

# Modified fluctuation-dissipation theorem for general non-stationary states and application to the Glauber–Ising chain

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Received 4 August 2011

Accepted 26 September 2011

Published 24 October 2011

Online at [stacks.iop.org/JSTAT/2011/P10025](http://stacks.iop.org/JSTAT/2011/P10025)

[doi:10.1088/1742-5468/2011/10/P10025](https://doi.org/10.1088/1742-5468/2011/10/P10025)

**Abstract.** In this paper, we present a general derivation of a modified fluctuation-dissipation theorem (MFDT) valid near an arbitrary non-stationary state for a system obeying Markovian dynamics. We show that the method for deriving modified fluctuation-dissipation theorems near non-equilibrium stationary states used by Prost *et al* (2009 *Phys. Rev. Lett.* **103** 090601) is generalizable to non-stationary states. This result follows from both standard linear response theory and from a transient fluctuation theorem, analogous to the Hatano–Sasa relation. We show that this modified fluctuation-dissipation theorem can be interpreted at the trajectory level using the notion of stochastic trajectory entropy, in a way which is similar to what has been done recently in the case of the MFDT near non-equilibrium steady states (NESS). We illustrate this framework with two solvable examples: the first example corresponds to a Brownian particle in a harmonic trap subjected to a quench of temperature and to a time-dependent stiffness; the second example is a classic model of coarsening systems, namely the 1D Ising model with Glauber dynamics.

**Keywords:** exact results, spin glasses (theory), fluctuations (theory), stochastic processes (theory)

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**1. Introduction**

It is a general rule that as a system gets smaller its fluctuations increase. As a consequence, for small systems (like a colloidal particle or a biomolecule), thermodynamic quantities like work [1, 2] and heat are only defined in a statistical sense. Exact relations between the statistical distributions of these thermodynamic quantities, known as fluctuation relations, were obtained about a decade ago. Such ideas have led to the emergence of a new field, concerned with the specificity of thermodynamics for small systems, which has been called stochastic thermodynamics.

Fluctuation relations hold very generally for a large class of systems and arbitrarily far from equilibrium [1], [3]–[7]. They provide fresh ideas for revisiting an old but central issue of statistical physics, namely the origin of irreversibility of macroscopic systems. Furthermore, within the linear regime, these fluctuations relations lead to various new modified fluctuation-dissipation theorems (MFDT), which are interesting and valuable extensions of the classical fluctuation-dissipation theorem [8]–[11].

Recently, three main routes have emerged for constructing such generalizations of the fluctuation-dissipation theorem:

- In the first route initiated by Cugliandolo *et al* [12] and continued by Lippiello *et al* [13] and Diezemann [14], the response function is written as a sum of a time derivative of the correlation function (similar to the equilibrium FDT) and an additive function, called the asymmetry, which vanishes under equilibrium conditions. A physical interpretation for this asymmetry has been lacking for many years, until, recently, Baiesi *et al* [15] proposed a different separation of the response function, in which one part is associated with a new concept called the frenesy. This frenesy contains the time symmetric part of the non-equilibrium fluctuations.
- In the second route initiated by Speck and Seifert [16], the modifications of the fluctuation-dissipation theorem can be related to the so-called local velocity, which originates in the local currents present in the non-equilibrium situation. This route was further extended and generalized by Ch  trite *et al* [17] who also provided the MFDT with a Lagrangian frame interpretation [18]. These ideas were then confirmed experimentally using colloidal particles confined to circular trajectories [19, 20]. In the end, it appears that the first and second routes are closely related and can be unified through the introduction of stochastic derivatives [21].
- In the third approach developed by Prost *et al* [22] (see also [17]), the modified fluctuation theorem valid near non-equilibrium steady states (NESS) takes the standard equilibrium form except that it involves a new observable, a function of the non-equilibrium steady state. This new observable is the system stochastic entropy, which here must be evaluated in the NESS [23, 24].

Surprisingly, while the first and second approaches provide ways to construct generalizations of the fluctuation dissipation for cases where the system is initially in an arbitrary non-equilibrium state, the third approach seems limited to systems close to a non-equilibrium stationary state. One objective of the present paper is to show that the third approach can also be extended to systems close to a general non-stationary state, in a way which is closely related to the first approach. In view of this, it appears that the three approaches provide closely related formulations for generalizing the fluctuation-dissipation theorem to non-equilibrium situations.

Such generalizations could potentially lead to a broad range of applications to non-stationary or driven systems, such as glasses, spin glasses, coarsening systems, granular media and dense colloidal systems, for which violations of the fluctuation-dissipation theorem have been an active topic for many years [12]–[14], [25]. Another more recent but promising field of application of these ideas concerns biological systems [26]–[28], where the application of MFDT could possibly lead to new methods for probing these complex media.

In section 2, we show how to derive a MFDT classically, first using linear response theory, and then using more recent methods based on fluctuation relations. We then discuss an interpretation of the MFDT within stochastic thermodynamics, according to which the MFDT can be formulated in terms of a particular form of stochastic entropy. In section 3, we illustrate our framework with two pedagogical examples. In the first example, a Brownian particle placed in a harmonic potential is subjected to a quench of temperature and to a time-dependent stiffness, and in the second example, a 1D Ising chain obeying Glauber dynamics is subjected to a quench of temperature and then probed with a magnetic field.

## 2. A modified fluctuation-dissipation theorem (MFDT) for general non-stationary states

### 2.1. Stochastic modelling and definitions

In the following, we derive a modified fluctuation-dissipation theorem (MFDT) for a system which is initially (at time 0) in a general non-stationary state. The evolution of the system at all times is assumed to follow a continuous-time Markovian dynamics of a pure jump type [29]. The rate of transition for jumping from a configuration  $c$  to a configuration  $c'$  is denoted as  $w_t(c, c')$ , where the subscript  $t$  indicates that we allow for time-dependent rates. We denote as  $\rho_t(c)$  the probability of being in state  $c$  at time  $t$ . This quantity obeys the unperturbed master equation

$$\frac{d\rho_t(c)}{dt} = \sum_{c'} [\rho_t(c')w_t(c', c) - \rho_t(c)w_t(c, c')] \quad (1)$$

which can be written equivalently as

$$\frac{d\rho_t(c)}{dt} = \sum_{c'} \rho_t(c')L_t(c', c), \quad (2)$$

in terms of the time-dependent Markovian generator  $L_t(c', c)$  defined by

$$L_t(c', c) = w_t(c', c) - \delta(c, c') \sum_{c''} w_t(c', c''). \quad (3)$$

At time  $t = 0$ , an arbitrary but given time-dependent perturbation  $h_t$  is applied to the system, and we denote by  $P_t(c, [h_t])$  the probability of observing the system in the state  $c$  at a time  $t$  in the presence of this perturbation. The notation  $[h_t]$  emphasizes that the dependence is functional with respect to the perturbation. The evolution of the system for  $t > 0$  is controlled by the generator  $L_t^{h_t}$ , which is defined like in equation (3) provided the rates  $w_t(c, c')$  are replaced by perturbed rates  $w_t^{h_t}(c, c')$ . This generator can be expanded to first order in  $[h_t]$ :

$$L_t^{h_t} = L_t + h_t N_t. \quad (4)$$

In the following, it does matter whether the unperturbed dynamics is autonomous or not. If this dynamics is non-autonomous, i.e. proceeds from the application of a protocol, we still denote the generator by  $L_t$  without specifying this protocol explicitly. In this case, the application of the perturbation  $h_t$  for  $t > 0$  can be seen as an additional protocol.

Let us introduce  $\pi_t(c, h)$  as the probability of observing the system in the state  $c$  at a time  $t > 0$  in the presence of a constant (time-independent) perturbation  $h$ , which obeys

$$\left( \frac{\partial \pi_t}{\partial t} \right) (c, h) = \sum_{c'} \pi_t(c', h) L_t^h(c', c). \quad (5)$$

A key object for the following discussion is  $\pi_t(c, h_t)$ , which is constructed from  $\pi_t(c, h)$  by replacing the time-independent constant  $h$  by the value of the perturbation at time  $t$ , namely  $h_t = h(t)$ . In the particular case where the perturbed dynamics with a constant  $h$  is time independent (i.e.  $L_t^h \equiv L^h$ ), the subscript  $t$  in  $\pi_t(c, h)$  may be dropped, and  $\pi(c, h_t)$  becomes the ‘accompanying’ distribution introduced in [30]. We emphasize that  $\pi_t(c, h_t)$

depends only on the perturbation at time  $t$ , unlike  $P_t(c, [h_t])$  which depends functionally on the protocol history of the perturbation. The dynamics of  $\pi_t(c, h_t)$  is given by

$$\frac{d}{dt}(\pi_t(c, h_t)) = \sum_{c'} \pi_t(c', h_t) L_t^{h_t}(c', c) + \dot{h}_t \frac{\partial \pi_t(c, h_t)}{\partial h_t} \quad (6)$$

$$= \sum_{c'} \pi_t(c', h_t) [L_t^{h_t}(c', c) - \delta(c', c) \dot{h}_t \partial_{h_t} \psi_t(c, h_t)], \quad (7)$$

where in the second equation one has introduced  $\psi_t(c, h_t) = -\ln \pi_t(c, h_t)$  and an initial condition  $\rho_0(c) = P_0(c, 0) = \pi_0(c, 0)$  has been assumed.

The response function associated with a dynamic observable  $A_t(c_t, h_t)$  reads, for a perturbation applied at an earlier time  $t' > 0$ ,

$$R(t, t') = \left. \frac{\delta \langle A_t(c_t, h_t) \rangle_{[h_t]}}{\delta h_{t'}} \right|_{h \rightarrow 0}. \quad (8)$$

where  $\langle \dots \rangle_{[h_t]}$  represents an average with respect to the perturbed dynamics, and  $\delta/\delta h_{t'}$  is our notation for functional derivatives. We derive below a general formulation of a modified dissipation theorem for non-stationary states, which reads for  $t > t' > 0$

$$R(t, t') = -\frac{d}{dt'} \langle \partial_h \psi_{t'}(c_{t'}, h) |_{h \rightarrow 0} A_t(c_t, h_t) \rangle. \quad (9)$$

This relation qualifies as a modified fluctuation dissipation because the response function is now expressed in terms of a correlation function of observables with respect to the unperturbed dynamics, denoted as  $\langle \dots \rangle$  [10]. Note that when  $A_t(c_t, h_t) = \partial_{h_t} \psi_t(c_t, h_t)$ , the modified fluctuation-dissipation theorem takes a more symmetric form, derived in [22], under some specific conditions: these conditions are that the initial state must be stationary and that the dynamics followed by the system at constant perturbation  $h$  must be time independent ( $L_t^h \equiv L^h$ ). Thus, as announced in section 1, the present derivation extends the result developed in this reference to a more general initial condition and a more general dynamics.

## 2.2. Derivation of the MFDT from linear response theory

This section shows how the modified fluctuation-dissipation theorem of equation (9) follows from standard linear response theory. Starting from the master equation given above, one can generate a Dyson-type equation for the perturbed propagator  $P(ct|c't')$ , which is a fundamental result of linear response theory [9, 10, 30]. This propagator  $P(ct|c't')$  represents the probability of finding the system in the state  $c$  at time  $t$  given that it was in the state  $c'$  at time  $t'$  according to the perturbed dynamics, while  $\rho(ct|c't')$  denotes the corresponding propagator for the unperturbed dynamics. When taken to first order in  $[h_t]$ , the Dyson equation [31] for the propagator reads

$$P(ct|c_0 0) = \rho(ct|c_0 0) + \int_0^t dt' h_{t'} \sum_{c'', c'} \rho(ct|c't') N_{t'}(c'', c') \rho(c''t'|c_0 0). \quad (10)$$

We then multiply this equation by an arbitrary observable, which we denote here as  $A_t(c)$  as shorthand notation for  $A_t(c, h_t)$ . After integrating over the initial distribution  $\rho_0(c_0)$ ,

one obtains

$$\langle A_t(c_t) \rangle_{[h_t]} = \langle A_t(c_t) \rangle + \int_0^t dt' h_{t'} \sum_{c, c'} A_t(c) \rho(ct, c't') B_{t'}(c') \quad (11)$$

where we used the notation  $\rho(ct, c't') = \rho(ct|c't')\rho_{t'}(c')$ , and  $B$  is the operator such that

$$B_{t'}(c') = \rho_{t'}(c')^{-1} \sum_{c''} N_{t'}(c'', c') \rho_{t'}(c''). \quad (12)$$

The main point of introducing  $B$  is that it allows us to write the response function in terms of a correlation function of two observables with respect to the unperturbed dynamics [10]:

$$R(t, t') = \langle B_{t'}(c_{t'}) A_t(c_t) \rangle. \quad (13)$$

We can now use an expansion of the distribution  $\pi_t$  to first order in  $h$ ,  $\pi_t(c, h) = \pi_t^{(0)}(c) + h\pi_t^{(1)}(c)$ . It follows from the master equation that the zeroth-order solution is  $\pi_t^{(0)}(c) = \rho_t(c)$ , while the first-order solution is

$$\frac{\partial \pi_t^{(1)}(c)}{\partial t} = \sum_{c'} \pi_t^{(0)}(c') N_t(c', c) + \pi_t^{(1)}(c') L_t(c', c). \quad (14)$$

Thus, the observable  $B$  defined above can be written as

$$B_t(c) = \pi_t^{(0)}(c)^{-1} \sum_{c'} N_t(c', c) \pi_t^{(0)}(c'), \quad (15)$$

$$= \pi_t^{(0)}(c)^{-1} \left( \frac{\partial}{\partial t} \pi_t^{(1)}(c) - \sum_{c'} L_t(c', c) \pi_t^{(1)}(c') \right). \quad (16)$$

After substituting this in equation (13), one obtains

$$R(t, t') = \sum_{c, c'} A_t(c) \rho(ct|c't') \left( \frac{\partial}{\partial t'} \pi_{t'}^{(1)}(c') - \sum_{c''} L_{t'}(c'', c') \pi_{t'}^{(1)}(c'') \right). \quad (17)$$

This form can be further transformed using the property that the unperturbed propagator  $\rho(ct|c't')$  satisfies the backward Kolmogorov equation [32]

$$\partial_{t'} \rho(ct|c't') = - \sum_{c''} L_{t'}(c'', c') \rho(ct|c't'), \quad (18)$$

so in the end,

$$R(t, t') = \frac{d}{dt'} \left( \sum_{c, c''} A_t(c) \rho(ct|c''t') \pi_{t'}^{(1)}(c'') \right), \quad (19)$$

which leads to equation (9) after using the relation  $\pi_{t'}^{(1)}(c)/\pi_{t'}^{(0)}(c) = \partial_h \ln \pi_{t'}(c, h)|_{h \rightarrow 0}$ .

### 2.3. Derivation of the MFDT from fluctuation relations

For each path trajectory, we introduce the following functional:

$$\mathcal{Y}_t = \int_0^t \dot{h}_\tau \partial_{h_\tau} \psi_\tau(c_\tau, h_\tau) d\tau. \quad (20)$$

This functional  $\mathcal{Y}_t$  has already appeared in [21, 33] but in a different form. In the appendix A, we explain the connections between the different formulations. The advantage of writing  $\mathcal{Y}_t$  in the form of equation (20), besides its simplicity, is that the similarity with the functionals introduced by Jarzynski [1] and Hatano and Sasa [7] is then very apparent.

In the same spirit as in the seminal works of Jarzynski and of Hatano and Sasa, we consider, below, averages over trajectories with a weight  $\mathcal{Y}_t$ . To perform such averages, we introduce the joint probability of being in the configuration  $c$  at time  $t$  with a value  $\mathcal{Y}$  for the quantity  $\mathcal{Y}_t$ ,  $P_t(c, \mathcal{Y})$ , which is defined by

$$P_t(c, \mathcal{Y}) = \langle \delta(c - c_t) \delta(\mathcal{Y} - \mathcal{Y}_t) \rangle_{[h_t]}. \quad (21)$$

This quantity obeys the following master equation:

$$\frac{\partial P_t(c, \mathcal{Y})}{\partial t} = \sum_{c'} P_t(c', \mathcal{Y}) L_t^{h_t}(c', c) - \dot{h}_t \frac{\partial \psi_t(c, h_t)}{\partial h_t} \frac{\partial P_t(c, \mathcal{Y})}{\partial \mathcal{Y}}, \quad (22)$$

which can be solved through the Laplace transform. Defining  $\hat{P}_t(c, \gamma) = \int d\mathcal{Y} P_t(c, \mathcal{Y}) e^{-\gamma \mathcal{Y}}$ , we obtain

$$\frac{\partial \hat{P}_t(c, \gamma)}{\partial t} = \sum_{c'} \hat{P}_t(c', \gamma) L_t^{h_t}(c', c) - \dot{h}_t \gamma \frac{\partial \psi_t(c, h_t)}{\partial h_t} \hat{P}_t(c, \gamma). \quad (23)$$

Thus, the equation satisfied by  $\hat{P}_t(c, \gamma = 1)$  is identical to the equation (7) satisfied by  $\pi_t(c, h_t)$ . Furthermore, since  $P_0(c, \mathcal{Y}) = P_0(c) \delta(\mathcal{Y})$ , the two functions have identical initial conditions  $\hat{P}_0(c, 1) = \int d\mathcal{Y} P_0(c, \mathcal{Y}) \exp(-\mathcal{Y}) = P_0(c) = \pi_0(c, 0)$ . Therefore, these two functions must be identical; in other words,  $\hat{P}_t(c, 1) = \pi_t(c, h_t)$ . Using the definition of the Laplace transform, it follows from this equality that

$$\pi_t(c, h_t) = \langle \delta(c - c_t) e^{-\mathcal{Y}_t} \rangle_{[h_t]}, \quad (24)$$

an equation which can be called a Feynman–Kac formula [34, 35].

By multiplying equation (24) by an arbitrary observable  $A_t(c, h_t)$  and integrating over  $c$ , one obtains the following generalization of the Hatano–Sasa relation:

$$\langle A_t(c_t, h_t) e^{-\mathcal{Y}_t} \rangle_{[h_t]} = \int dc \pi_t(c, h_t) A_t(c, h_t) = \langle A_t(c_t, h_t) \rangle_{\pi_t}, \quad (25)$$

where in the last equality,  $\langle \dots \rangle_{\pi_t}$  denotes the average with respect to  $\pi_t(c, h_t)$ . In the particular case where the initial condition is stationary and provided that the perturbed dynamics at constant perturbation  $h$  is time independent, the Hatano–Sasa relation [7] is recovered from equation (25) for the particular case of a constant observable  $A_t = 1$ .

We now consider a small variation with respect to the perturbation  $h_t$ . From the definition of  $\mathcal{Y}_t$  in equation (20), it follows that this quantity is small, of order 1 in  $h_t$ ,



and thus  $e^{-\mathcal{Y}_t} \simeq 1 - \mathcal{Y}_t$ . Therefore, we have

$$\langle A_t(h_t) \rangle_{\pi_t} \simeq \langle A_t(c_t, h_t) \rangle_{[h_t]} - \left\langle \int_0^t \dot{h}_\tau \partial_{h_\tau} \psi_\tau(c_\tau, h_\tau) A_t(c_t, h_t) d\tau \right\rangle_{[h_t]}, \quad (26)$$

$$\simeq \langle A_t(c_t, h_t) \rangle_{[h_t]} - \int_0^t d\tau \dot{h}_\tau \langle \partial_h \psi_\tau(c_\tau, h) |_{h \rightarrow 0} A_t(c_t, h_t) \rangle, \quad (27)$$

where in the last equation, we have approximated the derivative with respect to  $h_\tau$  by a derivative with respect to  $h$ , an approximation which is correct to first order with respect to the perturbation, and at the same order in perturbation we have replaced the perturbed average by an unperturbed one. Taking into account that the functional derivative of the lhs of equation (27) with respect to  $h_{t'}$  vanishes for  $t' < t$ , and rewriting the second term of the rhs using an integration by parts, we obtain equation (9).

#### 2.4. Interpretation of the MFDT in terms of trajectory entropy

In section 2.1, we have introduced a key quantity, namely  $\psi_t(c, h) = -\ln \pi_t(c, h)$ . When properly evaluated on a specific trajectory  $[c_t, h_t]$ , the function  $\psi_t(c_t, h_t)$  gives access to the functional  $\mathcal{Y}_t$  defined in equation (20), and when evaluated at small constant  $h$  on a trajectory  $[c_t]$ , it allows one to calculate the response in the MFDT according to equation (9). Clearly, this quantity must be closely related to the stochastic entropy introduced in [36]. Indeed, the stochastic system entropy is defined as

$$s_t(c_t, [h_t]) = -\ln P_t(c_t, [h_t]), \quad (28)$$

and therefore depends functionally on the perturbation  $[h_t]$ . In contrast to that, the system entropy which is needed here is evaluated using a constant perturbation  $[h]$ :

$$s_t(c_t, [h]) = -\ln P_t(c_t, [h]) = -\ln \pi_t(c_t, h) = \psi_t(c_t, h). \quad (29)$$

We now focus on the trajectories taken by the system, which can be described by a set of discrete values  $\mathcal{C} = \{c_0, c_1, \dots, c_N\}$ , with the convention that the system is in state  $c_0$  at time 0 and in state  $c_N$  at time  $t$ . Furthermore, the transition from state  $c_{j-1}$  to state  $c_j$  occurs at the jumping times  $\tau_j$ . The stochastic system entropy can be decomposed as  $s_t(c_t, [h_t]) = -s_t^r(c_t, [h_t]) + s_t^{\text{tot}}(c_t, [h_t])$ , in terms of the reservoir entropy production  $s_t^r(c_t, [h_t])$  (also called the medium entropy in [36]) and the total entropy production  $s_t^{\text{tot}}(c_t, [h_t])$ .

The system entropy is a state function, which means that

$$\Delta s_t(c_t, [h_t]) = -\ln P_t(c_t, [h_t]) + \ln P_0(c_0, h_0). \quad (30)$$

In contrast to that, the reservoir entropy and the total entropy are not state functions, but are trajectory-dependent quantities, which can be written as

$$\begin{aligned} \Delta s_t^r(c_t, [h_t]) &= \sum_{j=1}^N \ln \frac{w_{\tau_j}^{h_j}(c_{j-1}, c_j)}{w_{\tau_j}^{h_j}(c_j, c_{j-1})}, \\ \Delta s_t^{\text{tot}}(c_t, [h_t]) &= \sum_{j=1}^N \ln \frac{P_{\tau_j}(c_{j-1}, [h_t]) w_{\tau_j}^{h_j}(c_{j-1}, c_j)}{P_{\tau_j}(c_j, [h_t]) w_{\tau_j}^{h_j}(c_j, c_{j-1})} - \int_0^t d\tau (\partial_\tau \ln P_\tau)(c_\tau, [h_\tau]), \end{aligned} \quad (31)$$



where  $h_j$  is the value of the control parameter at the jump time  $\tau_j$ . Between the jumps,  $s_t^r$  is a constant function of the time while  $s_t$  and  $s_t^{\text{tot}}$  are in general non-constant but continuous functions of the time. All these functions,  $s_t$ ,  $s_t^r$  and  $s_t^{\text{tot}}$ , are discontinuous at the jump times  $\tau_j$ .

When adapted to the case of a constant perturbation  $[h]$ , the above decomposition of the system entropy leads to two terms in the MFDT. Starting from equation (9) together with equation (30), one obtains

$$R(t, t') = -\frac{d}{dt'} \langle \partial_h \Delta s_{t'}(c_{t'}, [h])|_{h \rightarrow 0} A_t(c_t) \rangle = R_{\text{eq}}(t, t') - R_{\text{neq}}(t, t'), \quad (32)$$

where

$$R_{\text{eq}}(t, t') = \frac{d}{dt'} \langle \partial_h \Delta s_{t'}^r(c_{t'}, [h])|_{h \rightarrow 0} A_t(c_t) \rangle, \quad (33)$$

and

$$R_{\text{neq}}(t, t') = \frac{d}{dt'} \langle \partial_h \Delta s_{t'}^{\text{tot}}(c_{t'}, [h])|_{h \rightarrow 0} A_t(c_t) \rangle. \quad (34)$$

This decomposition of the MFDT contains two terms: the first term  $R_{\text{eq}}(t, t')$ , which is analogous to the equilibrium FDT in a sense made more precise below, and the second term  $R_{\text{neq}}(t, t')$ , which represents an additive correction. Such a decomposition has been discussed by several authors following the original work of [16]. Note that the interpretation of the MFDT or standard FDT in terms of trajectory entropies is more recent [15, 37]; in the context of non-equilibrium stationary states this has been done in [23, 24]. The present decomposition is very similar to that one, but here the total entropy production replaces the so-called adiabatic entropy production [38], because the non-adiabatic part is non-zero and contributes to the second term  $R_{\text{neq}}(t, t')$ .

*Reformulation of the MFDT using local currents.* Let us rewrite more explicitly the two terms above for the MFDT without the time derivatives present in equations (32)–(34), instead using local currents. For the first term, using the expression for the reservoir entropy of equation (31), we have

$$\langle \Delta s_{t'}^r(c_{t'}, [h]) A_t(c_t) \rangle = \int_0^{t'} ds \sum_{c, c', c''} \rho_s(c) w_s(c, c') \ln \left[ \frac{w_s^h(c, c')}{w_s^h(c', c)} \right] \rho(c'' | c' s) A_t(c''), \quad (35)$$

which implies that

$$\begin{aligned} \frac{d}{dt'} \langle \partial_h \Delta s_{t'}^r(c_{t'}, [h])|_{h \rightarrow 0} A_t(c_t) \rangle &= \sum_{c, c', c''} \rho_{t'}(c) w_{t'}(c, c') \partial_h \ln \frac{w_{t'}^h(c, c')}{w_{t'}^h(c', c)} \Big|_{h \rightarrow 0} \rho(c'' | c' t') A_t(c''), \\ &\equiv \langle j_{t'}(c_{t'}) A_t(c_t) \rangle = R_{\text{eq}}(t, t'), \end{aligned} \quad (36)$$

where  $j_{t'}$  denotes the local current given by

$$j_{t'}(c') = \sum_c \frac{\rho_{t'}(c)}{\rho_{t'}(c')} w_{t'}(c, c') \partial_h \ln \frac{w_{t'}^h(c, c')}{w_{t'}^h(c', c)} \Big|_{h \rightarrow 0}. \quad (37)$$

A property of this local current is that its average represents a physical current:

$$\langle j_{t'}(c_{t'}) \rangle = \sum_{c, c'} J_{t'}(c', c) \partial_h \ln w_{t'}^h(c', c) \Big|_{h \rightarrow 0}, \quad (38)$$

with  $J_{t'}(c', c) = \rho_{t'}(c') w_{t'}(c', c) - \rho_{t'}(c) w_{t'}(c, c')$ , the unperturbed probability current between the states  $c$  and  $c'$ .

The same strategy can be used to transform the second term in the MFDT.

$$\begin{aligned} \langle \Delta s_{t'}^{\text{tot}}(c_{t'}, [h]) A_t(c_t) \rangle &= \int_0^{t'} ds \sum_{c, c', c''} \rho_s(c) w_s(c, c') \ln \left[ \frac{\pi_s(c, h) w_s^h(c, c')}{\pi_s(c', h) w_s^h(c', c)} \right] \rho(c'' | c' s) A_t(c'') \\ &+ \int_0^{t'} d\tau \sum_{c', c''} \rho_\tau(c') (\partial_\tau \psi_\tau)(c', h) \rho(c'' | c' \tau) A_t(c''), \end{aligned} \quad (39)$$

which implies

$$\begin{aligned} \frac{d}{dt'} \langle \partial_h \Delta s_{t'}^{\text{tot}}(c_{t'}, [h]) \Big|_{h \rightarrow 0} A(c_t) \rangle &= \sum_c \left\langle \frac{\rho_{t'}(c)}{\rho_{t'}(c')} w_{t'}(c, c') \partial_h \ln \frac{\pi_{t'}(c, h) w_{t'}^h(c, c')}{\pi_{t'}(c', h) w_{t'}^h(c', c)} \Big|_{h \rightarrow 0} A_t(c_t) \right\rangle \\ &+ \langle \partial_h (\partial_{t'} \psi_{t'}) (c_{t'}, h) \Big|_{h \rightarrow 0} A_t(c_t) \rangle. \end{aligned} \quad (40)$$

One can rewrite this in a more compact form in terms of another local current  $\nu_{t'}$  such that

$$\frac{d}{dt'} \langle \partial_h \Delta s_{t'}^{\text{tot}}(c_{t'}, [h]) \Big|_{h \rightarrow 0} A(c_t) \rangle \equiv \langle \nu_{t'}(c_{t'}) A_t(c_t) \rangle = R_{\text{neq}}(t, t'), \quad (41)$$

where

$$\nu_{t'}(c') = \sum_c \frac{J_{t'}(c', c)}{\rho_{t'}(c')} \partial_h \ln w_{t'}^h(c', c) \Big|_{h \rightarrow 0}. \quad (42)$$

Note that this equation, together with equation (38), implies that the two currents  $\nu_{t'}(c')$  and  $j_{t'}(c_{t'})$  have the same average. In the end, the MFDT takes the form

$$R(t, t') = \langle (j_{t'}(c_{t'}) - \nu_{t'}(c_{t'})) A_t(c_t) \rangle. \quad (43)$$

We emphasize that the decomposition of equations (37)–(42) is a general result, which does not rely on any assumption about the form of the rates. Note that the response function can thus be written in a way which does not contain a time derivative provided two local currents  $j_{t'}$  and  $\nu_{t'}$  are introduced. This comes at the price that there is not a unique decomposition of this type [24].

When additional assumptions are available about the transition rates, the two terms can be further transformed. One such assumption is a generalized detailed balance relation of the form

$$\frac{w_t^{h_t}(c, c')}{w_t^{h_t}(c', c)} = \frac{w_t(c, c')}{w_t(c', c)} \exp(h_t d_t(c, c')), \quad (44)$$

where  $d_t(c, c')$  represents the variation of a physical quantity conjugate to  $h_t$  during the transition from state  $c$  to  $c'$  at time  $t$ . In this case, the local current  $j_{t'}$  can be simplified as

$$j_{t'}(c') = \sum_c \frac{\rho_{t'}(c)}{\rho_{t'}(c')} w_{t'}(c, c') d_t(c, c'). \quad (45)$$

One particular usual choice of transition rates compatible with equation (44) is

$$w_{t'}^{h_{t'}}(c, c') = w_{t'}(c, c') \exp\left(\beta h_{t'} \frac{O(c') - O(c)}{2}\right), \quad (46)$$

where  $O(c)$  represents a physical time-independent observable, and  $d(c, c') = O(c') - O(c)$  is its variation between states  $c$  and  $c'$ . See [21] for a general discussion on the various forms of perturbations in connection with the potential theory.

The reservoir entropy introduced in equation (31) only needs to be evaluated for a constant perturbation  $[h]$ ; therefore,

$$\begin{aligned} \partial_h \Delta s_t^r(c_t, [h])|_{h \rightarrow 0} &= \sum_{j=1}^N \partial_h \ln \frac{w_{\tau_i}^h(c_{j-1}, c_j)}{w_{\tau_i}^h(c_j, c_{j-1})} \Big|_{h \rightarrow 0} = \beta \sum_{j=1}^N (O(c_j) - O(c_{j-1})), \\ &= \beta (O(c_N) - O(c_0)) = \beta (O(c_t) - O(c_0)). \end{aligned} \quad (47)$$

Substituting this in the first term of the MFDT, namely, equation (33), one obtains

$$R_{\text{eq}}(t, t') = \beta \frac{d}{dt'} \langle O(c_{t'}) A(c_t) \rangle, \quad (48)$$

which is a form similar to the equilibrium FDT. Indeed, in the case of a perturbation around an equilibrium state, the average  $\langle \dots \rangle$  becomes an equilibrium average, and the equilibrium form of FDT is recovered. We explain in appendix A how the second term  $R_{\text{neq}}(t, t')$  can be transformed in the continuous-space limit for the particular case of a nearest-neighbour random walk.

In the end, the present framework provides a way to interpret the MFDT at the level of trajectory entropies in the case where a system is perturbed near an arbitrary non-equilibrium state. Thus, this framework generalizes the results obtained in [23] for non-equilibrium stationary states.

### 3. Illustrative examples

While the derivations above concern discontinuous pure jump Markov processes, the results are more general and their proofs are transposable for continuous diffusion processes [10]. For this reason, we provide, in the following, two illustrative examples of each kind: the first one corresponds to a continuous process of the Langevin type while the second one corresponds to a discrete jump process (Glauber dynamics).

### 3.1. A particle obeying Langevin dynamics in a harmonic potential with time-dependent stiffness and subjected to a quench of temperature

We consider a particle in one dimension and in a harmonic potential obeying Langevin dynamics:

$$\dot{x}_t = -\frac{k_t}{\gamma}x_t + \frac{h_t}{\gamma} + \eta_t \quad \text{with} \quad \langle \eta_t \eta_{t'} \rangle = \frac{2T_t}{\gamma} \delta(t - t') \quad \text{and} \quad \langle \eta_t \rangle = 0, \quad (49)$$

where  $\eta_t$  is a Gaussian white noise,  $k_t$  a time-dependent spring constant,  $\gamma$  a friction coefficient and  $T_t$  the time-dependent temperature of the bath, which starts from  $T_0$  at  $t = 0$  and ends at  $T_{t_f}$  at  $t = t_f$ . As a result of the non-stationary bath and of the time-dependent spring constant, the system at time  $t > 0$  is not in equilibrium although it is assumed to be at equilibrium at  $t = 0$ . We denote by  $h_t$  an additional external perturbing force. For this system, one can compute explicitly, provided that the spring constant is integrable on interval  $[0, t]$ , the position at time  $t$ :

$$x_t = x_0 e^{-\int_0^t d\tau k_\tau/\gamma} + e^{-\int_0^t d\tau k_\tau/\gamma} \int_0^t d\tau \left( \frac{h_\tau}{\gamma} + \eta_\tau \right) e^{\int_0^\tau d\tau' k_{\tau'}/\gamma}, \quad (50)$$

which is also a random Gaussian variable thanks to the linearity in  $\eta$  and because the probability distribution of  $x_0$  is the equilibrium Gaussian one at  $T_0$ , i.e.  $\rho_0(x_0) = \exp(-k_0 x_0^2/2T_0)/Z$  with  $Z$  the partition function. From equation (50), we obtain

$$\mu_t = \langle x_t \rangle_{[h_t]} = \int_0^t d\tau \frac{h_\tau}{\gamma} \exp\left(-\int_\tau^t d\tau' k_{\tau'}/\gamma\right), \quad (51)$$

$$\sigma_t^2 = \langle x_t^2 \rangle_{[h_t]} - \langle x_t \rangle_{[h_t]}^2, \quad (52)$$

$$= \langle x_0^2 \rangle \exp\left(-2 \int_0^t d\tau k_\tau/\gamma\right) + \int_0^t d\tau \frac{2T_\tau}{\gamma} \exp\left(-2 \int_\tau^t du k_u/\gamma\right). \quad (53)$$

The functional derivative of equation (51) with respect to  $h_{t'}$  gives directly the response function

$$R(t, t') = \left. \frac{\delta \langle x_t \rangle_{[h_t]}}{\delta h_{t'}} \right|_{h \rightarrow 0} = \frac{1}{\gamma} \exp\left(-\int_{t'}^t d\tau k_\tau/\gamma\right). \quad (54)$$

As we show below, this result can also be recovered from the MFDT of equation (9). Since  $x_t$  is a Gaussian variable, we deduce from this that the probability density function at time  $t$  is

$$P_t(x, [h_t]) = \frac{1}{(2\pi\sigma_t^2)^{1/2}} \exp\left[\frac{-1}{2\sigma_t^2} \left(x - \int_0^t d\tau \frac{h_\tau}{\gamma} \exp\left(-\int_\tau^t d\tau' k_{\tau'}/\gamma\right)\right)^2\right]. \quad (55)$$

We note that  $\sigma_t^2$  does not depend on the perturbation  $[h]$  but  $\mu_t$  does. To obtain the probability density function  $\pi_t(x, h)$ , we just make the perturbation constant using  $\pi_t(x, h) = P_t(x, [h])$ , so

$$\pi_t(x, h) = \frac{1}{(2\pi\sigma_t^2)^{1/2}} \exp\left[\frac{-1}{2\sigma_t^2} \left(x - \frac{h}{\gamma} \int_0^t d\tau \exp\left(-\int_\tau^t d\tau' k_{\tau'}/\gamma\right)\right)^2\right]. \quad (56)$$

Note that  $\pi_t(x, h_t)$  is indeed distinct from  $P_t(x, [h_t])$  as emphasized from the beginning. Since we now have the key function  $\pi_t$ , we can compute the response function and the functional  $\mathcal{Y}_t$ .

To obtain the response, we calculate  $\psi_t(x, h) = -\ln \pi_t(x, h)$  and its derivative with respect to  $h$ :

$$\partial_h \psi_t(x, h) = -\frac{1}{\sigma_t^2} (x - hI_t) I_t, \quad (57)$$

where

$$I_t = \frac{1}{\gamma} \int_0^t d\tau e^{(-\int_\tau^t d\tau' k_{\tau'}/\gamma)}, \quad (58)$$

resulting in

$$\langle \partial_h \psi_{t'}(x_{t'}) x_t \rangle |_{h \rightarrow 0} = -\frac{I_{t'}}{\sigma_{t'}^2} \langle x_t x_{t'} \rangle |_{h \rightarrow 0}. \quad (59)$$

This last expression requires the two-time correlation function for  $x$  that can be obtained from equation (50), assuming  $t' < t$ :

$$\langle x_{t'} x_t \rangle = \mu_t \mu_{t'} + \sigma_{t'}^2 \exp\left(-\int_{t'}^t d\tau k_\tau/\gamma\right). \quad (60)$$

Since  $\mu_t = \mu_{t'} = 0$  in the limit  $h \rightarrow 0$ , we obtain

$$\langle \partial_h \psi_{t'}(x_{t'}) x_t \rangle |_{h \rightarrow 0} = -I_{t'} \exp\left(-\int_{t'}^t d\tau k_\tau/\gamma\right), \quad (61)$$

$$= -\frac{1}{\gamma} \int_0^{t'} d\tau e^{(\int_0^\tau d\tau' k_{\tau'}/\gamma)} \exp\left(-\int_0^t d\tau k_\tau/\gamma\right). \quad (62)$$

Now, by taking the opposite of the time derivative with respect to  $t'$ , we obtain the response function of equation (54), which provides a verification of equation (9) for this particular example. Note that the response function does not have time translational symmetry (it is not solely a function of the time difference  $t - t'$ ) in the general case where the spring constant is time dependent. Furthermore, this response function has the property of being independent of the protocol of temperature variation.

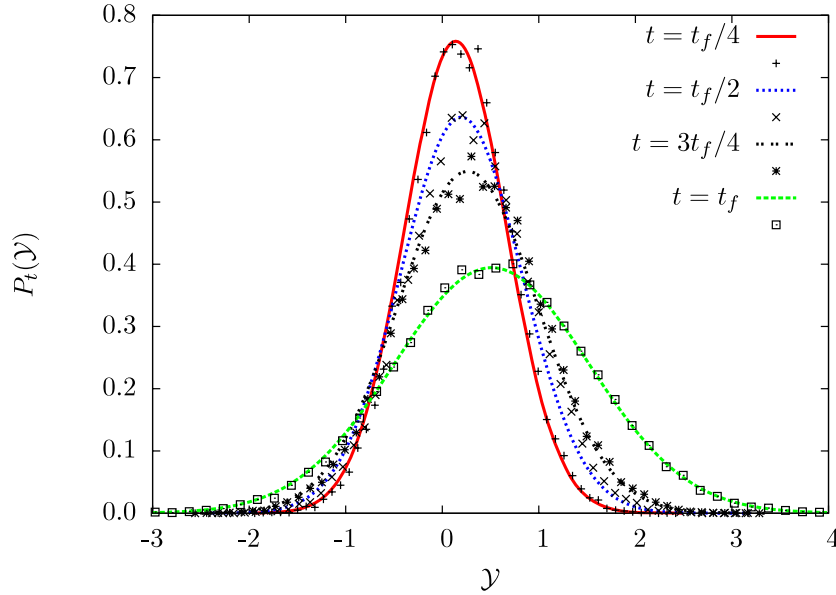
We obtain  $\mathcal{Y}_t$  from equations (20) and (57)

$$\mathcal{Y}_t = -\int_0^t d\tau \frac{\dot{h}_\tau I_\tau}{\sigma_\tau^2} (x_\tau - h_\tau I_\tau), \quad (63)$$

which is linear in  $x_\tau$  and hence is also a Gaussian variable. Its mean value and variance of  $\mathcal{Y}_t$  are respectively

$$\langle \mathcal{Y}_t \rangle = \int_0^t d\tau \frac{\dot{h}_\tau I_\tau}{\sigma_\tau^2} (h_\tau I_\tau - \mu_\tau), \quad (64)$$

$$\langle \mathcal{Y}_t^2 \rangle - \langle \mathcal{Y}_t \rangle^2 = \int_0^t d\tau \int_0^t d\tau' \frac{\dot{h}_\tau I_\tau \dot{h}_{\tau'} I_{\tau'}}{\sigma_\tau^2 \sigma_{\tau'}^2} (\langle x_\tau x_{\tau'} \rangle - \mu_\tau \mu_{\tau'}). \quad (65)$$



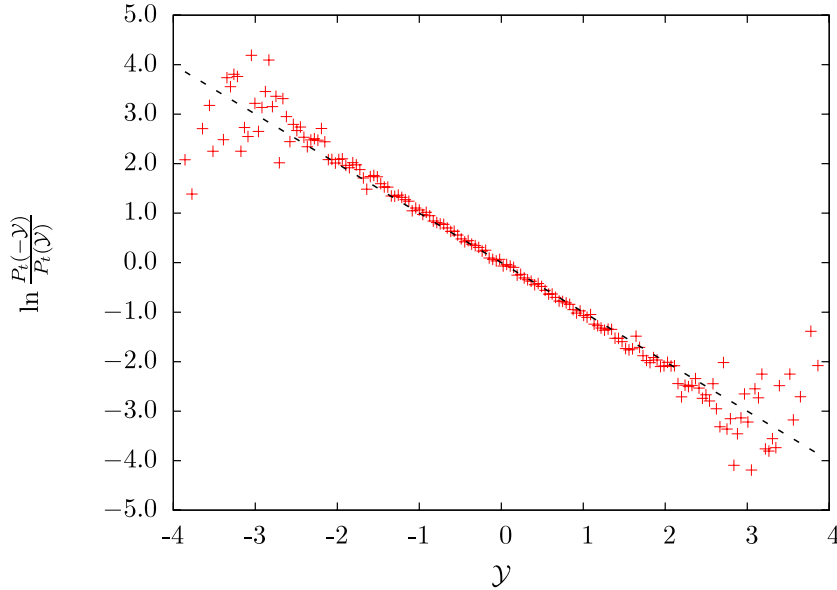
**Figure 1.** Probability density function  $P_t(\mathcal{Y})$  at four different times  $t$  for a particle in a harmonic trap obeying Langevin dynamics. The symbols represent an estimate of  $P_t(\mathcal{Y})$  based on  $10^5$  trajectories of total duration  $t_f = 5.12$ ; the solid line shows the Gaussian probability density which has the mean and variance given respectively by equations (64) and (65). The system is at  $t > 0$  in a non-equilibrium state due to an imposed time-dependent spring constant  $k_t = 5 + 2.5 \sin(\pi t/t_f)$ , and a time-dependent heat bath temperature, which is such that it is  $T_0 = 5$  at  $t = 0$  and  $T_t = 1$  for  $t > 0$ . The friction coefficient is  $\gamma = 1$ . This system is further perturbed by a force, according to the protocol  $h_t = 5 \sin(\pi t/t_f)$ .

At the end, after transforming equation (65) using an integration by parts, we find that  $\langle \mathcal{Y}_t^2 \rangle - \langle \mathcal{Y}_t \rangle^2 = 2 \langle \mathcal{Y}_t \rangle$ . Since  $\mathcal{Y}_t$  is a Gaussian process, this relation implies that the probability density function  $P_t(\mathcal{Y})$  satisfies the following detailed fluctuation relation:

$$\frac{P_t(\mathcal{Y})}{P_t(-\mathcal{Y})} = \exp(\mathcal{Y}), \quad (66)$$

which we have also confirmed through a numerical determination of the distribution of  $\mathcal{Y}_t$  as shown in figures 1 and 2.

Remarkably, this detailed fluctuation relation holds although no dual process has been invoked. Thus, this relation differs in an essential way from the generalized Crooks theorem given in [21] (see equation (133)). Another difference between equation (66) and the generalized Crooks theorem of that reference is that in the generalized Crooks relation, the initial condition in the forward or backward processes are taken according to the distribution  $\pi_0(x, h_0)$  and  $\pi_t(x, h_t)$  respectively. In contrast to this, in equation (66) the initial condition in which the system is prepared results from the application of unperturbed dynamics. For this reason, one may say that equation (66) is closer to a relation of the Bochkov and Kuzovlev type than to a Crooks relation [39].



**Figure 2.** Numerical test of the detailed fluctuation relation of equation (66) satisfied by the probability distribution of  $\mathcal{Y}$ . Here  $t = t_f = 5.12$ , and all the other parameters are the same as in figure 1.

Finally, we would like to emphasize that equation (66) is a very general result for linear Langevin dynamics. We have checked in appendix B that equation (66) can indeed be extended to a general multidimensional linear Langevin dynamics.

### 3.2. The 1D Ising model with Glauber dynamics

*3.2.1. Introduction.* We now move to a more complex system with many interacting degrees of freedom, which will allow for phase transitions and ordering phenomena absent from the previous example. The system is the Ising–Glauber chain in 1D, which when subjected to a temperature quench, is a paradigm for coarsening dynamics [40]. For this system, explicit exact expressions for the correlation and response functions have been obtained; and the ratio between these two quantities admits a non-trivial limit, which is a universal quantity in the case of a quench to the critical temperature [41]. Multipoint correlation functions have also been calculated analytically in order to test theoretical ideas about the dynamic heterogeneities of glasses [42, 43]. For more general spin systems, the correlations or response cannot be obtained analytically, but the response function has been shown to be related to correlations characteristic of the non-perturbed system [13, 14, 44, 45], a conceptual progress but also a definite advantage for numerical simulations as compared to previous methods. In the following, we illustrate the framework of a modified fluctuation-dissipation theorem presented in the previous sections for the Glauber–Ising chain subjected to a quench of temperature. Using analytical calculations, we first show that we can recover the known exact response function using this formalism. We then present some numerical simulations to confirm the theoretical expectations.



3.2.2. *Definition of the rates.* This Ising–Glauber chain is made of  $L$  Ising spins  $\sigma_i = \pm 1$  with  $i = 1, \dots, L$  in one dimension, and is described by the following Hamiltonian:

$$\mathcal{H}(\{\sigma\}) = -J \sum_{i=1}^L \sigma_i \sigma_{i+1} - H_m \sigma_m, \quad (67)$$

where  $J$  is the coupling constant and  $H_m$  a magnetic field which acts on the spin  $m$ . We assume periodic boundary conditions. The magnetic field  $H_m$  will be the only control parameter. In principle, we could allow for many control parameters corresponding to magnetic fields present on any lattice site, but since we are mainly interested in the linear response regime, we restrict ourselves to the case where this magnetic field only acts on the spin  $m$ . The Ising chain is assumed to be initially in equilibrium at an infinite temperature. At  $t = 0$ , it is subjected to an instantaneous quench which brings the temperature to  $T$ , and the system to a non-equilibrium state. The system further evolves after the quench from the time 0 to the time  $t'$  where the small magnetic field  $H_m$  is turned on. Therefore we assume that  $H_m(t) = H_m \theta(t - t')$ , with  $t > 0$  and  $\theta$  the Heaviside function.

The probability of finding the system in the state  $\{\sigma\} = \{\sigma_1, \dots, \sigma_L\}$  at time  $t$ ,  $P_t(\{\sigma\})$ , obeys the following master equation:

$$\frac{\partial P_t(\{\sigma\})}{\partial t} = - \sum_i w^{H_m}(\{\sigma\}, \{\sigma\}^i) P_t(\{\sigma\}) + \sum_i w^{H_m}(\{\sigma\}^i, \{\sigma\}) P_t(\{\sigma\}^i), \quad (68)$$

where  $w^{H_m}(\{\sigma\}, \{\sigma\}^i)$  is the rate of jumping from the configuration  $\{\sigma\}$  to the configuration  $\{\sigma\}^i = \{\sigma_1, \dots, -\sigma_i, \dots, \sigma_L\}$ .

Following [41], we choose the rates in the presence or absence of a field to be given respectively by

$$w^{H_m}(\{\sigma\}, \{\sigma\}^i) = \frac{\alpha}{2} (1 - \sigma_i \tanh(\beta J (\sigma_{i-1} + \sigma_{i+1}) + \beta H_m \delta_{im})), \quad (69)$$

$$w(\{\sigma\}, \{\sigma\}^i) = \frac{\alpha}{2} \left( 1 - \sigma_i \frac{\gamma}{2} (\sigma_{i-1} + \sigma_{i+1}) \right), \quad (70)$$

where  $\alpha$  denotes the inverse characteristic timescale of the transitions (which we take below to be equal to 1),  $\gamma = \tanh(2\beta J)$  and  $\beta = 1/T$ , the inverse of the temperature after the quench. Note that these rates depend on time (for  $t > 0$ ) only via the control parameter  $H_m$ . It is important to also realize that this form of the rates is just one of the possible choices compatible with the detailed balance condition, which imposes that

$$\frac{w^H(\{\sigma\}, \{\sigma'\})}{w^H(\{\sigma'\}, \{\sigma\})} = \frac{e^{-\beta \mathcal{H}(\{\sigma'\})}}{e^{-\beta \mathcal{H}(\{\sigma\})}}. \quad (71)$$

Other forms are possible; for instance, equation (46) corresponds to a different acceptable choice, which is the one made in [13] while equations (69) and (70) are the choice of [41, 45].

3.2.3. *Analytical verification of the MFDT.* Unlike in the previous example of a particle obeying Langevin dynamics, where the non-stationary probability distribution  $\pi_t$  was analytically solvable even in the presence of the perturbation due to the assumption of a harmonic potential, in the present problem, the probability distribution  $\pi_t(\{\sigma\}, H_m)$  for finding the system in an arbitrary configuration  $\{\sigma\}$  at time  $t$  with a constant magnetic

field  $H_m$  applied from  $t = 0$  is difficult to obtain even in the absence of a magnetic field ( $H_m = 0$ ). As we show below, this distribution is not required for evaluating the response function for a general one spin-dependent observable  $A(\sigma_n)$ , because in this particular case only the reduced one-spin distribution  $\pi_t(\sigma_n, H_m) = \sum_{\{\sigma\} \neq \sigma_n} \pi_t(\{\sigma\}, H_m)$  is needed and, fortunately, this reduced distribution can be calculated analytically. We define the response function as

$$R_{n-m}(t, t') = T \left. \frac{\delta \langle A(\sigma_n(t)) \rangle_{[H_m]}}{\delta H_m(t')} \right|_{H_m \rightarrow 0}, \quad (72)$$

which contains an extra factor  $T$  with respect to the definition used in previous sections. The introduction of this extra factor is purely a matter of convenience, but historically it was introduced in order to provide the response function with a well defined limit when  $T \rightarrow 0$ . According to the MFDT of equation (9), this response function is

$$\begin{aligned} R_{n-m}(t, t') &= T \frac{d}{dt'} \sum_{\sigma_n, \{\sigma'\}} A(\sigma_n) \rho(\sigma_n t | \{\sigma'\} t') \rho_{t'}(\{\sigma'\}) \left. \frac{\partial_{H_m} \pi_{t'}(\{\sigma'\}, H_m)}{\pi_{t'}(\{\sigma'\}, H_m)} \right|_{H_m \rightarrow 0}, \\ &= T \frac{d}{dt'} \sum_{\sigma_n, \{\sigma'\}} A(\sigma_n) \rho(\sigma_n t | \{\sigma'\} t') \partial_{H_m} \pi_{t'}(\{\sigma'\}, H_m) \Big|_{H_m \rightarrow 0}. \end{aligned}$$

where in the last step, we used  $\pi_{t'}(\{\sigma'\}, 0) = \rho_{t'}(\{\sigma'\})$ .

To progress, we need an explicit expression for the propagator  $\rho(\sigma_n t | \{\sigma'\} t')$ , which is the probability of finding the system with spin  $n$  in state  $\sigma_n$  at time  $t$  in the unperturbed dynamics given that the system was in the state  $\{\sigma'\}$  at time  $t'$ . In fact, this propagator is directly related to the average magnetization at time  $t$  when the system starts in the state  $\{\sigma'\}$  at time  $t'$ , namely  $\langle \sigma_n(t) \rangle_{\{\sigma\}(t')=\{\sigma'\}}$ :

$$\rho(\sigma_n t | \{\sigma'\} t') = \frac{1}{2} (\sigma_n \langle \sigma_n(t) \rangle_{\{\sigma\}(t')=\{\sigma'\}} + 1). \quad (73)$$

From equation (68), one can show that this average magnetization in the absence of an applied field satisfies [40]

$$\langle \sigma_n(t) \rangle_{\{\sigma\}(t')=\{\sigma'\}} = \sum_k G_{n-k}(t - t') \sigma'_k, \quad (74)$$

where  $G_k(t)$  is the Green function of the problem. In the following, we consider the thermodynamic limit  $L \rightarrow \infty$ , in which case, the Green function can be written as  $G_k(t) = e^{-t} I_k(\gamma t)$ , in terms of  $I_k$  the modified Bessel function. It follows from the above two equations that

$$\rho(\sigma_n t | \{\sigma'\} t') = \frac{\sigma_n}{2} \sum_k \sigma'_k G_{n-k}(t - t') + \frac{1}{2}. \quad (75)$$

After substituting this expression in equation (72), one obtains

$$R_{n-m}(t, t') = T \left. \frac{d}{dt'} \sum_{\sigma_n, \{\sigma'\}} A(\sigma_n) \partial_{H_m} \pi_{t'}(\{\sigma'\}, H_m) \right|_{H_m \rightarrow 0} \left[ \frac{\sigma_n}{2} \sum_k \sigma'_k G_{n-k}(t - t') + \frac{1}{2} \right]. \quad (76)$$

The fact that the observable  $A(\sigma_n)$  only depends on a single spin  $\sigma_n$  leads to a simplification of the expression above since it is possible to sum over all spins in  $\{\sigma'\}$

except for the  $k$ th spin of that set. The second term in the bracket of equation (76) vanishes due to the normalization condition  $\sum_{\{\sigma'\}} \pi_{t'}(\{\sigma'\}, H_m) = 1$ , and we obtain

$$R_{n-m}(t, t') = T \frac{d}{dt'} \sum_k \sum_{\sigma'_k, \sigma_n} \frac{1}{2} \sigma'_k \sigma_n A(\sigma_n) G_{n-k}(t-t') \partial_{H_m} \pi_{t'}(\sigma'_k, H_m) |_{H_m \rightarrow 0}. \quad (77)$$

This equation shows that the response now only depends on the reduced one-spin distribution  $\pi_t(\sigma_n, H_m) = \sum_{\{\sigma\} \neq \sigma_n} \pi_t(\{\sigma\}, H_m)$ , which can be obtained analytically at first order in the applied field  $H_m$  from the magnetization. Indeed, in the presence of a field, the magnetization at a time  $t > 0$  is

$$\langle \sigma_n(t) \rangle_{\{\sigma\}(0)=\{\sigma'\}} \simeq \sum_k G_{n-k}(t) \sigma'_k + \beta H_m \int_0^t G_{n-m}(t-t') K(t') dt', \quad (78)$$

where  $K(t) = 1 - \gamma^2/2(1 + \langle \sigma_{n+1}(t) \sigma_{n-1}(t) \rangle)$ , a correlation function which is known analytically. For instance, in the case of a  $T = 0$  quench where  $\gamma(t) = 1$  for  $t > 0$ , we have  $K(t) = e^{-2t}(I_0(2t) + I_2(2t) + 2I_1(2t))/2$  [46].

Since the chain is initially at infinite temperature, the average magnetization of spin  $k$  at time  $t = 0$  vanishes for all  $k$ . It follows that the first term of equation (78) vanishes when the averaging over the initial condition is performed. Using equation (75), we obtain the distribution  $\pi_t(\sigma_n, H_m)$  at first order in the applied field:

$$\pi_t(\sigma_n, H_m) \simeq \frac{1}{2} + \frac{1}{2} \beta H_m \sigma_n \int_0^t G_{n-m}(t-t') K(t') dt'. \quad (79)$$

We can use this distribution to write the response function in equation (77) in a more explicit form. After summing over  $\sigma'_k$ , we have

$$\begin{aligned} R_{n-m}(t, t') &= \frac{d}{dt'} \sum_{k, \sigma_n} \frac{1}{2} \sigma_n A(\sigma_n) G_{n-k}(t-t') \int_0^{t'} G_{k-m}(t'-u) K(u) du \\ &= \frac{A(1) - A(-1)}{2} \frac{d}{dt'} \int_0^{t'} \sum_k G_{n-k}(t-t') G_{k-m}(t'-u) K(u) du \\ &= \frac{A(1) - A(-1)}{2} G_{n-m}(t-t') K(t'), \end{aligned} \quad (80)$$

which does indeed agree with the response function obtained in [41] in the case  $A(\sigma_n) = \sigma_n$ .

Besides recovering the known response function of the Glauber–Ising chain, we can also investigate the separate contributions of the two local currents  $j_{t'}$  and  $\nu_{t'}$  introduced in section 2.4. It is straightforward to show using the detailed balance condition of equation (71) that the first term in the MFDT, namely  $R_{\text{eq}}(t, t')$ , can be written in a form similar to that of the equilibrium FDT (note that in the equation below, the index  $j$  represents the discrete times when jumps occur):

$$\partial_{H_m} \Delta S_t^r(\{\sigma\}_t, [h]) |_{H_m=0} = \sum_{j=1}^N \partial_{H_m} \ln \frac{w_{\tau_j}^{H_m}(\{\sigma\}(j-1), \{\sigma\}(j))}{w_{\tau_j}^{H_m}(\{\sigma\}(j), \{\sigma\}(j-1))} \Big|_{H_m=0} \quad (81)$$

$$= \beta \sum_{j=1}^N (\sigma_m(j) - \sigma_m(j-1)), \quad (82)$$

$$= \beta (\sigma_m(N) - \sigma_m(0)) = \beta (\sigma_m(t) - \sigma_m(0)). \quad (83)$$

Substituting this in equation (33), one obtains the equilibrium part of the response for a general multi-spin observable  $A(\{\sigma\}_t)$ :

$$R_{\text{eq}}(t, t') = \beta \frac{d}{dt'} \langle A(\{\sigma\}_t) \sigma_m(t') \rangle. \quad (84)$$

Alternately, one can also recover this result through an evaluation of the local current  $j_{t'}$  using equation (37).

For the term associated with the local current  $\nu(t')$ , one can show either from the decomposition of the stochastic entropy into two terms or from equations (41) and (42) that the expected part of the MFDT is recovered, or in other words that  $R_{\text{neq}}(t, t') = R(t, t') - R_{\text{eq}}(t, t')$ , where  $R(t, t')$  is the response given in equation (80) for the case of a one-spin observable  $A(\sigma_n)$ . In the literature [13, 14], the term  $R_{\text{neq}}(t, t')$  is called the asymmetry; its precise form depends on the specific form of the rates (unlike the first term which only depends on the ratio of forward to backward rates) and it vanishes under equilibrium conditions. The present derivation also makes it clear that there is in principle a physical observable associated with this term, namely  $\Delta s_t^{\text{tot}}(\{\sigma\}_t, [h])$ .

### 3.3. Numerical verification

As mentioned above, the distribution  $\pi_t(\{\sigma\}, H_m)$  does not seem to be accessible analytically. In order to test our framework, we have thus determined these distributions numerically from simulations for various values of  $H_m$  and  $\{\sigma\}$ . Then, we calculated the response function  $R(t, t')$  via equation (9) and using trajectories which were simulated according to the dynamics in the absence of a magnetic field. In figures 3 and 4, the integrated response defined by

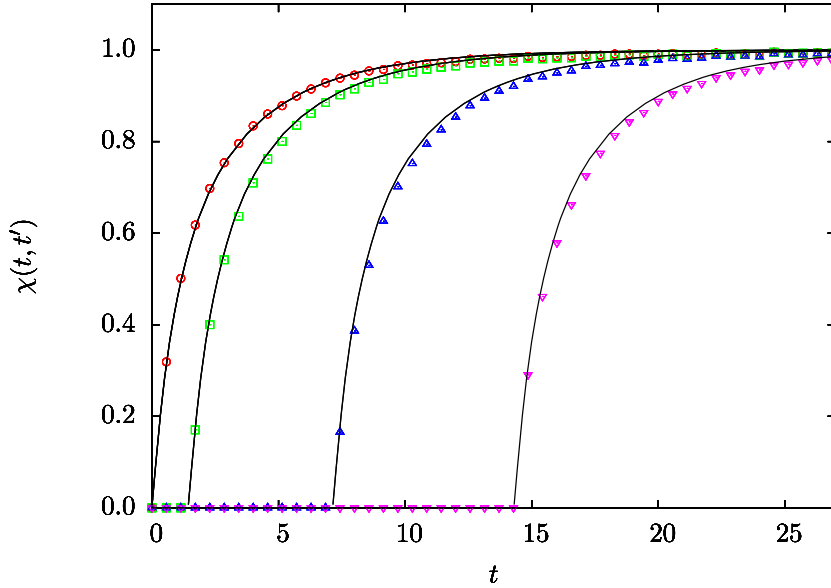
$$\chi_{n-m}(t, t') = \int_{t'}^t d\tau R_{n-m}(t, \tau), \quad (85)$$

is shown, where the symbols represent the response function obtained from such simulations at zero field via equation (9) and the solid line is the analytical expression obtained from equation (80). Since equation (80) takes a simple form in Laplace space [41], this solid line was obtained through a numerical inverse Laplace transform of that equation.

In the simulations at zero field, we have used a small system size of  $L = 14$ . It is difficult to go to significantly larger sizes with the present algorithm, because the numerical determination of the distribution  $\pi_t(\{\sigma\}, H_m)$  rapidly becomes a difficult task in large systems given that the configuration space grows as  $2^N$ .

## 4. Conclusion

In this paper, we have presented a framework which can be used to generalize the fluctuation-dissipation theorem to non-equilibrium systems obeying Markovian dynamics. We have shown, using alternatively the linear response theory or a first-order development of a fluctuation relation, that the main result of [22] for systems in a non-equilibrium stationary state is generalizable to systems which are near non-stationary states. This generalization is important because it restores a fluctuation-dissipation theorem to the

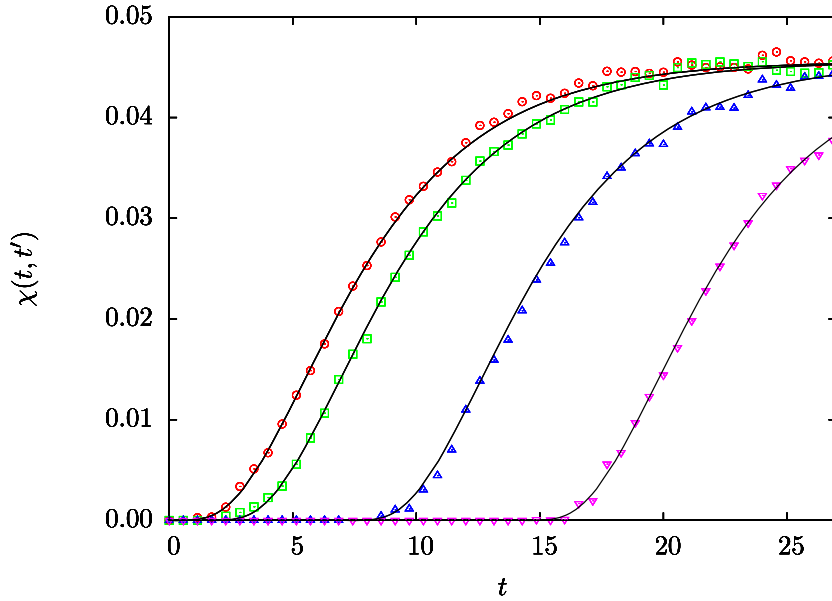


**Figure 3.** Integrated response functions  $\chi(t, t')$  versus time  $t$  for a step protocol for the magnetic field  $H_m = 0.05$  starting at various values of the waiting time  $t'$  after the initial quench at  $t = 0$ . The different values of  $t'$  are  $t' = 0$  for circles,  $t' = 1.43$  for squares,  $t' = 7.14$  for triangles and  $t' = 14.3$  for inverted triangles. The parameters are the following:  $J = 0.5$ ,  $\alpha = 1$ ,  $T = 1$  and  $L = 14$ . The response is calculated for the same spin as the magnetic field is applied to, here  $n = m = 3$ . The averages have been done with  $10^6$  trajectories of 400 time steps of length  $dt = 0.07$ . The continuous lines stand for the integrated response obtained analytically, while the symbols were obtained from the MFDT of equation (9) using the unperturbed dynamics.

form of a unique correlation between physical observables and it opens up many new possibilities for applying this framework to experiments.

In fact, this framework is applicable to systems which are prepared in a non-equilibrium state, and which are then further probed through the application of a time-dependent control parameter. This situation is typically the one encountered in studies of slowly relaxing or ageing systems, but it is also frequently encountered in biological systems. One outcome of our approach is that it is possible to replace this complicated problem by a somewhat simpler problem, namely the problem of determining the probability distribution  $\pi_t(c, h)$  for finding the system in a non-equilibrium state  $c$  but with a time-independent perturbation  $h$ . Having to consider only a time-independent perturbation for probing a non-equilibrium system should be a definite advantage from both experimental and theoretical points of view.

Our study of a Brownian particle in a harmonic potential and subjected to a quench of temperature raises the question of the validity of fluctuation relations for particles in contact with a non-equilibrium bath. These kinds of studies may be important for understanding the non-equilibrium fluctuations of a Brownian particle confined in a gel as in the experiment of [47]. We hope that our work will trigger further studies on the applications of stochastic thermodynamics to the characterization of non-equilibrium



**Figure 4.** Integrated response functions  $\chi(t, t')$  versus time  $t$  under the same conditions as in figure 3, except that now the response is evaluated for a different spin ( $n = 7$ ), not that where the magnetic field is applied ( $n = 3$ ), and the averages have been done with  $10^7$  trajectories.

systems, and in particular for non-equilibrium systems which result from contact with a non-equilibrium bath or from a coarsening process.

### Acknowledgments

We acknowledge stimulating discussions with F Krzakala and B Wynants.

### Appendix A. The link with previous formulations

#### Equivalence between different expressions for the work-like functional

As mentioned in the main text, the functional  $\mathcal{Y}_t$  defined in equation (20) has appeared before in [33]. We explain here how to make contact with the different forms. The functional denoted here as  $\mathcal{Y}_t$  corresponds to the one called  $W_t$  in equation (24) of this reference. In order to see this correspondence, one should choose the quantity defined as  $f_t$  in [33] to be equal to  $\pi_t(c, h_t)$ . With this choice,

$$W_t \equiv \int_0^t d\tau \left( \pi_\tau(\cdot, h_\tau)^{-1} (L_\tau^{h_\tau})^\dagger [\pi_\tau(\cdot, h_\tau)] - \frac{\partial}{\partial \tau} (\ln \pi_\tau(\cdot, h_\tau)) \right) (c_\tau) \quad (\text{A.1})$$

$$\begin{aligned} &= \int_0^t d\tau \left( \pi_\tau(c_\tau, h_\tau)^{-1} (\partial_\tau \pi_\tau)(c_\tau, h_\tau) - (\partial_\tau \ln \pi_\tau)(c_\tau, h_\tau) - \dot{h}_\tau \partial_{h_\tau} \ln \pi_\tau(c_\tau, h_\tau) \right) \\ &= \int_0^t d\tau \dot{h}_\tau \partial_{h_\tau} \ln \pi_\tau(c_\tau, h_\tau), \end{aligned} \quad (\text{A.2})$$

where in the first line, the dagger stands for adjoint operator and the  $\cdot$  indicates that the expression is to be understood as a matrix product before evaluation on the trajectory  $c_\tau$ . This calculation shows that  $W_t$  does indeed coincide with  $\mathcal{Y}_t$  defined in equation (20).

### Equivalence between the discrete and continuous formulations of the MFD

In [17, 33], a modified fluctuation dissipation has been derived for general continuous diffusion processes. Here we show that the present framework formulated for discrete jump processes leads to the same results, when the appropriate continuous limit of the master equation is taken [48].

We shall assume the same parametrization of the rates as is given in equation (46). The equilibrium contribution in the response,  $R_{\text{eq}}(t, t')$ , can be written in the same way as in [33]; therefore, we shall focus on the other non-equilibrium contribution, namely  $R_{\text{neq}}(t, t')$ . For simplicity, let us consider a nearest-neighbour random walk on a 1D lattice, in which a random walker at position  $m$  can only jump to neighbouring sites  $m \pm 1$ . We denote the actual distance between all the sites by  $\epsilon$ . The master equation is

$$\frac{d}{dt}\rho_t(m) = J_t(m+1, m) - J_t(m-1, m) \quad (\text{A.3})$$

with  $J_t(m, m+1) = \rho_t(m)w_t(m, m+1) - \rho_t(m+1)w_t(m+1, m)$ . Using the Taylor expansion  $\rho_t(m \pm 1) = \rho_t(m) \pm \partial_m \rho_t(m)$ , we can rewrite the discrete currents as

$$J_t(m, m \pm 1) = w_t(m, m \pm 1)\rho_t(m) - w_t(m \pm 1, m)(\rho_t(m) \pm \partial_m \rho_t(m)). \quad (\text{A.4})$$

To establish a link between these discrete currents and the current arising in the corresponding Fokker–Planck equation, we introduce the notation  $u_t(m+1) = w_t(m, m+1) - w_t(m+1, m)$  and  $2D_t(m+1) = w_t(m, m+1) + w_t(m+1, m)$ , so the discrete currents are

$$J_t(m, m+1) = \rho_t(m)(u_t(m) + \partial_m u_t(m)) - w_t(m+1, m)\partial_m \rho_t(m) \quad (\text{A.5})$$

$$J_t(m, m-1) = -\rho_t(m)u_t(m) + w_t(m-1, m)\partial_m \rho_t(m). \quad (\text{A.6})$$

In the continuous limit,  $\epsilon \rightarrow 0$ ,

$$\begin{aligned} \rho_t(m) &\sim \epsilon \rho_t(x), & \partial_m &\sim \epsilon \partial_x, \\ u_t(m) &\sim u_t(x)/\epsilon, & D_t(m) &\sim D_t(x)/\epsilon^2, \end{aligned} \quad (\text{A.7})$$

and the discrete currents can be related to the current  $J_t(x)$  entering the Fokker–Planck equation:

$$J_t(m, m \pm 1) \sim \pm u_t(x)\rho_t(x) \mp D_t \partial_x \rho_t(x) = \pm J_t(x). \quad (\text{A.8})$$

Let us recall the expression given in equation (42) for the local current  $\nu_{t'}(m)$ :

$$\nu_{t'}(m) = \frac{J_{t'}(m, m+1)\partial_h w_{t'}^h(m, m+1)|_{h \rightarrow 0}}{\rho_{t'}(m)} \quad (\text{A.9})$$

$$+ \frac{J_{t'}(m, m-1)\partial_h w_{t'}^h(m, m-1)|_{h \rightarrow 0}}{\rho_{t'}(m)}. \quad (\text{A.10})$$



Using the expression for the rates given in equation (46), we have  $\partial_h w_{t'}^h(m, m \pm 1) = (O(m \pm 1) - O(m))\beta/2 \sim \pm\beta\epsilon\partial_x O(x)/2$ . Inserting this in the above equation and using equation (A.8), one obtains the following expression for the local current  $\nu_{t'}$  in the continuous limit:

$$\nu_{t'}(x) = \frac{\beta J_{t'}(x)}{\rho_{t'}(x)} \partial_x O(x). \quad (\text{A.11})$$

Combining this with the first term in the response given in equation (48), one recovers from this the response function given in [33], namely

$$R(t, t') = \beta \frac{d}{dt'} \langle A(x_t) O(x_{t'}) \rangle - \beta \left\langle A(x_{t'}) \frac{J_{t'}(x_{t'})}{\rho_{t'}(x_{t'})} \partial_{x_{t'}} O(x_{t'}) \right\rangle. \quad (\text{A.12})$$

## Appendix B. Generalization to multidimensional linear Langevin dynamics

We now extend the results obtained in section 3.1 for Langevin dynamics with one degree of freedom to many dimensions. To that end, we start with the following multidimensional linear Langevin equation:

$$\dot{x}_t = N_t x_t + h_t + \eta_t, \quad (\text{B.1})$$

where  $\eta_t$  is a white noise such that  $\langle \eta_t \rangle = 0$  and  $\langle \eta_t \eta_{t'} \rangle = 2T_t \Gamma_t \delta(t - t')$ . We denote as  $T_t$  a time-dependent temperature, as  $\Gamma_t$  a positive and symmetric matrix, and as  $N_t$  an arbitrary matrix.

For any  $t \geq s$ , we introduce the following matrix:

$$T(t, s) = \overleftarrow{\exp} \left( \int_s^t du N_u \right), \quad (\text{B.2})$$

where the exponential is to be understood as time ordered. This matrix satisfies the useful identity that for any  $t \geq s \geq u$ ,  $T(t, s)T(s, u) = T(t, u)$ . We also introduce the matrix

$$D(t, s) = 2 \int_s^t du T(t, u) T_u \Gamma_u T^\dagger(t, u), \quad (\text{B.3})$$

where the dagger denotes the transpose of a matrix.

The solution of equation (B.1) is

$$x_t = T(t, 0)x_0 + \int_0^t ds T(t, s)(h_s + \eta_s). \quad (\text{B.4})$$

We assume that the initial condition  $x_0$  is distributed according to a Gaussian, with a characteristic mean  $m_0 \equiv \langle x_0 \rangle$ , and an initial covariance matrix  $V_0^{ij} = \langle (x_0 - \langle x_0 \rangle)^i (x_0 - \langle x_0 \rangle)^j \rangle$ . From equation (B.4), we deduce the mean

$$m_t = \langle x_t \rangle = T(t, 0)m_0 + \int_0^t ds T(t, s)h_s, \quad (\text{B.5})$$

and the symmetric matrix of the covariance

$$V_t = \langle (x_t - m_t)(x_t - m_t) \rangle = T(t, 0)V_0 T^\dagger(t, 0) + D(t, 0). \quad (\text{B.6})$$

As in the 1D case, we note that in this case too,  $V_t$  is independent of the perturbation  $h_t$ . Since  $x_t$  is a Gaussian random variable, its distribution is

$$P_t(x, [h_t]) = (\det(2\pi V_t))^{-1/2} \exp(-\frac{1}{2}(x - m_t)V_t^{-1}(x - m_t)), \quad (\text{B.7})$$

and therefore

$$\pi_t(x, h_t) = (\det(2\pi V_t))^{-1/2} \exp(-\frac{1}{2}(x - \tilde{m}_t)V_t^{-1}(x - \tilde{m}_t)), \quad (\text{B.8})$$

with

$$\tilde{m}_t = T(t, 0)m_0 + \left( \int_0^t ds T(t, s) \right) h_t. \quad (\text{B.9})$$

Since

$$\partial_{h_t} (\ln \pi_t(x, h_t)) = \left( \int_0^t ds T^\dagger(t, s) \right) V_t^{-1}(x - \tilde{m}_t), \quad (\text{B.10})$$

the functional  $\mathcal{Y}$  of interest here takes the form

$$\mathcal{Y}_T = - \int_0^T dt \dot{h}_t \left( \int_0^t ds T^\dagger(t, s) \right) V_t^{-1}(x_t - \tilde{m}_t). \quad (\text{B.11})$$

From this, we obtain the average

$$\langle \mathcal{Y}_T \rangle = - \int_0^T dt \dot{h}_t \left( \int_0^t ds T^\dagger(t, s) \right) V_t^{-1}(m_t - \tilde{m}_t), \quad (\text{B.12})$$

$$= \int_0^T dt \dot{h}_t \left( \int_0^t ds T^\dagger(t, s) \right) V_t^{-1} \left( \int_0^t du T(t, u) (h_t - h_u) \right), \quad (\text{B.13})$$

and so

$$\mathcal{Y}_T - \langle \mathcal{Y}_T \rangle = - \int_0^T dt \dot{h}_t \left( \int_0^t ds T^\dagger(t, s) \right) V_t^{-1}(x_t - m_t). \quad (\text{B.14})$$

Through explicit calculation, one can verify that

$$\begin{aligned} & \langle (\mathcal{Y}_T - \langle \mathcal{Y}_T \rangle) (\mathcal{Y}_T - \langle \mathcal{Y}_T \rangle) \rangle \\ &= 2 \int_0^T dt \dot{h}_t \left( \int_0^t ds T^\dagger(t, s) \right) V_t^{-1} \int_0^t dt' T(t, t') \left( \int_0^{t'} ds T(t', s) \right) \dot{h}_{t'}. \end{aligned} \quad (\text{B.15})$$

Using an integration by parts, one can check that

$$\int_0^t dt' T(t, t') \left( \int_0^{t'} ds T(t', s) \right) \dot{h}_{t'} = \int_0^t dt' T(t, t') (h_t - h_{t'}). \quad (\text{B.16})$$

Taken together, these two equations imply the relation

$$\langle (\mathcal{Y}_T - \langle \mathcal{Y}_T \rangle) (\mathcal{Y}_T - \langle \mathcal{Y}_T \rangle) \rangle = 2 \langle \mathcal{Y}_T \rangle. \quad (\text{B.17})$$

Since  $\mathcal{Y}_T$  is a Gaussian variable, the detailed fluctuation relation given in equation (66) follows from this for the general linear Langevin dynamics.

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