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Fluctuation relations in simple examples of non-equilibrium steady states

Raphaël Chetrite¹, Gregory Falkovich^{2,3} and Krzysztof Gawędzki¹

¹ Laboratoire de Physique, CNRS, ENS-Lyon, Université de Lyon, 46 Allée d'Italie, F-69364 Lyon, France

² Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel

³ KITP, University of California at Santa Barbara, Santa Barbara, CA 93106, USA

E-mail: raphael.chetrite@ens-lyon.fr, gregory.falkovich@weizmann.ac.il and kgawedzk@ens-lyon.fr

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Abstract. We discuss fluctuation relations in simple cases of non-equilibrium Langevin dynamics. In particular, we show that, close to non-equilibrium steady states with non-vanishing probability currents, some of these relations reduce to a modified version of the fluctuation–dissipation theorem. The latter may be interpreted as the equilibrium-like relation in the reference frame moving with the mean local velocity determined by the probability current.

Keywords: driven diffusive systems (theory), exact results, stationary states

Contents

1. Introduction	2
2. NESS in Langevin processes	5
2.1. NESS for resurrecting processes	7
2.2. NESS for forced diffusions on circle	7
3. Modified fluctuation–response relation	8
4. Modified fluctuation–dissipation theorem	8
4.1. Lagrangian-frame interpretation	9
4.2. Direct derivation	11
5. General fluctuation relations	11
6. Fluctuation relations in Langevin dynamics	13
6.1. Fluctuation relations for resurrecting processes	15
6.2. Fluctuation relations for forced diffusions on circle	16
7. Fluctuation relations close to NESS	16
7.1. Reduction of Crooks’ DFR to MFDT	17
7.2. Jarzynski–Hatano–Sasa equality and MFDT	18
8. Conclusions	20
Acknowledgments	20
Appendix A	20
Appendix B	23
References	24

1. Introduction

In statistical mechanics, the fluctuation–dissipation theorem (FDT) provides a simple relation in an equilibrium state between the response of the fixed-time averages to small time-dependent perturbations of the Hamiltonian and the dynamical correlations [34, 35]. Let $O^a(x)$ for $a = 1, \dots, A$ be a collection of (classical) observables. With the shorthand notation O_t^a for the single-time functions $O^a(x_t)$ of the dynamical process x_t , the response function and the two-time correlation function in a steady state are, respectively,

$$\mathcal{R}^{ab}(t - s) = \left. \frac{\delta}{\delta h_s} \right|_{h=0} \langle O_t^a \rangle_h \quad \text{and} \quad \mathcal{C}^{ab}(t - s) = \langle O_t^a O_s^b \rangle_0, \quad (1.1)$$

where $\langle - \rangle_h$ denotes the dynamical expectation obtained from the steady state by replacing the time-independent Hamiltonian $H(x)$ by a slightly perturbed time-dependent one $H(x) - h_t O^b(x)$. The FDT asserts that, when the unperturbed state is the equilibrium at inverse temperature β , then

$$\beta^{-1} \mathcal{R}^{ab}(t - s) = \partial_s \mathcal{C}^{ab}(t - s). \quad (1.2)$$

Such a direct relation between the response and correlation functions is violated in systems out of equilibrium and a lot of interest in the research on non-equilibrium statistical mechanics was devoted to such violations. In particular, they were studied intensively for glassy systems [13, 10, 7], for colloidal suspensions [3, 19], for granular matter [2, 6] and for biophysical systems [39, 27]. In recent years, it has been realized that the FDT, as well as the Green–Kubo relation, another linear response law of the equilibrium regime, are special cases of more general fluctuation relations that hold also far from equilibrium. Such relations pertain either to non-stationary transient situations [16, 28] or to stationary regimes [20]. In particular, the so-called Jarzynski equality [28] for the dynamics with a time-dependent Hamiltonian reduces to the FDT for tiny time variations [8].

In the present paper, we revisit the violations of the FDT in simple examples of non-equilibrium steady states (NESS) for systems with few degrees of freedom evolving according to the Langevin equation, possibly including non-conservative forces, see, e.g., [42, 14, 41, 25, 23, 44]. For such systems, we show a modified fluctuation–dissipation theorem (MFDT) that may be written in the form

$$\beta^{-1}\mathcal{R}_L^{ab}(t, s) = \partial_s\mathcal{C}_L^{ab}(t, s) \quad (1.3)$$

similar to the equilibrium relation, somewhat in the spirit of [44]. Above, $\mathcal{R}_L^{ab}(t, s)$ and $\mathcal{C}_L^{ab}(t, s)$ denote the response function and the dynamical correlation function in the Lagrangian frame moving with the mean local velocity $\nu_0(x)$ of the NESS. The Lagrangian-frame functions are obtained by replacing in the definitions (1.1) the time-independent observables $O^a(x)$ by the time-dependent ones $O^a(t, x)$ that evolve according to the advection equation

$$\partial_t O^a(t, x) + \nu_0(x) \cdot \nabla O^a(t, x) = 0, \quad (1.4)$$

i.e. are frozen in the Lagrangian frame. In the equilibrium, the mean local velocity $\nu_0(x)$ vanishes and the MFDT (1.3) becomes the FDT (1.2).

The other goal of the present work is to explain how the MFDT (1.3) may be obtained from more general fluctuation relations by restricting them to the regime close to NESS, similarly as for the case of the equilibrium FDT. Before doing that, we recall different fluctuation relations holding arbitrarily far from stationarity and equilibrium in Langevin systems and their perturbations. Our discussion follows with minor modifications the recent exposition [8].

The general results presented in this paper apply, in particular, to two types of one-dimensional systems with NESS that we shall call type 1 and type 2. The type 1 systems undergo an overdamped Langevin dynamics described by the stochastic differential equation (SDE) on the line:

$$\dot{x} = -\partial_x H(x) + \zeta \quad (1.5)$$

with a Hamiltonian $H(x) = ax^k + \dots$, for odd $k \geq 3$ and the white noise ζ_t with the covariance

$$\langle \zeta_t \zeta_s \rangle = 2\beta^{-1}\delta(t - s). \quad (1.6)$$

The Gibbs density $e^{-\beta H}$ is not normalizable here, so it does not give rise to an invariant probability measure. In fact, for $a > 0$, the process x_t solving equation (1.5) escapes in finite time to $-\infty$. It has, however, a realization with trajectories that reappear

immediately from $+\infty$, see appendix A. Such a resurrecting process has a non-Gibbsian invariant probability measure with constant probability flux. The processes of that kind arise naturally in the context of simple hydrodynamical and disordered systems that we recall below. The reader more interested in the abstract discussion of NESS for Langevin dynamics may skip the following two paragraphs.

The hydrodynamical model giving rise to equation (1.5) with a cubic Hamiltonian describes particles with inertia moving in one-dimensional Kraichnan's random velocities [31, 18]. Such velocities $v_t(r)$ form a Gaussian ensemble with mean zero and covariance:

$$\langle v_t(r)v_s(r') \rangle = \delta(t-s)D(r-r'). \quad (1.7)$$

The evolution of the inertial particles is described by the SDE [4]:

$$\dot{r} = u, \quad \dot{u} = \frac{1}{\tau}(-u + v_t(r)), \quad (1.8)$$

where τ is the Stokes time measuring the time delay of the particles relative to the flow motion. The separation between two infinitesimally close trajectories satisfies then the equations

$$\frac{d}{dt}\delta r = \delta u, \quad \frac{d}{dt}\delta u = \frac{1}{\tau}(-\delta u + \delta r \partial_r v_t(r)), \quad (1.9)$$

where one may replace $(1/\tau)\partial_r v_t(r)$ on the right-hand side by a white noise ζ_t with the covariance (1.6) for $\beta^{-1} = -(1/2\tau^2)\partial_r^2 D(0)$. For the ratio $x = \delta u/\delta r$, one then obtains the SDE:

$$\dot{x} = -x^2 - \frac{1}{\tau}x + \zeta \quad (1.10)$$

of the form (1.5) for $H(x) = \frac{1}{3}x^3 + (1/2\tau)x^2$. The resurrecting solutions x_t jumping instantaneously from $-\infty$ to $+\infty$ correspond here to the solutions for $(\delta r, \delta u)$, where δr passes through zero with a non-vanishing speed, i.e. to the crossing of close particle trajectories with faster particles overcoming slower ones (this is allowed in this model of a dilute particle suspension with no pressure and no back-reaction on the flow [17]). The top Lyapunov exponent for the random dynamical system (1.8) is obtained as the mean value of x (which is the temporal logarithmic derivative of $|\delta r|$) in the invariant non-Gibbsian probability measure of the resurrecting process [46].

The above story is a variation of a much older story [22, 38] of the one-dimensional Anderson localization in the stationary Schrödinger equation:

$$-\frac{d^2}{dr^2}\psi + V\psi = E\psi \quad (1.11)$$

with a δ -correlated potential $V(r)$. For $x = ((d/dr)\psi)/\psi$, one obtains the equation

$$\frac{d}{dr}x = -x^2 - E + V \quad (1.12)$$

that may be viewed as a stochastic evolution equation (1.5) with $H(x) = \frac{1}{3}x^3 + Ex$ and $\zeta = V$ if r is replaced by t . The resurrecting trajectories correspond here to wavefunctions with nodes. The invariant measure with constant flux for such an SDE was already used

in [22]. The substitution $x \rightarrow x + 1/2\tau$, $E \rightarrow -1/4\tau^2$, $V \rightarrow \zeta$ turns equation (1.12) into equation (1.10) (provided that one replaces r by t).

The type 2 one-dimensional systems with NESS covered by our discussion are obtained by adding a non-conservative force to the Langevin dynamics. More specifically, we shall consider a particle which moves on a circle according to the SDE:

$$\dot{x} = -\partial_x H(x) + G(x) + \zeta, \quad (1.13)$$

where, as before, ζ_t is the white noise with covariance (1.6). The above dynamics pertains again to the overdamped regime where it is the particle velocity rather than the particle acceleration that is proportional to the force. The angular coordinate x will be taken modulo 2π . We shall assume that $H(x + 2\pi) = H(x)$ and $G(x + 2\pi) = G(x)$ but $\int_0^{2\pi} G(x)dx \neq 0$ so that the force G is not a gradient and it drives the system out of equilibrium. Equation (1.13) was used, for example, to describe the motion of a colloidal particle in an optical trap [43]. It was discussed recently in [40] in a context similar to the one of this work.

The present paper is organized as follows. Section 2, returns to the discussion of stationary Langevin diffusion processes, presenting more details on the one-dimensional systems with explicit non-Gibbsian invariant measures [8, 40]. For such systems, we examine in section 3 the simplest fluctuation–response relation that describes the change of the invariant measure under a small time-independent variation of the Hamiltonian. In section 4, we prove the MFDT (1.3) that holds around NESS of the Langevin-type dynamics, in particular, in the one-dimensional cases with explicit invariant measures. Section 5 is devoted to a brief presentation of general fluctuation relations for SDEs [32, 37, 24, 33, 8]. These are specified for the Langevin systems under consideration in section 6. In particular, we describe the Crooks detailed fluctuation relation [12] and the Hatano–Sasa [24] version of the Jarzynski equality [29, 30], both holding arbitrarily far from stationarity. In section 7, we return to the MFDT, showing that it may be viewed as a limiting case around the stationary situation of the Crooks transient fluctuation relation or, in a special case, of the Jarzynski–Hatano–Sasa equality. Finally, after brief conclusions, in appendix A, we collect some facts about the one-dimensional processes with explosion. Appendix B illustrates the MFDT by an explicit calculation for the Langevin particle driven by a constant force along a circle.

2. NESS in Langevin processes

The general stationary dynamics that we consider is described by the Langevin equation in d dimensions with an external force:

$$\dot{x}^i = -\Gamma^{ij}\partial_j H(x) + \Pi^{ij}\partial_j H(x) + G^i(x) + \zeta^i, \quad (2.1)$$

where Γ is a constant non-negative matrix and Π an antisymmetric one, H is the Hamiltonian, G the external force and the white noise ζ_t has the covariance

$$\langle \zeta_t^i \zeta_s^j \rangle = 2\beta^{-1}\Gamma^{ij}\delta(t - s). \quad (2.2)$$

The deterministic force $-\Gamma\nabla H$ decreases the energy, driving the solution towards the minimum of H , if it exists, whereas the noise mimics the effect of a thermal bath. We have added the Hamiltonian force $\Pi\nabla H$ that preserves the energy in order to cover systems

governed by Langevin–Kramers equations [32] or Fermi–Pasta–Ulam chains [15]. The generator L of the process x_t satisfying the SDE (2.1) is defined by the relation

$$\partial_t \langle f(x_t) \rangle = \langle (Lf)(x_t) \rangle. \quad (2.3)$$

It is a second-order differential operator:

$$L = ((-\Gamma + \Pi)(\nabla H) + G) \cdot \nabla + \beta^{-1} \nabla \cdot \Gamma \nabla \quad (2.4)$$

in the vector notation, with the formal adjoint

$$L^\dagger = -\nabla \cdot ((-\Gamma + \Pi)(\nabla H) + G) + \beta^{-1} \nabla \cdot \Gamma \nabla. \quad (2.5)$$

The transition probabilities $P_t(x, dy)$ of the process satisfy the evolution equation:

$$\partial_t P_t(x, dy) = L_x P_t(x, dy), \quad (2.6)$$

with the subscript in L_x indicating that L acts on the variable x . The dynamics of the mean instantaneous density ϱ_t of the process x_t is generated by the adjoint operator L^\dagger and takes the form of the continuity equation

$$\partial_t \varrho_t = L^\dagger \varrho_t = -\nabla \cdot j_t \quad (2.7)$$

with the current⁴

$$j_t = (-\Gamma(\nabla H) + \Pi(\nabla H) + G - \beta^{-1}(\Gamma - \Pi)\nabla)\varrho_t. \quad (2.8)$$

Following [24], let us introduce the mean local velocity $\nu_t = \varrho_t^{-1} j_t$. With the use of the velocity field, the above continuity equation may be rewritten in the hydrodynamical form as the advection equation for the density ϱ_t :

$$(\partial_t + \nabla \cdot \nu_t)\varrho_t = 0, \quad (2.9)$$

stating that ϱ_t is annihilated by the convective derivative or, in other words, that it evolves as the density of Lagrangian particles whose trajectories obey the ordinary differential equation:

$$\dot{x} = \nu_t(x). \quad (2.10)$$

For an invariant density, the corresponding current is conserved: $\nabla \cdot j = 0$. If for the density ϱ the current j itself vanishes then one says that the dynamics (2.2) satisfies the detailed balance relative to ϱ . The detailed balance holds relative to the Gibbs density $e^{-\beta H}$ if $G = 0$ (this was ensured by the addition of the term $\beta^{-1} \Pi \nabla \varrho$ to the current). When $G \neq 0$, the invariant density is not known explicitly, in general, even if it exists. There are, however, special cases of processes satisfying the SDE (2.2), where one may obtain an analytic formula for a non-Gibbsian invariant density. Let us list two examples.

⁴ For convenience, we have included in the current the term $\beta^{-1} \Pi \nabla \varrho$.

2.1. NESS for resurrecting processes

The first, type 1, example is obtained for the Langevin equation on the line. In this case, any force is a gradient so that, upon setting for simplicity $\Gamma = 1$, the dynamical equation becomes the SDE (1.5) with the covariance of the white noise given by (1.6). The detailed balance holds here relative to the Gibbs density $e^{-\beta H}$ since the corresponding current vanishes. Such a density may, however, be not normalizable, hence not leading to an invariant probability measure. Let us look closer at various possibilities by considering the case of a polynomial Hamiltonian with the highest degree term equal to ax^k [8]. For $k = 0$ or $k = 1$, the solution x_t of equation (1.5) is, up to a linear change of variables, a Brownian motion, which does not have an invariant probability measure. For even $k \geq 2$ and $a > 0$, the Gibbs measure $\mu(dx) = Z^{-1}e^{-\beta H(x)}dx$ provides the unique invariant probability measure of the process x_t . If $a < 0$, however, then the Gibbs density is not normalizable. In this case the process escapes with probability 1 to $\pm\infty$ in a finite time and it has no invariant probability measure. For odd $k \geq 3$, the Gibbs density $e^{-\beta H(x)}$ is also not normalizable. Changing eventually x to $-x$, we may assume that $a > 0$. In this case, the process x_t escapes in finite time to $-\infty$ but it has a realization with the trajectories that reappear immediately from $+\infty$, as discussed in appendix A. Such a resurrecting process has a unique invariant probability measure:

$$\mu(dx) = \frac{1}{Z} \left(\int_{-\infty}^x e^{\beta H(y)} dy \right) e^{-\beta H(x)} dx \equiv \varrho_H(x) dx, \quad (2.11)$$

where Z is the (positive) normalization constant. The density ϱ_H of μ behaves as $(Za\beta kx^{k-1})^{-1}$ when $x \rightarrow \pm\infty$, see the estimate (A.17) in appendix A. It corresponds to the constant current $j = -(\beta Z)^{-1}$ with the flux towards negative x . The situation provides one of the simplest examples of NESS. In particular, the inertial particle in the one-dimensional Kraichnan flow and the Anderson localization in the one-dimensional δ -correlated potential lead naturally to the resurrecting processes corresponding to $k = 3$, as discussed in section 1.

2.2. NESS for forced diffusions on circle

The second, type 2, model with an explicit analytic expression for the invariant non-Gibbsian measure is the perturbed one-dimensional Langevin equation (1.13) on the unit circle. Now, the unique invariant probability measure is given by the formula [42, 25, 40]

$$\mu(dx) = \frac{1}{Z} \left(\int_0^{2\pi} e^{\beta U(x,y)} dy \right) e^{-\beta H(x)} dx \equiv \varrho_H(x) dx \quad (2.12)$$

for $0 \leq x \leq 2\pi$, with

$$U(x, y) = H(y) + \theta(x - y) \int_y^x G(z) dz + \theta(y - x) \left(\int_0^x G(z) dz + \int_y^{2\pi} G(z) dz \right). \quad (2.13)$$

Also here the density ϱ_H of the measure μ corresponds to a constant probability current:

$$j = \frac{1}{\beta Z} \left(e^{\beta \int_0^{2\pi} G(z) dz} - 1 \right). \quad (2.14)$$

In the following, we shall see how the presence of the probability flux in NESS deforms the usual fluctuation relations.

3. Modified fluctuation–response relation

As a warm-up, let us see what is the form taken by the most elementary fluctuation–response relation [36] in the one-dimensional systems with NESS that we discussed above. The set-up of the fluctuation–response relation is as follows. One prepares the system in the far past in the invariant state with probability density ϱ_H which is given by equations (2.11) and (2.12) for the type 1 and type 2 systems, respectively. At $t = 0$, the Hamiltonian H is perturbed by a small time-independent potential V (vanishing sufficiently fast when $x \rightarrow \pm\infty$ for type 1 and periodic for type 2), leading to the change $H \mapsto H' = H + V$. The system evolves then and converges toward the new steady state with the probability density $\varrho_{H'}$. The fluctuation–response relation compares the initial and the final averages of an observable $O_t \equiv O(x_t)$:

$$\langle O_0 \rangle = \int O(x) \varrho_H(x) dx \quad \text{and} \quad \langle O_\infty \rangle = \int O(x) \varrho_{H'}(x) dx. \quad (3.1)$$

By a straightforward differentiation of the explicit formulae for the invariant densities, one obtains the identity

$$\langle O_\infty \rangle = \langle O_0 \rangle - \beta[\langle O_0(V_0 - \widehat{V}_0) \rangle - \langle O_0 \rangle \langle V_0 - \widehat{V}_0 \rangle], \quad (3.2)$$

up to terms of the second order in V , where

$$\widehat{V}(x) = \frac{\int_{-\infty}^x V(y) e^{\beta H(y)} dy}{\int_{-\infty}^x e^{\beta H(y)} dy} \quad \text{and} \quad \widehat{V}(x) = \frac{\int_0^{2\pi} V(y) e^{\beta U(x,y)} dy}{\int_0^{2\pi} e^{\beta U(x,y)} dy} \quad (3.3)$$

for systems of type 1 and type 2, respectively. Equation (3.2) deforms the usual fluctuation–response relation around the Gibbs state [36] by replacing V by $(V - \widehat{V})$ with the averaged potential \widehat{V} dependent on the initial Hamiltonian H . Note that, in the type 2 case, \widehat{V} is again periodic.

4. Modified fluctuation–dissipation theorem

Coming back to the general case, let us consider a system prepared at negative times in the steady state of the stationary Langevin dynamics (2.1). This forces the time zero value x_0 of the corresponding process to be distributed according to the invariant probability measure $\mu_0(dx) = \varrho_0(x) dx$. At $t = 0$, one switches on a non-stationary perturbation, taking the Hamiltonian for the positive times to be equal to

$$H_t(x) = H(x) - \sum_a h_{a,t} O^a(x), \quad (4.1)$$

where $h_{a,t}$ carry the time dependence and functions $O^a(x)$ (the ‘observables’) are supposed to vanish sufficiently fast when $|x| \rightarrow \infty$ or, for type 2 systems, to be periodic. We denote by $\langle \mathcal{F} \rangle_h$ the corresponding expectation, with $\langle \mathcal{F} \rangle_0$ referring to the non-perturbed case. The expression

$$\langle \mathcal{F} R_t^a \rangle_0 = \left. \frac{\delta}{\delta h_{a,t}} \right|_{h=0} \langle \mathcal{F} \rangle_h. \quad (4.2)$$

defines the response correlations. To shorten further the notations, let us set

$$\langle O_t^a R_s^b \rangle_0 \equiv \mathcal{R}^{ab}(t-s), \quad \langle O_t^a O_s^b \rangle_0 \equiv \mathcal{C}^{ab}(t-s), \quad \theta(t-s) \langle O_t^a B_s^b \rangle_0 \equiv \mathcal{B}^{ab}(t-s) \quad (4.3)$$

for $O_t \equiv O(x_t)$ and the induced observable

$$B^b = \varrho_0^{-1} j_0 \cdot \nabla O^b \quad (4.4)$$

with j_0 standing for the current (2.8) corresponding to the invariant density ϱ_0 . Note that, for the one-dimensional NESS with constant probability current j_0 , the observable $B^b = j_0 \varrho_0^{-1} \partial_x O^b$ has the constant probability flux as an explicit factor. Remark that, by causality, the response function $\mathcal{R}^{ab}(t-s)$ vanishes for $s \geq t$. We shall show the following modified version of the fluctuation–dissipation theorem (MFDT) holding for $t > s$:

$$\beta^{-1} \mathcal{R}^{ab}(t-s) = \partial_s \mathcal{C}^{ab}(t-s) - \mathcal{B}^{ab}(t-s). \quad (4.5)$$

The second term on the right-hand side of equation (4.5) is new compared to the FDT around the equilibrium steady state. Indeed, in the equilibrium case, the external force $G = 0$ and $\varrho_0 = Z^{-1} e^{-\beta H}$ is the normalized Gibbs factor with $j_0 = 0$ so that $B^b = 0$ and the MFDT (4.5) reduces to the standard equilibrium form (1.2).

4.1. Lagrangian-frame interpretation

What the formula (4.5) for the response function means is better understood by rewriting it with the explicit form of the right-hand side as

$$\begin{aligned} \beta^{-1} \mathcal{R}^{ab}(t-s) &= \int dx O^b(x) \varrho_0(x) \partial_s P_{t-s}(x, dy) O^a(y) \\ &\quad - \int dx (j_0 \cdot \nabla O^b)(x) P_{t-s}(x, dy) O^a(y) \\ &= \int dx O^b(x) (\partial_s + \nabla \cdot \nu_0(x)) \int \varrho_0(x) P_{t-s}(x, dy) O^a(y), \end{aligned} \quad (4.6)$$

where $P_t(x, dy)$ denotes the stationary transition probabilities of the unperturbed process and we have integrated by parts to obtain the second equality, setting $\varrho_0^{-1} j_0 \equiv \nu_0$. Note that the time derivative ∂_s of the equilibrium relation has been replaced by the convective derivative $\partial_s + \nabla \cdot \nu_0(x)$ which acts on the first component of the joint probability density function of the time s and time t values of the stationary process x_t . This suggests that the MFDT (4.5) should take the equilibrium form in the Lagrangian frame moving with the stationary mean local velocity $\nu_0(x)$.

To render this interpretation more transparent, let us replace the time-independent observables $O^a(x)$ by the time-dependent ones $O^a(t, x)$ evolving according to the advection equation (1.4). We shall define the Lagrangian-frame response function and correlation function by

$$\mathcal{R}_L^{ab}(t, s) = \frac{\delta}{\delta h_{b,s}} \Big|_{h=0} \langle O_t^a(t) \rangle_h, \quad \mathcal{C}_L^{ab}(t, s) = \langle O_t^a(t) O_s^b(s) \rangle_0, \quad (4.7)$$

where $\langle - \rangle_h$ denotes now the expectation referring to the Hamiltonian $H_t(x) = H(x) - \sum_a h_{t,a} O^a(t, x)$ and $O_t^a(t)$ with the double time dependence is the shorthand notation for

$O(t, x_t)$. Writing the MFDT (4.5) with the explicit right-hand side given by equation (4.6) for the observables $O^a(t, x)$, one casts this relation into the form

$$\begin{aligned} \beta^{-1} \mathcal{R}_L^{ab}(t, s) &= \int dx O^b(s, x) \varrho_0(x) \partial_s P_{t-s}(x, dy) O^a(t, y) \\ &\quad - \int dx \varrho_0(x) (\nu_0(x) \cdot \nabla O^b(s, x)) P_{t-s}(x, dy) O^a(t, y) \\ &= \partial_s \int dx O^b(s, x) \varrho_0(x) P_{t-s}(x, dy) O^a(t, y) = \partial_s \mathcal{C}_L^{ab}(t, s), \end{aligned} \quad (4.8)$$

where the last equality follows from the advection equation (1.4). This proves the identity (1.3) announced in the introduction. We should stress that, in spite of the similarity between that relation and the equilibrium FDT (1.2), in general it is not true that the dynamical process x_t viewed in the Lagrangian frame of the velocity field ν_0 is governed by an equilibrium Langevin equation, although this is what happens in the simple example considered in appendix B.

For the Langevin process on the circle, a fluctuation–dissipation relation for velocities similar to (4.5) was discussed in [44], see equation (11) there, with the interpretation similar in spirit, but not in form, to the above one, see the subsequent discussion there. One of the consequences of the fluctuation–dissipation relation of [44] linking the effective diffusivity and mobility was checked experimentally in [5], see also [26].

It is sometimes more interesting, especially for applications, to re-express the fluctuation–dissipation relations in terms of the frequency space quantities. Let

$$\hat{\mathcal{C}}^{ab}(\omega) = \int_{-\infty}^{\infty} e^{i\omega(t-s)} \mathcal{C}^{ab}(t-s) dt = \int_s^{\infty} e^{i\omega(t-s)} \mathcal{C}^{ab}(t-s) dt + \int_s^{\infty} e^{-i\omega(t-s)} \mathcal{C}^{ba}(t-s) dt, \quad (4.9)$$

$$\hat{\mathcal{R}}^{ab}(\omega) = \int_s^{\infty} e^{i\omega(t-s)} \mathcal{R}^{ab}(t-s) dt, \quad \hat{\mathcal{B}}^{ab}(\omega) = \int_s^{\infty} e^{i\omega(t-s)} \mathcal{B}^{ab}(t-s) dt. \quad (4.10)$$

Note that $\hat{\mathcal{R}}^{ab}(\omega)$ measures the response to a small time-dependent potential of frequency ω . The MFDT (4.5) is equivalent to the relation

$$i\omega \hat{\mathcal{C}}^{ab}(\omega) = \beta^{-1} (\hat{\mathcal{R}}^{ab}(\omega) - \hat{\mathcal{R}}^{ba}(-\omega)) + \hat{\mathcal{B}}^{ab}(\omega) - \hat{\mathcal{B}}^{ba}(-\omega) \quad (4.11)$$

in the frequency space.

In general, assuming that the transition probabilities $P_t(x, dy)$ converge at long times to the invariant measure, all three terms of equation (4.5) tend to zero when $(t-s) \rightarrow \infty$. Mimicking the idea employed with success for disordered systems [13, 14, 7], their relative proportions, or the relative proportions of the corresponding terms in equation (4.11), could be used to define dynamical temperatures that would, in general, depend also on the observables involved. We discuss those proportions in a simple case of the Langevin dynamics on a circle with a constant force in appendix B.

We propose three derivations of the result (4.5): the first one direct, that we shall present now, and the next two ones from the general fluctuation relations that will be discussed in the subsequent sections.

4.2. Direct derivation

The beginning of the argument is quite standard, see, e.g., [1] or section 2.3.2 of [41]. By the definition of the response correlations

$$\mathcal{R}^{ab}(t-s) = \frac{\delta}{\delta h_{b,s}} \Big|_{h=0} \int O^a(y) \varrho_t(y) dy, \quad (4.12)$$

where ϱ_t is the density obtained by the perturbed dynamical evolution (2.7) from ϱ_0 . Using the explicit form (2.8) of the current, one obtains by the first-order perturbation the relation

$$\varrho_t(y) dy = \varrho_0(y) dy - \sum_b \int_0^t h_{b,s} ds \int dx [\nabla \cdot ((\Gamma - \Pi)(\nabla O^b) \varrho_0)](x) P_{t-s}(x, dy) + \mathcal{O}(h^2). \quad (4.13)$$

Consequently, for $t > s$,

$$\mathcal{R}^{ab}(t-s) = - \int dx [\nabla \cdot ((\Gamma - \Pi)(\nabla O^b) \varrho_0)](x) P_{t-s}(x, dy) O^a(y). \quad (4.14)$$

Now, a straightforward although somewhat tedious algebra shows that

$$\beta^{-1} \nabla \cdot ((\Gamma - \Pi)(\nabla O^b) \varrho_0) = L^\dagger(O^b \varrho_0) + \nabla \cdot (O^b j_0) = L^\dagger(O^b \varrho_0) + j_0 \cdot \nabla O^b, \quad (4.15)$$

where the adjoint generator L^\dagger is given by equation (2.5) and in the last equality we have used the conservation of the current j_0 . Substituting this identity into equation (4.14) and integrating the term with L^\dagger by parts, we obtain the relation

$$\beta^{-1} \mathcal{R}^{ab}(t-s) = - \int dx [(O^b \varrho_0)(x) L_x + (j_0 \cdot \nabla O^b)(x)] P_{t-s}(x, dy) O^a(y) \quad (4.16)$$

which, together with equation (2.6), implies the MFDT (4.5).

5. General fluctuation relations

In [8], two of us discussed arbitrary diffusion processes in d dimensions defined by the Stratonovich SDE:

$$\dot{x} = u_t(x) + v_t(x), \quad (5.1)$$

where $u_t(x)$ is a time-dependent deterministic vector field (a drift) and $v_t(x)$ is a Gaussian random vector field with mean zero and covariance:

$$\langle v_t^i(x) v_s^j(y) \rangle = \delta(t-s) D_t^{ij}(x, y). \quad (5.2)$$

The Langevin equation (2.1) provides a special example of such an SDE. For the processes solving equation (5.1), we showed, combining the Girsanov and the Feynman-Kac formulae, a detailed fluctuation relation (DFR):

$$\mu_0(dx) P_T(x; dy, dW) e^{-W} = \mu'_0(dy^*) P'_T(y^*; dx^*, d(-W)), \quad (5.3)$$

where

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

- (2) $\mu'_0(dx) = \varrho'_0(x) dx$ 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 \mathcal{W}_T of the same process on the interval $[0, T]$ (described later),
- (4) $P'_T(x; dy, dW)$ is the similar joint probability distribution for the backward process.

The key behind the DFR is the action of a time inversion on the forward system. First, such an inversion acts on time and space by an involution

$$(t, x) \mapsto (T - t, x^*). \quad (5.4)$$

The above involution may be extended to the action $x \mapsto \tilde{x}$, on trajectories by the formula $\tilde{x}_t = x_{T-t}^*$ and, further, to the action on functionals of trajectories $\mathcal{F} \mapsto \tilde{\mathcal{F}}$ by setting $\tilde{\mathcal{F}}[x] = \mathcal{F}[\tilde{x}]$. Second, to recover a variety of fluctuation relations discussed in the literature [32, 37, 11, 12, 28, 45, 9], we allow for a non-trivial behavior of the drift $u_t(x)$ under the time inversion, dividing it into two parts, $u = u_+ + u_-$, with u_+ transforming as a vector field under the space-time involution (5.4) and u_- as a pseudo-vector field, i.e. defining

$$u_{T-t, \pm}^i(x^*) = \pm(\partial_k x^{*i})(x) u_{t, \pm}^k(x), \quad u' = u'_+ + u'_-. \quad (5.5)$$

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

$$\dot{x} = u'_t(x) + v'_t(x) \quad (5.6)$$

and is, in general, different from the naive time inversion \tilde{x}_t of the forward process. The functional \mathcal{W}_T involved in the DFR depends explicitly on the densities ϱ_0 and ϱ_T , where the latter is defined by the relation $\mu'_0(dx^*) = \varrho_T(x) dx$:

$$\mathcal{W}_T = -\Delta_T \ln \varrho + \int_0^T \mathcal{J}_t dt \quad (5.7)$$

with the notation $\Delta_T \ln \varrho \equiv \ln \varrho_T(x_T) - \ln \varrho_0(x_0)$. In the above formula

$$\mathcal{J}_t = 2\hat{u}_{t,+}(x_t) \cdot d_t^{-1}(x_t)(\dot{x}_t - u_{t,-}(x_t)) - (\nabla \cdot u_{t,-})(x_t), \quad (5.8)$$

where $d_t^{ij}(x) = D_t^{ij}(x, x)$ and $\hat{u}_{t,+}^i(x) = u_{t,+}^i(x) - \frac{1}{2} \partial_{y^j} D_t^{ij}(x, y)|_{y=x}$. The time integral in equation (5.7) should be taken in the Stratonovich sense. The functional \mathcal{W}'_T for the backward process is defined in the same way, setting $\mu_0(dx^*) = \varrho'_T(x) dx$. One has the relation

$$\mathcal{W}'_T = -\tilde{\mathcal{W}}_T, \quad (5.9)$$

where the tilde denotes the involution of trajectory functionals introduced before.

The quantity \mathcal{J}_t has the interpretation of the rate of entropy production in the environment modeled by the thermal noise. When the density ϱ_T coincides with the density obtained from ϱ_0 by the dynamical evolution (2.7), where now the current

$$j_t = (\hat{u}_t - \frac{1}{2} d_t \nabla) \varrho_t \quad \text{with } \hat{u} = \hat{u}_+ + u_- \quad (5.10)$$

then the first contribution $-\Delta_T \ln \rho$ to \mathcal{W}_T may be interpreted as the change in the instantaneous entropy of the process. In this case, the functional \mathcal{W}_T becomes equal to the overall entropy production. Keep in mind that this is a fluctuating quantity which, in general, may take both positive and negative values.

The DFR (5.3) holds even if the measures μ_0 and μ'_0 are not normalized, or even not normalizable. For normalized initial measures, we denote by

$$\langle \mathcal{F} \rangle = \int \mathcal{F}[x] \mathcal{M}[dx] \quad \text{and} \quad \langle \mathcal{F}' \rangle = \int \mathcal{F}[x] \mathcal{M}'[dx] \quad (5.11)$$

the averages over the realizations of the forward and the backward process $x_t, 0 \leq t \leq T$, with x_0 distributed according to the probability measure μ_0 and μ'_0 , respectively. $\mathcal{M}[dx]$ and $\mathcal{M}'[dx]$ stand for the corresponding measures over the space of trajectories. One of the immediate consequences of the DFR (5.3) is the identity

$$\langle e^{-\mathcal{W}_T} \rangle = 1 \quad (5.12)$$

obtained by the integration of both sides. This is a generalization of the celebrated Jarzynski equality [29, 28]. The relation (5.12) implies the inequality $\langle \mathcal{W}_T \rangle \geq 0$ that has the form of the second law of thermodynamics stating the positivity of the average entropy production. The Jarzynski equality, however, provides also information about an exponential suppression of the events with negative entropy production in non-equilibrium systems for which $\langle \mathcal{W}_T \rangle > 0$.

With a little more work [8] involving a multiple superposition of the FDR (5.3), the latter may be cast into the Crooks form [12]:

$$\langle \mathcal{F} e^{-\mathcal{W}_T} \rangle = \langle \tilde{\mathcal{F}}' \rangle. \quad (5.13)$$

In terms of the trajectory measures $\mathcal{M}[dx]$ and $\mathcal{M}'[dx]$, this becomes the identity

$$\tilde{\mathcal{M}}'[dx] = e^{-\mathcal{W}_T[x]} \mathcal{M}[dx], \quad (5.14)$$

where $\tilde{\mathcal{M}}'[dx] = \mathcal{M}'[d\tilde{x}]$. Equation (5.14) permits us to interpret the expectation of \mathcal{W}_T as the relative entropy of the trajectory measures:

$$\langle \mathcal{W}_T \rangle = \int \ln \frac{\mathcal{M}[dx]}{\tilde{\mathcal{M}}'[dx]} \mathcal{M}[dx] \equiv S(\mathcal{M} | \tilde{\mathcal{M}}'), \quad (5.15)$$

in line with the above entropic interpretation of the functional \mathcal{W}_T .

6. Fluctuation relations in Langevin dynamics

Let us specify the DFR (5.3) to the case of Langevin dynamics (2.2) with, possibly, time-dependent Hamiltonian H and external force G . A canonical choice of the time inversion for such a system takes

$$u_+ = -\Gamma \nabla H, \quad u_- = \Pi \nabla H + G \quad (6.1)$$

and a linear involution $x^* = Rx$ such that $R\Gamma R^t = \Gamma$ and $R\Pi R^t = -\Pi$ [8]. For example, for the Langevin–Kramers dynamics in the phase space, the usual R changes the sign of momenta. The backward dynamics has now the same form as the forward one, with the time-reversed Hamiltonian $H'_t(x) = H_{T-t}(Rx)$ and the time-reversed

external force $G'_t(x) = -RG_{T-t}(Rx)$. In this case, we may use the Gibbs densities $\varrho_t(x) = Z_t^{-1}e^{-\beta H_t(x)} = \varrho'_{T-t}(Rx)$ at the initial and final times, with Z_t standing for the partition function $\int e^{-\beta H_t(x)} dx$ if the integral is finite and $Z_t = 1$ otherwise. A straightforward calculation gives

$$\mathcal{W}_T = \ln(Z_T/Z_0) + \int_0^T (\beta \partial_t H_t + \beta G_t \cdot \nabla H_t - \nabla \cdot G_t)(x_t) dt. \quad (6.2)$$

For normalizable Gibbs factors this is often called the ‘dissipative work’. \mathcal{W}'_T is given by the same expression, with H_t and G_t replaced by H'_t and G'_t .

Another useful choice of the time inversion is based on the eventual knowledge of the densities ϱ_t corresponding to the conserved currents with $\nabla \cdot j_t = 0$. Note that such densities would be left invariant by the evolution (2.7) if the time dependence of the Hamiltonian and of the external force were frozen to the instantaneous values H_t and G_t . One takes

$$u_+ = \beta^{-1} \Gamma \nabla \ln \varrho, \quad u_- = -\Gamma \nabla (H + \beta^{-1} \ln \varrho) + \Pi \nabla H + G. \quad (6.3)$$

With the linear involution $x^* = Rx$ as above, the backward process has

$$u'_+ = \beta^{-1} \Gamma \nabla \ln \varrho', \quad u'_- = \Gamma \nabla (H' + \beta^{-1} \ln \varrho') + \Pi \nabla H' + G', \quad (6.4)$$

where $\varrho'_t(x) = \varrho_{T-t}(Rx)$ and H' and G' are as before. The current corresponding to the density ϱ'_t satisfies

$$j'_t(x) = -Rj_{T-t}(Rx). \quad (6.5)$$

It is also conserved. Such a time inversion (for $R = 1$) was considered in [24] and, more explicitly, in [9]. In [8], it was called the current reversal. The DFR (5.3) holds now for

$$\mathcal{W}_T = - \int_0^T (\partial_t \ln \varrho_t)(x_t) dt \quad (6.6)$$

and \mathcal{W}'_T given by the same expression with ϱ_t replaced by ϱ'_t . The Jarzynski equality (5.12) for this case (assuming that ϱ_t are normalized) was first proven by Hatano and Sasa in [24]. Note that, if $G = 0$, then the current corresponding to the densities $\varrho_t = Z_t^{-1}e^{-\beta H_t}$ is conserved and with this choice of ϱ_t the two-time inversions coincide. In particular, for $G = 0$ and the time-independent $H_t \equiv H$, the functional \mathcal{W}_T identically vanishes and the DFR reduces to the equality

$$e^{-\beta H(x)} dx P_T(x, dy) = e^{-\beta H(y)} dy P'_T(y^*, dx^*) \quad (6.7)$$

which is a more global version of the detailed balance relation. On the right-hand side, one may replace P'_T by P_T for $\Pi = 0$ and $x^* \equiv x$ because, in that case, the forward and backward processes have the same distribution.

6.1. Fluctuation relations for resurrecting processes

Let us consider the process solving the SDE (1.5), with $H_t(x) = ax^k + o(|x|^k)$ at large $|x|$ with odd $k \geq 3$ and $a > 0$. We admit a mild time dependence of $H_t(x)$ disappearing when $x \rightarrow \pm\infty$. The corresponding process still has a resurrecting version, as in the stationary case described in appendix A. Let us discuss first the canonical time inversion with the trivial involution $x^* = x$ leading to the backward process of the same type with $H_t(x)$ replaced by $H'_t(x) = H_{T-t}(x)$. The definition of the functional \mathcal{W}_T for the resurrecting process requires a little care in order to account for the contributions from the jumps from $-\infty$ to $+\infty$. This may be done by compactifying the line to a circle, writing $x = \cot \theta$ for θ modulo π . One has

$$\int_0^T \mathcal{J}_t dt = -\beta \int_0^T (\partial_x H_t)(x_t) \dot{x}_t dt = \int_0^T (\beta k a \cot^{k-1} \theta_t + \dots) \sin^{-2} \theta_t \dot{\theta}_t dt \quad (6.8)$$

and the integral diverges to $+\infty$ whenever θ_t passes from the negative to the positive values, i.e. whenever x_t jumps from $-\infty$ to $+\infty$. Upon taking $\varrho_t = e^{-\beta H_t}$, we infer that

$$\mathcal{W}_T = \begin{cases} \int_0^T (\partial_t H_t)(x_t) dt & \text{if } x_t \text{ has no rebirths for } 0 < t < T, \\ +\infty & \text{otherwise,} \end{cases} \quad (6.9)$$

and similarly for \mathcal{W}'_T . As a result, the contributions of the rebirths to the DFR (5.3) trivially decouple, reducing the latter to the identity

$$e^{-H_0(x)} dx P_T^0(x; dy, dW) e^{-W} = e^{-H_T(y)} dy P_T^0(x; dy, d(-W)) \quad (6.10)$$

between the joint distributions of the endpoints and of the functional $\mathcal{W}_T = \int_0^T (\partial_t H_t)(x_t)$ (usually called the ‘Jarzynski work’), or of its counterpart \mathcal{W}'_T , in the processes without rebirths. In the stationary case, equation (6.10) reduces to the detailed balance relation

$$e^{-\beta H(x)} dx P_t^0(x, dy) = e^{-\beta H(y)} dy P_t^0(y, dx), \quad (6.11)$$

which ensures upon integration over x that the process without rebirths preserves the infinite measure $e^{-\beta H(x)} dx$.

On the other hand, one could use for the same SDE with the resurrecting solution the current reversal based on the splitting

$$u_{t,+} = \beta^{-1} \partial_x \ln \varrho_{H_t}, \quad u_{t,-} = -\partial_x (H_t + \beta^{-1} \ln \varrho_{H_t}) \quad (6.12)$$

of the drift $-\partial_x H_t$, with the density ϱ_{H_t} given by equation (2.11). The use of the involution $x^* = -x$ leads then to the backward process solving the SDE (1.5) with the Hamiltonian $H_t(x)$ replaced by

$$H'_t(x) = H_{T-t}(-x) - 2\beta^{-1} \ln \left(\int_{-\infty}^{-x} e^{\beta H_{T-t}(y)} dy \right). \quad (6.13)$$

From the estimate (A.17) in appendix A, one infers that $H'_t(x) = ax^k + o(|x|^k)$ for large $|x|$ (we have used the non-trivial spatial involution to keep a positive). Hence H'_t is of the same type as the Hamiltonian H_t for the forward process. In this case, the functionals \mathcal{W}_T and \mathcal{W}'_T are given by equation (6.6) with $\varrho_t(x)$ replaced by $\varrho_{H_t}(x)$ and $\varrho_{H_{T-t}}(-x) = \varrho_{H'_t}(x)$,

respectively, with no extra contributions from the rebirths. In the stationary case, one has $\mathcal{W}_T = 0 = \mathcal{W}'_T$ and the DFR (5.3) reduces to the modified detailed balance relation

$$\varrho_H(x) dx P_T(x, dy) = \varrho_H(y) dy P'_T(-y, d(-x)). \quad (6.14)$$

The latter links the transition probabilities of the resurrecting forward and backward processes and ensures upon integration over x or y that those processes preserve the probability measures $\varrho_H(x) dx$ and $\varrho_H(-x) dx$, respectively.

6.2. Fluctuation relations for forced diffusions on circle

Consider the process satisfying the SDE (1.13) with periodic Hamiltonian $H(x) = H(x + 2\pi)$ and external force $G(x) = G(x + 2\pi)$, both possibly time-dependent. The use of the canonical time inversion with the trivial involution $x^* = x$ leads to the backward process of the same type with $H'_t = H'_{T-t}$ and $G'_t = -G'_{T-t}$. The functionals \mathcal{W}_T and \mathcal{W}'_T are given here by the formula (6.2).

On the other hand, the use of the current reversal with the densities ϱ_{H_t} of equation (2.12) and the trivial inversion $x^* = x$ leads to the backward process satisfying the same SDE with H_t replaced by

$$H'_t(x) = -H_{T-t}(x) - 2\beta^{-1} \ln \varrho_{H_{T-t}} \quad (6.15)$$

and G_t by G'_t as above. The functionals \mathcal{W}_T and \mathcal{W}'_T are given now by equation (6.6) with $\varrho_t(x)$ equal to $\varrho_{H_t}(x)$ and $\varrho_{H'_t}(x) = \varrho_{H_{T-t}}(x)$, respectively. They vanish in the stationary case when the DFR (5.3) reduces again to the modified detailed balance relation

$$\varrho_H(x) dx P_T(x, dy) = \varrho_H(y) dy P'_T(y, dx). \quad (6.16)$$

Note that, in general, to obtain the DFR for the probability distributions on the circle, one should sum both sides of the relation (5.3) pertaining to the motion on the line, over the shifts of x or y by $2\pi n$ with integer n (both summations amount to the same). To get the Jarzynski equality, one has to integrate the relation obtained this way over x and y from 0 to 2π . Another simple remark is that for the SDE (1.13) on the circle, one may always assume that the external force G is constant by changing $G(x)$ to $\bar{G} = (1/2\pi) \int_0^{2\pi} G(x) dx$ and by subtracting $\int_0^x (G(y) - \bar{G}) dy$ from $H(x)$. Such a change does not affect the DFR (5.3) obtained by the current reversal that uses only the invariant densities and it modifies in a simple way the DFR obtained from the canonical time inversion because in the latter we used the Gibbs measures for the initial and final distributions.

7. Fluctuation relations close to NESS

As promised, we shall show here that the MFDT (4.5), proven directly in section 4.1, may also be derived by reducing the Crooks' DFR for the current reversal, and, in a special case, the Jarzynski–Hatano–Sasa equality, to the situations close to NESS.

7.1. Reduction of Crooks' DFR to MFDT

Let us consider the DFR (5.13) for the Langevin dynamics (2.1) with $\mathcal{F} = O_t^a$ and $0 < t < T$, the backward dynamics determined by the current reversal and, for simplicity, the trivial space involution $x^* \equiv x$, see section 6. It is

$$\langle O_t^a e^{-W_T} \rangle = \langle O_{T-t}^a \rangle'. \quad (7.1)$$

We shall assume that the time dependence of the Hamiltonian is given by equation (4.1) with $h_{a,0} = 0 = h_{a,T}$, and that the external force G is time-independent. Let, as above, $\varrho_0(x) dx$ denote the invariant probability measure of the unperturbed process (assumed to exist). By ϱ_t , we shall denote now the normalized densities whose current j_t , given by equation (2.8), is conserved, i.e. such that

$$L^\dagger \varrho_t - \sum_a h_{t,a} \nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_t) = 0. \quad (7.2)$$

Expanding

$$\varrho_t = \varrho_0 + \sum_a h_{t,a} \varrho_1^a + \mathcal{O}(h^2), \quad (7.3)$$

we obtain from equation (7.2) the relation

$$L^\dagger \varrho_1^a = \nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_0) \quad (7.4)$$

whose unique solution

$$\varrho_1^a = (L^\dagger)^{-1} [\nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_0)] \quad (7.5)$$

is chosen by imposing the orthogonality of ϱ_1^a to the constant mode, required by the normalization of ϱ_t . For the current reversal, the functional \mathcal{W}_T is given by equation (6.6) so that

$$\begin{aligned} \mathcal{W}_T &= - \sum_a \int_0^T (\partial_t h_{t,a}) (\varrho_0^{-1} \varrho_1^a)_t dt + \mathcal{O}(h^2) \\ &= \sum_a \int_0^T h_{t,a} \partial_t \{ \varrho_0^{-1} (L^\dagger)^{-1} [\nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_0)] \}_t dt + \mathcal{O}(h^2), \end{aligned} \quad (7.6)$$

where we have integrated once by parts over t . The application of the operator $(\delta/\delta h_{b,s})|_{h=0}$ for $0 < s < t$ to both sides of equation (7.1) gives the identity

$$\mathcal{R}^{ab}(t-s) - \langle O_t^a \partial_s \{ \varrho_0^{-1} (L^\dagger)^{-1} [\nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_0)] \}_s \rangle_0 = 0. \quad (7.7)$$

We have used the fact that, by causality, the right-hand side of equation (7.1) does not give the contribution because the perturbation is concentrated around time $(T-s) > (T-t)$. From the relation (7.7), it follows that

$$\begin{aligned} \mathcal{R}^{ab}(t-s) &= \langle O_t^a \partial_s \{ \varrho_0^{-1} (L^\dagger)^{-1} [\nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_0)] \}_s \rangle_0 \\ &= \partial_s \int dx \{ (L^\dagger)^{-1} [\nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_0)] \}(x) P_{t-s}(x, dy) O^a(y) \\ &= - \int dx \{ (L^\dagger)^{-1} [\nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_0)] \}(x) L_x P_{t-s}(x, dy) O^a(y) \\ &= - \int dx [\nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_0)](x) P_{t-s}(x, dy) O^a(y), \end{aligned} \quad (7.8)$$

which is equation (4.14) above. The rest of the proof of the identity (4.5) goes as before.

7.2. Jarzynski–Hatano–Sasa equality and MFDT

The standard FDT around the equilibrium Langevin dynamics (2.1) without the external force may be obtained by expanding the Jarzynski equality (5.12) for the Hamiltonian (4.1) up to second order in h , see [8]. The Jarzynski equality may then be viewed as an extension of the FDT to the case of Hamiltonians with arbitrary time dependence driving the system far from equilibrium. The natural question is whether this picture may be generalized to the case of the modified FDT (4.5) holding around NESS. We shall show here that the answer is a qualified yes.

Let us expand to second order in h the Hatano–Sasa version of the Jarzynski equality (5.12) obtained from the Crooks’ DFR (7.1) for the current reversal by replacing O^a by 1. We shall need to know the form of the densities ϱ_t with conserved current, i.e. satisfying equation (7.2), to second order in h . One has

$$\varrho_t = \varrho_0 + \sum_a h_{t,a} \varrho_1^a + \sum_{a,b} h_{t,a} h_{t,b} \varrho_2^{ab} + \mathcal{O}(h^3) \quad (7.9)$$

with ϱ_{1a} as before, see equation (7.5), and

$$\varrho_2^{ab} = (L^+)^{-1} [\nabla \cdot ((\Gamma - \Pi)(\nabla O^a) \varrho_1^b)]. \quad (7.10)$$

Expanding, in turn, the functional \mathcal{W}_T given by equation (6.6) to second order, we obtain

$$\begin{aligned} \mathcal{W}_T = & - \sum_a \int_0^T (\partial_t h_{t,a}) (\varrho_0^{-1} \varrho_1^a)_t dt - \sum_{a,b} \int_0^T (\partial_t (h_{t,a} h_{t,b})) (\varrho_0^{-1} \varrho_2^{ab} - \frac{1}{2} \varrho_0^{-2} \varrho_1^a \varrho_1^b)_t dt \\ & + \mathcal{O}(h^3), \end{aligned} \quad (7.11)$$

and, further,

$$\begin{aligned} \langle e^{-\mathcal{W}_T} \rangle_h - 1 = & \sum_a \int_0^T (\partial_t h_{t,a}) \langle (\varrho_0^{-1} \varrho_1^a)_t \rangle_h dt \\ & + \sum_{a,b} \int_0^T (\partial_t (h_{t,a} h_{t,b})) \langle (\varrho_0^{-1} \varrho_2^{ab} - \frac{1}{2} \varrho_0^{-2} \varrho_1^a \varrho_1^b)_t \rangle dt \\ & + \frac{1}{2} \sum_{a,b} \int_0^T \int_0^T (\partial_t h_{t,a}) (\partial_s h_{s,b}) \langle (\varrho_0^{-1} \varrho_1^a)_t (\varrho_0^{-1} \varrho_1^b)_s \rangle_0 dt ds + \mathcal{O}(h^3). \end{aligned} \quad (7.12)$$

The second term on the right-hand side integrates to zero because of the stationarity of the unperturbed expectation and the boundary conditions $h_{0,a} = 0 = h_{T,a}$. Expanding the remaining perturbed expectation in the first term on the right-hand side, we infer that

$$\begin{aligned} \langle e^{-\mathcal{W}_T} \rangle_h - 1 = & \sum_{a,b} \int_0^T \int_0^T (\partial_t h_{t,a}) h_{s,b} \langle (\varrho_0^{-1} \varrho_1^a)_t R_s^b \rangle_0 dt ds \\ & + \frac{1}{2} \sum_{a,b} \int_0^T \int_0^T (\partial_t h_{t,a}) (\partial_s h_{s,b}) \langle (\varrho_0^{-1} \varrho_1^a)_t (\varrho_0^{-1} \varrho_1^b)_s \rangle_0 dt ds + \mathcal{O}(h^3) = 0, \end{aligned} \quad (7.13)$$

where the last equality follows from the (generalized) Jarzynski equality (5.12). The integration by parts and the causality permit us to conclude that, for $t > s$,

$$\partial_t \left\langle (\varrho_0^{-1} \varrho_1^a)_t R_s^b \right\rangle_0 = \partial_t \partial_s \left\langle (\varrho_0^{-1} \varrho_1^a)_t (\varrho_0^{-1} \varrho_1^b)_s \right\rangle_0, \quad (7.14)$$

or, integrating once over t , that

$$\left\langle (\varrho_0^{-1} \varrho_1^a)_t R_s^b \right\rangle_0 = \partial_s \left\langle (\varrho_0^{-1} \varrho_1^a)_t (\varrho_0^{-1} \varrho_1^b)_s \right\rangle_0 \quad (7.15)$$

(we used the fact that both sides vanish for $t = s$). Note that equation (7.15) stays true if we add to ϱ_1^a any multiple of ϱ_0 , so that we may drop the normalization condition $\int \varrho_1^a(x) dx = 0$. From equations (7.5) and (4.15), it follows then that we may take

$$\varrho_0^{-1} \varrho_1^a = \beta(O^a - \widehat{O}^a) \equiv \beta A^a, \quad (7.16)$$

where the dressed observable

$$\widehat{O}^a = -\varrho_0^{-1} (L^\dagger)^{-1} [j_0 \cdot \nabla O^a]. \quad (7.17)$$

The identity (7.15) becomes now the relation

$$\begin{aligned} \beta^{-1} \langle A_t^a R_s^b \rangle_0 &= \partial_s \langle A_t^a A_s^b \rangle_0 = \partial_s \langle A_t^a O_s^b \rangle_0 - \partial_s \int dx (\widehat{O}^b \varrho_0)(x) P_{t-s}(x, dy) A^a(y) \\ &= \partial_s \langle A_t^a O_s^b \rangle_0 + \int dx (\widehat{O}^b \varrho_0)(x) L_x P_{t-s}(x, dy) A^a(y). \end{aligned} \quad (7.18)$$

Integrating by parts in the last term and using the definition (7.17), we finally obtain the identity

$$\begin{aligned} \beta^{-1} \langle A_t^a R_s^b \rangle_0 &= \partial_s \langle A_t^a O_s^b \rangle_0 - \int dx (j_0 \cdot \nabla O^b)(x) P_{t-s}(x, dy) A^a(y) \\ &= \partial_s \langle A_t^a O_s^b \rangle_0 - \langle A_t^a B_s^b \rangle_0 \end{aligned} \quad (7.19)$$

which is the MFDT (4.5) with the observable O^a replaced by A^a . It is a consequence of the identity (4.5) but, in general, it does not seem to be equivalent to it, except for the equilibrium case with vanishing external force and $\varrho_0 = Z^{-1} e^{-\beta H}$ when $j_0 = 0$, and $A^a = O^a$.

In the special cases of the one-dimensional NESS with constant current j_0 described above, the dressing (7.17) of the observables coincides with the one given by equations (3.3) for types 1 and 2, respectively. This may be easily seen by checking that, for the latter,

$$L^\dagger \varrho_0 \widehat{V} = -j_0 \partial_x V \quad (7.20)$$

with $\varrho_0 = \varrho_H$ given by equations (2.11) and (2.12).

8. Conclusions

We have discussed different fluctuation relations for the Langevin dynamics. Those included an extension of the fluctuation–dissipation theorem (FDT) (1.2), one of the most important relations of the (close to) equilibrium statistical mechanics, to the case of non-equilibrium steady states (NESS) of Langevin processes. The modified fluctuation–dissipation theorem (MFDT) (4.5) that holds around NESS has a new term containing the probability current but in the Lagrangian frame moving with the mean local velocity determined by the current, it takes the form (1.3) similar to that of the equilibrium FDT. We also pointed out that, similarly to the equilibrium FDT, the MFDT may be viewed as a limiting case of more general fluctuation relations that are valid arbitrarily far from the stationary situation, namely of the Crooks’ detailed fluctuation relation (5.13) for the backward dynamics with inverted probability current or of the Jarzynski–Hatano–Sasa equality (5.12). The general discussion was illustrated on two examples of one-dimensional systems with explicit non-equilibrium invariant measures.

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Appendix A

We return here to the case of the stationary SDE (1.5). The transition probabilities $P_t(x, dy)$ for the diffusion process solving this equation are given by the kernels of the exponential of the generator $L = \beta^{-1}\partial_x^2 - (\partial_x H)\partial_x$ of the process:

$$\int P_t(x, dy)f(y) = (e^{tL}f)(x), \tag{A.1}$$

see equation (2.6). One has the following relation:

$$L = -\beta^{-1}e^{(1/2)\beta H}Q^\dagger Q e^{-(1/2)\beta H} \tag{A.2}$$

for $Q = \partial_x + \frac{1}{2}\beta(\partial_x H)$, $Q^\dagger = -\partial_x + \frac{1}{2}\beta(\partial_x H)$. The Fokker–Planck operator:

$$\beta^{-1}Q^\dagger Q = -\beta^{-1}\partial_x^2 + \frac{1}{2}(\partial_x^2 H) + \frac{1}{4}\beta(\partial_x H)^2 \tag{A.3}$$

is a positive self-adjoint Hamiltonian of a supersymmetric quantum mechanics [47] so that the transition probabilities may be defined by the relation

$$P_t(x, dy) = e^{(1/2)\beta H(x)} e^{-t\beta^{-1}Q^\dagger Q}(x, dy) e^{-(1/2)\beta H(y)}. \tag{A.4}$$

If the Gibbs density is normalizable with $Z = \int e^{-H(x)}dx < \infty$ then $\psi_0(x) = Z^{-1/2}e^{-(1/2)\beta H(x)}$ provides the zero-energy ground state of the Fokker–Planck Hamiltonian. Such a ground state is supersymmetric: $Q\psi_0 = 0$. The transition probabilities given by equation (A.4) are correctly normalized here: $\int P_t(x, dy) = 1$. However, for the Hamiltonian $H(x) = ax^k + o(|x|^k)$ at large $|x|$ with either even $k \geq 2$ and $a < 0$ or odd $k \geq 3$, the ground state ψ_0 of $\beta^{-1}Q^\dagger Q$ is not given by $e^{-(1/2)\beta H(x)}$ but has positive energy $E_0 > 0$ and breaks the supersymmetry: $Q\psi_0 \neq 0$. In these cases, the transition

probabilities (A.4) are not normalized for $t > 0$ with

$$1 > \int P_t(x, dy) \underset{t \text{ large}}{\sim} e^{-E_0 t}. \quad (\text{A.5})$$

The defect $(1 - \int P_t(x, dy))$ gives the probability that the diffusion process x_t solving the SDE (1.5) and starting at time zero at x escapes by time t to $\pm\infty$. Writing $P_t(x, dy) \equiv P_t(x, y) dy$, the probability that the escape happens between times s and $s + ds$ may be expressed as

$$\begin{aligned} \left(-\frac{d}{ds} \int P_s(x, dy) \right) ds &= \left(-\int \partial_y [(\beta^{-1} \partial_y + (\partial_y H)(y)) P_s(x, y)] dy \right) ds \\ &= \left(-(\beta^{-1} \partial_y + (\partial_y H)(y)) P_s(x, y) \Big|_{y=-\infty}^{y=\infty} \right) ds \end{aligned} \quad (\text{A.6})$$

with the $y = \pm\infty$ terms determining the rates of escape to $\pm\infty$, respectively.

Let us concentrate on the case with $H(x) = ax^k + o(|x|^k)$, for odd $k \geq 3$ and $a > 0$, denoting the transition probabilities of equation (A.4) by $P_t^0(x, dy)$. Although they are not given by a closed analytic expression, their time integral, equal to the Green kernel of L , is

$$\int_0^\infty ds P_s^0(x, dy) = (-L)^{-1}(x, dy) = \beta \left(\int_{-\infty}^{\min(x,y)} e^{\beta H(z)} dz \right) e^{-\beta H(y)} dy. \quad (\text{A.7})$$

From the last formula, we infer that

$$\lim_{y \rightarrow \pm\infty} \int_0^\infty ds (\beta^{-1} \partial_y + (\partial_y H)(y)) P_s^0(x, dy) = \delta_{1, \mp 1} \quad (\text{A.8})$$

so that the process x_t escapes here only to $-\infty$ (this is already true if we ignore the noise in equation (1.5)). On the other hand, since the limit of the right-hand side of equation (A.7) when $x \rightarrow +\infty$ exists, it follows that the transition probabilities from $x = +\infty$:

$$P_t^0(\infty, dy) = \lim_{x \rightarrow +\infty} P_t^0(x, dy) \quad (\text{A.9})$$

are finite, non-zero measures. One may then define a resurrecting version of the process x_t solving the SDE (1.5). The trajectories of such a process reappear immediately at $+\infty$ after almost surely reaching $-\infty$. The resurrecting process is Markov and its transition probabilities are

$$P_t(x, dy) = \sum_{n=0}^\infty P_t^n(x, dy), \quad (\text{A.10})$$

where $P_t^n(x, dy)$ are the transition probabilities with exactly n jumps from $-\infty$ to $+\infty$. They are given by the recursion relation:

$$P_t^{n+1}(x, dy) = -\int_0^t ds \left(\frac{d}{ds} \int P_s^0(x, dy) \right) P_{t-s}^n(\infty, dy). \quad (\text{A.11})$$

In terms of the Laplace transforms:

$$\hat{P}_\omega^n(x, dy) = \int_0^\infty e^{-t\omega} P_t^n(x, dy), \quad (\text{A.12})$$

the recursion (A.11) becomes the equality

$$\hat{P}_\omega^{n+1}(x, dy) = \left(1 - \omega \int \hat{P}_\omega^0(x, dz)\right) \hat{P}_\omega^n(\infty, dy) \quad (\text{A.13})$$

which may be easily solved by iteration:

$$\hat{P}_\omega^n(x, dy) = \left(1 - \omega \int \hat{P}_\omega^0(x, dz)\right)^n \hat{P}_\omega^0(\infty, dy) \quad (\text{A.14})$$

for $n \geq 1$. Re-summing the geometric progression, one obtains for the Laplace transform of the transition probabilities of the resurrecting process the expression

$$\hat{P}_\omega(x, dy) = \hat{P}_\omega^0(x, dy) + \frac{\left(1 - \omega \int \hat{P}_\omega^0(x, dz)\right) \hat{P}_\omega^0(\infty, dy)}{\omega \int \hat{P}_\omega^0(\infty, dz)}. \quad (\text{A.15})$$

Note that $\hat{P}_\omega^0(x, dy)$ is analytic in ω for $\text{Re } \omega > -E_0$ but $\hat{P}_\omega(x, dy)$ has a pole at $\omega = 0$ with the residue

$$\mu(dy) = \frac{\hat{P}_0^0(\infty, dy)}{\int \hat{P}_0^0(\infty, dz)} = \frac{1}{Z} \left(\int_{-\infty}^y e^{\beta H(z)} dz \right) e^{-\beta H(y)} dy, \quad (\text{A.16})$$

where Z is the normalization constant. This is the invariant probability measure (2.11) of the resurrecting process.

Let us finish by estimating the behavior of the density of the invariant measure $\mu(dy)$ when $|y| \rightarrow \infty$. We shall show that

$$\int_{-\infty}^y e^{\beta(H(z)-H(y))} dz = \frac{1}{a\beta k y^{k-1}} + o(y^{-k+1}). \quad (\text{A.17})$$

To this end, we rewrite the latter integral as

$$\int_0^\infty e^{-\beta(H(y)-H(y-z))} dz = y^{1-k} \int_0^\infty e^{-\beta(H(y)-H(y-uy^{1-k}))} du. \quad (\text{A.18})$$

We take $H(y) = ay^k + h(y)$ and assume that $h(y)$ is smooth and that

$$h(y)|y|^{-k} \xrightarrow{|y| \rightarrow \infty} 0, \quad (\partial_y h)(y)|y|^{-k+1} \xrightarrow{|y| \rightarrow \infty} 0. \quad (\text{A.19})$$

First note that for $z > 0$

$$a(y^k - (y-z)^k) = a \sum_{l=1}^k (-1)^{l+1} C_k^l y^{k-l} z^l \geq 3\epsilon z y^{k-1} + \epsilon z^k \quad (\text{A.20})$$

if $\epsilon > 0$ is small enough. Next, for $0 < z < \frac{1}{2}|y|$ and $|y|$ large enough

$$|h(y) - h(y-z)| \leq z |\partial_y h(y - \vartheta z)| \leq \epsilon z y^{k-1} \quad (\text{A.21})$$

and for $z \geq \frac{1}{2}|y|$

$$|h(y) - h(y-z)| \leq C + \epsilon z^k \quad (\text{A.22})$$

so that, altogether,

$$H(y) - H(y-z) \geq -C + \epsilon z y^{k-1} = -C + \epsilon u \quad (\text{A.23})$$

for $z = uy^{1-k}$. It is also easy to see that for each $u > 0$

$$\lim_{|y| \rightarrow \infty} [H(y) - H(y - uy^{1-k})] = aku. \quad (\text{A.24})$$

The estimate (A.17) follows then from the dominant convergence theorem applied to the integral on the right-hand side of equation (A.18).

Appendix B

We shall illustrate here the MFDT (4.5) and its version (4.11) in the frequency space on the simple example of the one-dimensional Langevin equation (1.13) on a circle with the Hamiltonian $H = 0$ and a constant force G . In this case, equation (1.13) has, of course, the explicit solution

$$x_t = x_0 + Gt + \sqrt{2\beta^{-1}}W(t) \quad (\text{B.1})$$

for the standard Brownian motion $W(t)$. Since x is the angular variable, the above process possesses an invariant probability measure with the constant density $\varrho_0 = 1/2\pi$. The corresponding current $j_0 = (1/2\pi)G$ is constant and so is the local mean velocity $v_0 = G$. The transition probabilities of the process have the form of the shifted and periodized heat kernel:

$$\begin{aligned} P_t(x, dy) &= \sqrt{\frac{\beta}{4\pi t}} \sum_{n=-\infty}^{\infty} e^{-(\beta|x-y+Gt+2\pi n|^2)/4t} dy = \frac{1}{2\pi} \sum_{n=-\infty}^{\infty} e^{in(x-y+Gt)-\beta^{-1}n^2 t} dy \\ &= \frac{1}{2\pi} \vartheta_3 \left(\frac{1}{2}(x-y+Gt), e^{-t/\beta} \right), \end{aligned} \quad (\text{B.2})$$

where $\vartheta_3(z, q)$ is the Jacobi theta function [21]. For the (real) observables $O^a(x) = \sum_n \hat{O}_n e^{inx}$, one obtains easily the equalities:

$$\begin{aligned} \mathcal{R}^{ab}(t-s) &= \theta(t-s) \sum_n O_n^a O_{-n}^b n^2 e^{(inG-\beta^{-1}n^2)(t-s)}, \\ \mathcal{C}^{ab}(t-s) &= \sum_n O_n^a O_{-n}^b e^{inG(t-s)-\beta^{-1}n^2|t-s|}, \\ \mathcal{B}^{ab}(t-s) &= -iG\theta(t-s) \sum_n O_n^a O_{-n}^b n e^{(inG-\beta^{-1}n^2)(t-s)}. \end{aligned} \quad (\text{B.3})$$

The result for the Fourier transforms (4.9) and (4.10) follows immediately:

$$\begin{aligned} \hat{\mathcal{R}}^{ab}(\omega) &= \sum_n O_n^a O_{-n}^b \frac{n^2}{\beta^{-1}n^2 - i\omega - inG}, \\ \hat{\mathcal{C}}^{ab}(\omega) &= \sum_n O_n^a O_{-n}^b \frac{2\beta^{-1}n^2}{\beta^{-2}n^4 + (\omega + nG)^2}, \\ \hat{\mathcal{B}}^{ab}(\omega) &= - \sum_n O_n^a O_{-n}^b \frac{inG}{\beta^{-1}n^2 - i\omega - inG}. \end{aligned} \quad (\text{B.4})$$

The modified FDT (4.5) or (4.11) are clearly satisfied. Taking $a = b$, one may define the factors

$$X^a(t-s) = \frac{\mathcal{R}^{aa}(t-s)}{\beta \partial_s \mathcal{C}^{aa}(t-s)} \quad \text{for } t > s, \quad \hat{X}^a(\omega) = \frac{2 \operatorname{Im} \mathcal{R}^{aa}(\omega)}{\beta \omega \hat{\mathcal{C}}^{aa}(\omega)} \quad (\text{B.5})$$

that control the violation of the standard FDT [13, 14]. In particular, for $O^a(x)$ equal to $\sin(nx)$ or $\cos(nx)$ with $n \neq 0$, one obtains (replacing the superscript a by n in this case)

$$X^n(t-s) = \left[1 + \frac{\beta G}{n} \tan(nG(t-s)) \right]^{-1}, \quad \hat{X}^n(\omega) = \frac{\beta^{-2}n^4 + \omega^2 - n^2G^2}{\beta^{-2}n^4 + \omega^2 + n^2G^2}. \quad (\text{B.6})$$

Note that $X^n(0) = 1 = \hat{X}^n(\infty)$ but that these factors are not necessarily positive. This is also true for the ‘effective temperatures’:

$$T_{\text{eff}}^a(t-s) = \beta^{-1} X^a(t-s)^{-1}, \quad \hat{T}_{\text{eff}}^a(\omega) = \beta^{-1} \hat{X}^a(\omega)^{-1}. \quad (\text{B.7})$$

In particular, $T_{\text{eff}}^n(\omega)$ is positive only in the region where $\omega^2 > (n^2G^2 - \beta^{-2}n^4)$ and in this region it decreases with ω^2 approaching for $\omega^2 \rightarrow \infty$ the value β^{-1} .

The above calculations show that the equilibrium relation (1.2) between the response and correlation functions is strongly violated in the Langevin equation on a circle with a constant drift unless the drift vanishes. On the other hand, the drift may be removed altogether by passing into the frame moving with constant velocity $\nu_0 = G$, see equation (B.1). This is captured by our MFDT (1.3). Indeed, the solutions of the advection equation (1.4) are

$$O^a(t, x) = \sum_{n=-\infty}^{\infty} \hat{O}_n e^{in(x-Gt)} \quad (\text{B.8})$$

so that the Lagrangian-frame response and correlation functions are

$$\mathcal{R}_L^{ab}(t, s) = \mathcal{R}^{ab}(t-s)|_{G=0}, \quad \mathcal{C}_L^{ab}(t, s) = \mathcal{C}^{ab}(t-s)|_{G=0} \quad (\text{B.9})$$

and the MFDT (1.3) takes here the form of the equilibrium FDT holding for $G = 0$.

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