)\]

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 \(L_t^\dagger\) of the generator \(L_t\)

\[\partial_t \rho_t = L_t^\dagger \rho_t \quad (9)\]

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

\[\partial_t \rho_t = -\nabla \cdot \tilde{j}_t = -\nabla \cdot (\rho_t \tilde{v}_t) \quad \text{where} \quad \tilde{j}_t^i = (\tilde{u}_t^i - \beta_t^{-1} \Gamma_t^{ij} \nabla_j) \rho_t \quad \text{and} \quad \tilde{v}_t^i = \frac{\tilde{j}_t^i}{\rho_t}. \quad (10)\]

As was explained in \[\text{(8)}\], 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

\[j_t^i = j_t^i + \beta_t^{-1} \nabla_j (\Pi_t^{ij} \rho_t) \quad \text{and} \quad v_t^i = \frac{j_t^i}{\rho_t}. \quad (11)\]

which verify also the continuity equation \[\text{(10)}\] but vanish in the case of stationary Hamiltonian and temperature (i.e. \(H_t = H, \beta_t = \beta\)) for vanishing external force \(G_t = 0\).
3 Fluctuation relations and time inversion

In [5], 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 Sect. 3 of [7]. With the use of combined Girsanov and Feynman-Kac formulae, one obtains the detailed fluctuation relation (DFR)

\[ \mu_0(dx) P_T(x; dy, dW) e^{-W} = \mu_0^{\ast}(dy^\ast) P^{\ast}_T(y^\ast; dx^\ast, d(-W)), \]  

where

1. \( \mu_0(dx) = \rho_0(x) \, dx \) is the initial distribution of the original forward process (1).
2. \( \mu_0^{\ast}(dx) = \rho_0^\ast(x) \, dx \) 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 \( T \) 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),
4. \( P^{\ast}_T(x; dy, dW) \) is the similar joint probability distribution for the backward process.

The time inversion acts on time and space by an involution

\[ (t, x) \mapsto (t^\ast = T - t, x^\ast). \]  

(13)

Such an involution induces the action \( x \mapsto \overline{x} \) on trajectories by the formula \( \overline{x}_t = x_{T-t}^\ast \) and, further, the action on functionals of trajectories \( F \mapsto \overline{F} \) by setting \( \overline{F}[x] = F[\overline{x}] \). To recover various fluctuation relations discussed in the literature [23 31 10 25 46 8], one divides the drift part (1) of (1) 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:

\[ u^{\ast}_{T-t, \pm}(x^\ast) = \pm (\partial_k x^\ast)_k(x) u_{t, \pm}(x), \quad u^\ast = u_+^\ast + u_-^\ast. \]  

(14)

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

\[ \dot{x} = u_t^\ast(x) + \eta_t^\ast(x) \]  

(15)

and, in general, differs from the naive time inversion \( \overline{x}_t \) of the forward process. The functional \( W_T \) is given by the expression

\[ W_T = \ln \rho_0(x_0) - \ln(\rho_0^\ast(x_T^\ast)\sigma(x_T)) + \int_0^T J_t \, dt \]  

(16)

where \( \sigma(x) = \left| \det \left( \frac{\partial^2 x^\ast}{\partial x} \right) \right| \) 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

\[ J_t = \beta \tilde{u}_{t,+}(x_t) \cdot \Gamma_{t}^{-1}(x_t) (\dot{x}_t - u_{t,-}(x_t)) - (\nabla \cdot u_{t,-})(x_t). \]  

(17)

The time integral in eq. (16) is taken in the Stratonovich sense. When \( \mu_0^\ast(dx^\ast) = \mu_t(dx) \) then the boundary contribution \( \ln \rho_0(x_0) - \ln(\rho_0^\ast(x_T^\ast)\sigma(x_T)) \) 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 Principle gives us that the work \( T_T \) performed on the system can be expressed in term of \( J_t \):

\[ T_T = H_T(x_T) - H_0(x_0) + \int_0^T \frac{J_t}{\beta_t} \, dt. \]  

(18)
We can underline that in this setup, and contrary to the case of a the stationary bath [5], the work $T_r$ cannot be identified with the functional $W_r$ 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 $\mu_0$ and $\mu_0^*$ are not normalized, or even not normalizable. When they are normalized, let us denote by $\langle \cdot \rangle$ and by $\langle \cdot \rangle^*$ the expectations of functionals of, respectively, the forward and the backward process on the time interval $[0, T]$, with initial distributions $\mu_0$ and $\mu_0^*$. One of the immediate consequences of the DFR equation (12) is the (generalized) Jarzynski equality [25]

$$\langle e^{-W_r} \rangle = 1$$

(19)

obtained by the integration of the both sides of eq (12). It implies the inequality $\langle W_r \rangle \geq 0$ that has the form of the Second Law of Thermodynamics stating the positivity of the average entropy production. With a little more work [5], the DFR (12) may be cast into a form of the (generalized) Crooks relation [10]:

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

(20)

We will now restrict ourselves to the class of time inversions [14] such that there exists a non-stationary density $f_t$ such that

$$\tilde{u}_{+,t} = \beta_t^{-1} \Gamma_t \nabla \ln f_t$$

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

(21)

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

$$J_t = \dot{x}_t \cdot (\nabla \ln f_t)(x_t) + ((f_t)^{-1} L_{\pi_t}^T) f_t(x_t).$$

(22)

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

$$W_T = \int_0^T ((f_t)^{-1} L_{\pi_t}^T f_t - \partial_t \ln (f_t)) (x_t) \, dt .$$

(23)

We shall see that the fluctuation relations associated to this peculiar family of inversions are the natural generalizations of the FDT. We shall now describe particular cases in this family of time inversions.

### 3.1 Complete reversal [5]

As the function $f_t$ in (21) we take the instantaneous density function (i.e. $f_t = \rho_t$) of the forward process (1) distributed with initial condition $f_0$. Here, the functional (23) trivially vanishes $W_T = 0$ and the DFR (12) takes the form of the generalized detailed balance

$$\mu_0(dx) \, P_\pi(x; dy) = \mu_\pi(dy) \, P_\pi^T(y^*; dx^*) .$$

(24)

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

$$j_{\pi_t}^\ast(x^*) = - (\partial_t x^*)^k \, j_{\pi_t}^k(x) .$$

(25)

This inversion is employed in many articles in probability theory [21, 22, 23, 39, 38]. It corresponds to the vanishing overall entropy production.

### 3.2 Current reversal [8, 5]

Another useful choice of time inversion, called the current reversal is based on the choice $f_t = \pi_t$ where $\pi_t$ satisfies $L_{\pi_t}^T \pi_t = - \nabla \cdot \tilde{j}_t = 0$. In the case where $G_t = 0$, we have $\pi_t = \exp(-\beta_t (H_t - F_t))$ with $F_t$ the free energy (i.e. $\exp(-\beta_t F_t) = \int \exp(-\beta_t H_t)$). One can show [3] that $\pi_t(x) \equiv \rho_t^\ast(x)$. We will now restrict ourselvess to the class of time inversions (14) such that there exists a non-stationary density $f_t$ such that

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\[ \pi_t^*(x^*) \] is the density for the backward process which correspond to the conserved current \( \nabla \cdot j_t^* = 0 \) and that eq. (25) still holds. The functional (23) takes now the form

\[ W^c_{\pi_t} = -\int_0^T (\partial_t \ln \pi_t)(x_t) \, dt, \quad (26) \]

where the index "\text{ex}" stands for "excess" [13, 15]. For the backward process, the functional \( W^c_{\pi} \) is given by the same expression with \( \pi_t \) replaced by \( \pi_t^* \). The Jarzynski equality (19) for this case was first proven in one dimension in [21] and in the general case in [20, 22, 25].

### 3.3 Canonical inversion [5]

A natural choice for the system (4) 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. This corresponds to the choice \( \tilde{u}_{t,t} = -\Gamma_t \nabla H_t \) in (21). The functional (23) becomes

\[ W^c_{\pi} = -(\beta_T F_T - \beta_0 F_0) + \int_0^T \left[ \partial_t (\beta_t H_t) + \beta_t G_t \cdot \nabla H_t - \nabla \cdot G_t \right] (x_t) \, dt \quad (27) \]

where the index "\text{ci}" means "canonical inversion". The generalized Jarzynski equality (19) can be rewritten in the form:

\[ \left\langle \exp \left\{ -\int_0^T \left[ \partial_t (\beta_t H_t) + \beta_t G_t \cdot \nabla H_t - \nabla \cdot G_t \right] (x_t) \, dt \right\} \right\rangle = \exp \left\{ -(\beta_T F_T - \beta_0 F_0) \right\} \quad (28) \]

which permits to extract the difference of free energy out of a non-equilibrium experiment in a non-stationary bath (but the connexion with the work performed is lost). For example, for the Brownian particle (6), this functional takes the form:

\[ W^c_{\pi} = -(\beta_T F_T - \beta_0 F_0) + \int_0^T \left[ \beta_t \left( \frac{k_t}{2m_t} + V_t(q_t) \right) + \beta_t \left( -\frac{\mu_t \dot{q}_t}{m_t} + (\partial_t V_t)(q_t) + (f_t \nabla V_t)(q_t) \right) \right] \, dt \quad (29) \]

which, as compared to the functional which appears in the usual Jarzynski equality [23, 28], contains new terms proportional to the variation of temperature \( \partial_t \beta_t \) and of mass \( \partial_t m_t \). From the Jarzynski equality (19), one can deduce the inequality which constrains the evolution in a non-stationary bath

\[ \left\langle W^c_{\pi} \right\rangle \geq 0. \]

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

\[ \int_0^T (\partial_t \beta_t) \langle H(x_t) \rangle \, dt \geq (\beta_T F_T - \beta_0 F_0). \]

Let us consider now the overdamped particle in a 3-dimensional time-dependent harmonic potential. Such example admits an analytical computation of the distribution of the functional \( W^c_{\pi} \). We consider a non stationary bath \((\gamma_t, \beta_t)\) and the harmonic potential \( U_t(x) = \frac{k_t}{2}(x-a_t)^2 \) 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 \( \rho_0(x) = \exp(-\beta_0(U_0(x) - F_0)) = \left( \frac{C_0}{\beta_0} \right)^3 \exp \left( -\frac{C_0}{\beta_0} (x-a_0)^2 \right) \). 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 [31, 34]. The system satisfies the linear SDE

\[ \dot{x} = -\frac{k_t}{\gamma_t} (x-a_t) + \eta_t \quad \text{with} \quad \langle \eta_t^i \eta_t^j \rangle = \frac{2}{\beta_t \gamma_t} \delta(t-s) \delta^{ij}. \quad (30) \]
The functional $W_T^{ci} = W_T^{cz}$ 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 $\rho_0$ is Gaussian due to the linearity of (30). A straightforward calculation gives the mean

$$\langle W_T^{ci} \rangle = C \int_0^T dt \int_0^t \dot{a}_t \cdot \dot{a}_s \exp \left( -\int_s^t \frac{k_u}{\gamma_u} du \right) ds \quad (31)$$

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

$$V_T \equiv \left( W_T^{ci} - \langle W_T^{ci} \rangle \right)^2 = 2C \int_0^T dt \int_0^t \dot{a}_t \cdot \dot{a}_s \exp \left( -\int_s^t \frac{k_u}{\gamma_u} du \right) ds. \quad (32)$$

The distribution of $W_T^{ci}$ is then

$$P_T(W) = \frac{1}{\sqrt{2\pi V_T}} \exp \left( -\frac{(W - \langle W_T^{ci} \rangle)^2}{2V_T} \right) \quad (33)$$

and an elementary calculus shows that the Jarzynski equality (19) is equivalent to the fact that $V_T = 2 \langle W_T^{ci} \rangle$ which is evident from the comparison of (31) and (32).

### 3.4 New inversion

We choose for the function $f_t$ the mean instantaneous density $\rho'_t$ of another Langevin dynamics (1) which possesses the same parameters $\Gamma_t, \Pi_t, G_t$ but with another non-autonomous Hamiltonian $H'_t$ and another bath temperature $\beta'_t$. We note $L_t$ (resp. $L'_t$) the Markovian generators of the process with the Hamiltonian $H_t$ and the bath temperature $\beta_t$ (resp. $H'_t$ and $\beta'_t$). The functional (23) takes now the form

$$W_T = \int_0^T \left[ (\rho'_t)^{-1} (L_t - L'_t) \rho'_t \right](x_t) dt. \quad (34)$$

This new inversion will permit to recover new generalizations of the FDT around non-stationary non-equilibrium diffusions, see also [6, 7, 3].

### 4 Generalizations of the Fluctuation-Dissipation Theorem

As noted in [17, 31], the fluctuation relations may be viewed as extensions to the non-perturbative regime of the Green-Kubo and Onsager relations for the non-equilibrium transport coefficients valid within the linear response description of the vicinity of the equilibrium. Ref. [6] 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 to (26) or (27) gives in the second order of the Taylor expansion in $g$ the usual FDT. Ref. [7] showed that similar correspondence still holds around non-equilibrium steady states for a stationary dynamics with an external force (i.e. $G \neq 0$). In this case, it is the Crooks relation (20) associated to the functional (26) which gives the modified Fluctuation-Dissipation Theorem [4, 21] after the first order Taylor expansion in $g$. The second order expansion of Jarzynski equality (19) associated to the functional (26) 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 Sect 3.4.
4.1 FDT around non-stationary diffusions

We consider the system (31) with the Hamiltonian \( H_t(x) = H^0_t(x) - g_{a,t} O^a(x) \). Following Sect. 3.4, we choose \( f_i \) as the mean instantaneous density \( \rho^i_t \) of the unperturbed system with \( g = 0 \). The functional (34) becomes

\[
W_T = \int_0^T g_{b,s} \left( (\rho^0_s)^{-1} M^b_s \rho^0_s \right)_s \, ds \quad \text{with} \quad M^a_s = (\Gamma_s \nabla O^a - \Pi_s \nabla O^a) \cdot \nabla,
\]

where the subscript \(^+ \) on \((\rho^0_s)^{-1} M^b_s \rho^0_s \)

 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 the system (31) with the Hamiltonian \( H_t \), associated to a single time functional \( F[x] = O^a(x_t) \equiv O^a_t \) \((0 < t < T)\):

\[
\langle O^a_t \, e^{-W_T} \rangle = \langle O^a_{T-t} \rangle^R.
\]

We shall denote by \( \langle . \rangle_0 \) the average of the process with the dynamics driven by \( H^0_t \) and by \( L^0, v^0 \), respectively, its Markovian generator, its mean local velocity and its modified mean local velocity. The first order Taylor expansion

\[
\exp(-W_T) = 1 + \int_0^T g_{b,s} \left( (\rho^0_s)^{-1} M^b_s \rho^0_s \right)_s \, ds + \mathcal{O}(g^2)
\]

in (30) gives the relation

\[
\langle O^a_t \rangle_0 + \int g_{b,s} \delta \frac{\partial}{\partial g_{b,s}} \bigg|_{g=0} \langle O^a_t \rangle_0 \, ds - \int_0^T g_{b,s} \langle O^a_t (\rho^0_s)^{-1} M^b_s \rho^0_s \rangle_0 \, ds + \mathcal{O}(g^2) = \langle O^a_{T-t} \rangle^R.
\]

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

\[
\frac{\delta}{\delta g_{b,s}} \bigg|_{g=0} \langle O^a_t \rangle = \left\langle (\rho^0_s)^{-1} M^b_s \rho^0_s \right\rangle O^a_t \right|_{0}.
\]

A short calculation gives:

\[
(\rho^0_s)^{-1} M^b_s \rho^0_s = \beta_s v^0_s \cdot \nabla O^b - \beta_s L^0_s O^b + \Pi^j_s (\partial_j O^b) \partial_i \ln(\rho_s^0) + (\partial_i \Pi^j_s) \partial_j O^b
\]

\[
= \beta_s \left( 2 v^0_s \cdot \nabla - L^0_s \right) O^b - \beta_s v^0_s \cdot \nabla O^b + \Pi^j_s (\partial_j O^b) \partial_i \ln(\rho_s) + (\partial_i \Pi^j_s) \partial_j O^b
\]

\[
= \beta_s \left( 2 v^0_s \cdot \nabla - L^0_s \right) O^b - \beta_s v^0_s \cdot \nabla O^b.
\]

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

\[
\partial_s \langle O^b_s O^a_t \rangle_0 = \left\langle \left( (2 v^0_s \nabla - L^0_s) O^b_s \right) O^a_t \right\rangle_0.
\]

With (39), (41) and (40), we obtain the Modified Fluctuation-Dissipation Theorem :

\[
\partial_s \langle O^b_s O^a_t \rangle_0 = \frac{1}{\beta_s} \frac{\delta}{\delta g_{b,s}} \bigg|_{g=0} \langle O^a_t \rangle = \langle v^0_s \cdot \nabla O^b \rangle_0 O^a_t \right|_{0}.
\]

This is a generalization of the FDT around a non-equilibrium diffusion process in a stationary bath \( (\beta_s = \beta) \) of refs. [4] [5] [3] [11] and of FDT around non-stationary Langevin equation of ref. [52]. It may be also proven as in [6] using the fact that the diffusion process becomes an equilibrium one in the Lagrangian frame of its modified mean local velocity \( v^0_s \), verifying in that frame the usual FDT. The transformation of the latter back to the Eulerian (i.e. laboratory) frame leads to (42). Let us remark that here, similarly as in the stationary case discussed in [7], the Jarzynski equality (19) for the functional (34) leads upon the second order expansion in \( g \) to a particular case of the MFDT where the observable \( O^a_t \) is replaced by \( A^a = (\rho^0_t)^{-1} M^a_t \rho^0_t \) which is a (time-dependent) functional of \( O^a \).
The violation of the usual FDT can be parametrized by using \( T^{ff}(s,t,O^s) \) via the introduction of the so-called effective temperature \([12,9]\) defined by

\[
T^{eff}(s,t,O^s) = \frac{\partial_s \langle O_s^a O_t^b \rangle_0}{\delta g_{a,s}} \bigg|_{g=0} \langle O_t^b \rangle_0 = \frac{1}{\beta_s} + \frac{\langle (u^a_s \cdot \nabla O^b)_s O^b_t \rangle_0}{\delta g_{a,s}} \bigg|_{g=0} \langle O_t^b \rangle_0.
\] (43)

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 non-stationary bath.** The SDE which governs this system is

\[
\dot{x} = -\frac{k}{\gamma} x + \eta, \quad \text{with} \quad \langle \eta_t \eta_s \rangle = \frac{2T_i}{\gamma} \delta(t-s).
\] (44)

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

- **Instantaneous quench**
  \[
  T_i = \begin{cases} 
  T_i & \text{if } t = 0 \\
  T_f & \text{if } t > 0 \end{cases}
  \] (45)

- **Linear decrease of temperature**
  \[
  T_i = \begin{cases} 
  T_i + \frac{1}{\tau} (T_f - T_i) & \text{if } t \leq \tau \\
  T_f & \text{if } t \geq \tau \end{cases}
  \] (46)

Due to the linearity of eq. (44), one can compute explicitly the response of the position to an external perturbation \( V(x) = \frac{k}{2} x^2 \rightarrow V'_f(x) = V(x) - g_i x \) with \( g_0 = 0 \):

\[
\frac{\delta}{\delta g_s} \bigg|_{g=0} \langle x_t \rangle = \frac{1}{\gamma} \exp \left( -\frac{k}{\gamma} (t-s) \right).
\] (47)

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 2-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:

\[
\langle x_s x_t \rangle_0 = \frac{T_i - T_f}{k} \exp \left( -\frac{k}{\gamma} (s + t) \right) + \frac{T_f}{k} \exp \left( -\frac{k}{\gamma} (t-s) \right)
\] (48)

and for the linear decrease of temperature schedule:

\[
\langle x_s x_t \rangle_0 = \begin{cases} 
  \left( \frac{T_i}{k} + \frac{T_f - T_i}{k \tau} (s - \frac{\gamma}{2} t) \right) \exp \left( -\frac{k}{\gamma} (t-s) \right) + \frac{\gamma(T_f - T_i)}{2k^2 \tau} \exp(-\frac{k}{\gamma} (s+t)) & \text{if } s \leq \tau, s \leq t \\
  \frac{T_f}{k} \exp \left( -\frac{k}{\gamma} (t-s) \right) - \frac{\gamma(T_f - T_i)}{2k^2 \tau} \left( \exp(\frac{2k}{\gamma} \tau) - 1 \right) \exp \left( -\frac{k}{\gamma} (s+t) \right) & \text{if } \tau \leq s \leq t.
\end{cases}
\] (49)

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

\[
\langle x_s x_t \rangle_0 = \langle x_r^2 \rangle_0 \exp \left( -\frac{k}{\gamma} (t-s) \right) = \langle x_r^2 \rangle_0 \frac{\delta}{\delta g_s} \bigg|_{g=0} \langle x_t \rangle
\] (50)

holding for both cooling schedules. It shows that at very large times (i.e. \( t > s \gg \tau, \frac{k}{T_f} \)) the correlation functions (48) and (49) 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

\[ v^0_s(x) = \left( -\frac{k}{\gamma} + \frac{T_s}{\gamma \langle x_s^2 \rangle_0} \right) x \]

and the corrective term in the FDT (42) is

\[ \langle (v^0_s \cdot \nabla) x_s \rangle_0 = \langle (v^0_s) x_s \rangle_0 = \left( -\frac{k}{\gamma} + \frac{T_s}{\gamma \langle x_s^2 \rangle_0} \right) \langle x_s x_t \rangle_0 \]

Using the relations (50) and (52), the FDT (42) may be rewritten in this case as the identity

\[ \left( \partial_s + \frac{k}{\gamma} \right) \langle x_s x_t \rangle_0 = 2T_s \frac{\delta}{\delta g_s} \bigg|_{g=0} \langle x_t \rangle \]

which is easy to check directly.

The effective temperatures for the instantaneous quench \( T^e_{Q} \) and for the linear decrease of temperature schedule \( T^e_{LD} \) are:

\[ T^e_{Q}(s,t,x) = T_f + (T_f - T_i) \exp(-\frac{2k}{\gamma}s) \quad \text{if} \quad 0 < s \leq t \]

and

\[ T^e_{LD}(s,t,x) = \begin{cases} T_i + \frac{1}{2} \gamma (T_f - T_i) \left( s + \frac{\gamma}{2\kappa} \left( 1 - \exp(-\frac{2k}{\gamma}s) \right) \right) & \text{if} \quad 0 < s \leq \tau, \ s \leq t \vspace{0.5cm} \\
T_f + (T_f - T_i) \exp\left(-\frac{2k}{\gamma}s\right) \frac{\exp(\frac{2k}{\gamma}\tau)}{\frac{2k}{\gamma}} & \text{if} \quad \tau \leq s \leq t. \end{cases} \]

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 [12, 9]. For example, \( \lim_{s \to 0} T^e_{Q} = 2T_f - T_i \neq T_i \) and this expression can be negative if \( T_f < \frac{2k}{\gamma} \). The possibility to find negative effective temperature has been observed also in [7] and, for the kinetically constrained model, in [37]. 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, \( \lim_{s \to 0} T^e_{LD} = T_i \) and the effective temperature decreases from this value until time \( \tau \) when it reaches \( T^e_{LD}(\tau) = T_f + (T_f - T_i) \frac{1-\exp(\frac{-\tau}{\gamma})}{\frac{2k}{\gamma}} < T_f \). So, the problem with the initial time limit of \( T^e_{Q} \) 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 \( T^e_{Q} \) converges toward \( T_f \) by growing, the fact which is confirmed by (55). We represent below in Figure 1 typical joint evolution for the linear cooling schedule of the bath temperature \( T \) (the crosses) and the effective temperature \( T^e_{LD} \) (the solid lines) in the case where \( T_i = 300K, T_f = 200K \) and \( \frac{2k}{\gamma} = 1s^{-1} \) for \( \tau = 0.1s \) (red) \( \tau = 1s \) (blue) and \( \tau = 10s \) (black). The singularity of the limit \( \tau \to 0 \) is evident on this graph.

### 4.2 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 \( \beta_\tau^{-1} = (1 + g_\tau)\beta_0^{-1} \). We choose as the function \( f_\tau \) the mean instantaneous density \( \rho^0_s \) of the similar system with the constant bath temperature \( \beta_0^{-1} \). The functional \( \tilde{W}_T \) becomes

\[ W_T = \int_0^T g_s \left( (\rho^0_s)^{-1} M_s \rho^0_s \right) s ds \quad \text{with} \quad M_s = (\beta_0)^{-1} \left( \langle -\nabla j \Pi^{ij}_s \nabla \rangle \right) \nabla_i + \Gamma_s^{ij} \nabla_i \nabla_j \].

(56)
Figure 1: Red solid: $T_{LD}^{\text{eff}}$ for $\tau = 0.1\text{s}$. Red crosses: $T$ for $\tau = 0.1\text{s}$. Blue solid: $T_{LD}^{\text{eff}}$ for $\tau = 1\text{s}$. Blue crosses: $T$ for $\tau = 1\text{s}$. Black solid: $T_{LD}^{\text{eff}}$ for $\tau = 10\text{s}$. Black crosses: $T$ for $\tau = 10\text{s}$.

With the same reasoning as in Sect. 4.1, 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 $\beta_0$:

$$\frac{\delta}{\delta g_s} \bigg|_{g=0} \langle O_t \rangle = \langle \left( (\rho_s^0)^{-1} M^\dagger \rho_s^0 \right)_s O_t(x_t) \rangle_0. \quad (57)$$

Here, there does not seem to exist a simplification of this relation in the spirit of (42) and we cannot say more except for the case when the system with the bath temperature $\beta_0$ is an equilibrium one.

**Temperature pulse around equilibrium.** In the case where the system with the bath temperature $\beta_0$ 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 (27) takes the form:

$$W_{ci}^\text{ex} = W_{F}^\text{ex} = - (\beta T F_T - \beta_0 F_0) + \int_0^T \dot{\beta}_t H(x_t) \, dt. \quad (58)$$

We want to prove that the Taylor expansion in the second order of the Jarzynski equality (19) associated to this functional gives the usual Fluctuation-Dissipation Theorem for the energy (42):

$$\partial_s \langle H_s H_t \rangle_0 = \frac{1}{\beta_0 g_s} \frac{\delta}{\delta g_s} \bigg|_{g=0} \langle H_t \rangle. \quad (59)$$
The equality \[ \left\langle \exp \left( -\int_0^T \hat{\beta}_t H(x_t) \, dt \right) \right\rangle = \int \frac{\exp(-\beta_T H(x)) \, dx}{\int \exp(-\beta_H(x)) \, dx}. \] (60)

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 \( t \leq 0 \):

\[
\left\langle \exp \left( -\int_0^T \hat{\beta}_t H(x_t) \, dt \right) \right\rangle = \left\langle \exp \left( \beta_0 \int_0^T \hat{h}_t H(x_t) \, dt \right) + \mathcal{O}(h^3) \right\rangle
\]

\[
= \left( 1 + \beta_0 \int_0^T \hat{h}_t H(x_t) \, dt + \frac{\beta_0^2}{2} \int_0^T \int_0^T \hat{h}_t \hat{h}_s H(x_t) H(x_s) \, ds + \mathcal{O}(h^3) \right)
\]

\[
= 1 + \beta_0 \int_0^T \hat{h}_t \langle H(x_t) \rangle_0 \, dt + \beta_0 \int_0^T \int_0^T \hat{h}_t \hat{h}_s \delta(t-s) \, ds \int_0^t \frac{\delta}{\delta g_a} \bigg|_{g=0} \langle H_t \rangle \, du
\]

\[
+ \frac{\beta_0^2}{2} \int_0^T \int_0^T \hat{h}_t \hat{h}_s \langle H(x_t) H(x_s) \rangle_0 \, ds + \mathcal{O}(h^3),
\]

where the last equality was obtained by expressing \( h_u = \int_0^u \hat{h}_s \, ds \) in the second term and changing the order of integration over \( s \) and \( u \). Expansion of the right member of eq. (60) gives in turn

\[
\frac{\int \exp(-\beta_T H(x)) \, dx}{\int \exp(-\beta_H(x)) \, dx} = 1 + \beta_0 h_T \langle H \rangle_0 + \frac{1}{2} (\beta_0 h_T)^2 \langle H^2 \rangle_0 + \mathcal{O}(h^3)
\]

\[
= 1 + \beta_0 \int_0^T \hat{h}_t \langle H(x_t) \rangle_0 \, dt + \frac{\beta_0^2}{2} \int_0^T \int_0^T \hat{h}_t \hat{h}_s \langle H(x_t)^2 \rangle_0 \, ds.
\]

The comparison of the terms quadratic in \( \hat{h} \) leads to the identity

\[
\frac{1}{\beta_0} \int_s^t \frac{\delta}{\delta g_a} \bigg|_{g=0} \langle H_t \rangle = \langle H_t^2 \rangle_0 - \langle H_s H_t \rangle_0
\] (61)

for \( s \leq t \) which gives the relation (59) by the derivation with respect to \( s \). Once again the Jarzynski equality appears as a global version of the FDT.

5 Conclusions

We have discussed fluctuation relations for diffusion processes \([1]\) in a non-stationary 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 a relation \([28]\) which permits to extract the free energy difference in a non-equilibrium experiment. We proved that the fluctuation relations involving the functional \([34]\) are global versions of the Modified Fluctuation-Dissipation Theorem \((\text{MFDT})\) \([42]\) around a non-equilibrium diffusion extending the \(\text{MFDT}\) obtained before in \([7] [6]\) and of the usual \(\text{FDT}\) for energy \([59] [42]\) resulting from a pulse of temperature. On the way, in Sec 4.1 we illustrated the extended \(\text{MFDT}\) 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 \([12] [9]\) for such a system. One should underline that the interaction with a non-stationary 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 non-uniform temperature (i.e \( \beta(x) = \frac{1}{T(x)} \)) has been explained in \([49]\), for the unidimensional case, using a stationary non-equilibrium microscopic model of the type \([1]\) with \( G = 0 \) but with the thermophoretic force \( -\Gamma \frac{dT}{dx} \) added to the drift \([1]\). In the same spirit, many years ago, Landauer \([30]\) proposed a model with the wall temperature varying along a very narrow pipe filled with the Knudsen gas described by a stationary non-equilibrium microscopic model of the 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, other way to drive a system is to consider fluctuating coefficients in \([1]\), for example \([1]\) considered a fluctuating mass and \([44, 33, 48]\) a stochastic friction. It would be interesting to describe the fluctuation relations in those setups.

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