

Fluctuations and response from a Hatano and Sasa approach

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Abstract

In this commentary paper, we present some of the main ideas on deriving a modified fluctuation-dissipation theorem off equilibrium, which in the end can all be related to an approach based on a generalized Hatano–Sasa relation. This generalized Hatano–Sasa relation also contains an interesting inequality, which can be viewed as a generalization of the second law of thermodynamics to transitions between non-equilibrium states.

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1. Introduction

The fluctuation-dissipation theorem (FDT) is a cornerstone of statistical physics, since it allows one to extract linear response properties from the measurement of equilibrium fluctuations [1]. Beyond the equilibrium regime, the relation between the response and correlations does not take a simple and universal form. For this reason, the search for generalizations of the fluctuation-dissipation theorem off equilibrium has been an active topic both theoretically and experimentally for many years, as shown by a recent review [2]. On the experimental side, deviations from the fluctuation-dissipation theorem have been observed experimentally in many different systems: driven systems such as shaken granular matter, sheared fluids, manipulated colloidal systems, biological systems and glassy systems such as dense colloids or spin glasses [3–6].

For many years, these diverse systems appeared unrelated to each other and there was no sign of common universal features that could help construct a general theory. All this was changed fundamentally with the discovery of fluctuation relations, which are equalities valid for many classes of models and arbitrarily far from equilibrium, and which link distributions of various thermodynamic quantities [7–12]. Such results are particularly relevant for small systems, because small systems are characterized by large fluctuations. But, they are also useful for systems which are not small. Indeed, one can obtain through an expansion of the fluctuation theorems in the linear regime various modified fluctuation-dissipation theorems (MFDTs), which offer useful

generalizations of the fluctuation-dissipation theorem off equilibrium. Recently, three main routes have emerged to construct such generalizations.

- In the first route opened by Cugliandolo *et al* [3] and continued by Lippiello *et al* [4] and Diezemann [5], the response function is written as a sum of a time derivative of the correlation function (similar to the equilibrium FDT), plus an additive function, called the asymmetry, which vanishes under equilibrium conditions. Baiesi *et al* [13] took this idea much further by identifying the contribution of the entropy production in the response function and by introducing the traffic or dynamical activity which enters in the correlation function for the MFDT.
- In the second route opened by Speck and Seifert [14], the modified FDT near a non-equilibrium steady state (NESS) can be formulated in terms of the so-called local velocity, which originates from the local currents present in this NESS. A similar additive structure in MFDT was also found in the Harada–Sasa relation, which connects the energy dissipation with the violation of the FDT near an NESS [15], and in a modified Einstein relation found in studies of driven diffusive systems [16]. The idea of a local velocity was further extended to more general Langevin dynamics by Ch  trite *et al* [17], who also provided the MFDT with a Lagrangian frame interpretation [18]. In the end, it appears that the first and second routes are closely related [13, 19].
- In the third approach developed by Prost *et al* [20] based on an expansion of the Hatano–Sasa relation [12], the

modified fluctuation theorem valid near an NESS takes the standard equilibrium form except that it involves a new observable, a function of the NESS. This new observable is related to the system stochastic entropy, which here must be evaluated in the NESS [21, 22].

At first sight, the first and second approaches seem more general than the third one because they hold for general non-equilibrium situations, whereas the third one is limited to systems close to a non-equilibrium stationary state; but in fact, it is possible to extend also the third approach to a general non-equilibrium state as we have shown in [23]. In the end, it appears that the three approaches provide closely related formulations to generalize the FDT to non-equilibrium situations. This, of course, had to be the case since all these approaches lead in the end to the same response function.

In this commentary paper, we first recall the main ideas behind an MFDT using the third approach, which is based on a generalized Hatano–Sasa relation. This relation contains also an interesting inequality which generalizes the second law of thermodynamics to non-stationary states [24, 25]. We end the paper by a short discussion of possible experimental systems on which these ideas could be tested. This research has benefited from interactions with many participants during the Nordita program titled ‘Foundations and Applications of Non-equilibrium Statistical Mechanics’, which was conducted in September–October 2011.

2. The third route and the generalized Hatano–Sasa relation

We consider a system that is assumed to evolve according to a continuous-time Markovian dynamics of a pure jump type [26]. Let us introduce the transition rate $w_t(c, c')$ for the rate to jump from a state c to a state c' at time t . The subscript t in $w_t(c, c')$ describes processes that are non-stationary even in the absence of driving. The origin of such processes is arbitrary; they can result from a quench of some parameter like temperature or from an additional driving, which does not need to be specified. At time $t = 0$, an arbitrary driving protocol h_t is applied to the system, and we denote by $p_t(c, [h])$ the probability to observe the system in the state c at a time t in the presence of this driving. The evolution of the system for $t > 0$ is controlled by the generator $L_t^{h_t}$, which is defined as

$$L_t^{h_t}(c', c) = w_t^{h_t}(c', c) - \delta(c, c') \sum_{c''} w_t^{h_t}(c', c''), \quad (1)$$

where $w_t^{h_t}(c', c)$ is a transition rate in the presence of the driving $[h]$. Then $p_t(c, [h])$ is the solution of

$$\frac{dp_t(c, [h])}{dt} = \sum_{c'} p_t(c', [h]) L_t^{h_t}(c', c). \quad (2)$$

The notation $p_t(c, [h])$ emphasizes that this probability distribution depends functionally on the whole protocol history $[h]$ up to time t . We assume that, at $t = 0$ there is no driving, so that $p_0(c, [h_0]) = p_0(c)$. We also note that, in practice, the driving $[h]$ may not start immediately at $t = 0^+$

but may be turned on only later, after a certain time (such a delay corresponds to what is called the waiting time in the context of aging systems).

We now introduce a different probability distribution denoted by $\pi_t(c, h)$, which represents the probability to observe the system in the state c at a time $t > 0$ in the presence of a constant (time-independent) driving h . In other words, $\pi_t(c, h)$ can be constructed from $p_t(c, [h])$ by freezing the time dependence in the driving $[h]$. This distribution, which will play a key role in the following, obeys the master equation

$$\left(\frac{\partial \pi_t}{\partial t}\right)(c, h) = \sum_{c'} \pi_t(c', h) L_t^h(c', c). \quad (3)$$

We emphasize that when the function $\pi_t(c, h)$ is evaluated on a point (c_t, h_t) of the trajectory $[c]$, one obtains a quantity which is distinct from the full solution $p_t(c, [h])$, because of the functional dependence in the driving.

By evaluating $\pi_t(c, h)$ on a trajectory $[c]$, one can construct the following functional:

$$\mathcal{Y}[c] = - \int_0^t dt \dot{h}_\tau \partial_h \ln \pi_\tau(c_\tau, h_\tau), \quad (4)$$

which has clear similarities with the functionals introduced by Jarzynski [9] and Hatano and Sasa [12]. Using a Feynman–Kac approach, which has also played a central role in the derivation of the Jarzynski relation [27], we have shown in [23] that this functional \mathcal{Y} obeys a generalized Hatano–Sasa relation:

$$\langle A_t(c_t, h_t) \exp(-\mathcal{Y}[c]) \rangle_{[h]} = \langle A_t(c_t, h_t) \rangle_{\pi_t}, \quad (5)$$

where $A_t(c_t, h_t)$ is a dynamic observable which depends on the configuration c_t and on the value of the control parameter at time t , namely h_t . Note that $\langle \dots \rangle_{\pi_t}$ and $\langle \dots \rangle_{[h]}$ represent, respectively, averages with respect to $\pi_t(c, h_t)$ or with respect to the perturbed dynamics corresponding to $p_t(c, [h])$. It is important to point out that equation (5) holds in the absence of a thermodynamic structure (there is no need for a first law of thermodynamics for instance) and without a reference stationary state.

We now discuss the consequences of the above fluctuation relation for the linear response theory near an arbitrary non-equilibrium state. The response function associated with $A_t(c_t, h_t)$ reads, for a perturbation applied at an earlier time $t' > 0$:

$$R(t, t') = \left. \frac{\delta \langle A_t(c_t, h_t) \rangle_{[h]}}{\delta h_{t'}} \right|_{h \rightarrow 0}, \quad (6)$$

where $\delta/\delta h_{t'}$ is our notation for functional derivatives. From the functional derivative with respect to $h_{t'}$ of the linear expansion of equation (5) for small \mathcal{Y} , one obtains the following MFDT valid near an arbitrary non-equilibrium state [23], which reads for $t > t' > 0$:

$$R(t, t') = - \frac{d}{dt'} \left(\partial_h \psi_{t'}(c_{t'}, h) \Big|_{h \rightarrow 0} A_t(c_t, h_t) \right), \quad (7)$$

with $\psi_{t'}(c, h) = -\ln \pi_{t'}(c, h)$. Indeed, the functional derivative of the right-hand side of equation (5) vanishes because $t > t'$ and the left-hand side produces the above

result. It is immediate to check that the response function takes the familiar form of the standard FDT when the unperturbed dynamics is at equilibrium because in that case, the distribution π_t becomes the equilibrium Boltzmann distribution.

The above relation qualifies for 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 by $\langle \dots \rangle$ as in the standard FDT [28]. It is important to point out that equation (7) was first derived by this method in [20] for the particular choice $A_t(c_t, h_t) = \partial_{h_t} \psi_t(c_t, h_t)$, in which case the MFDT takes a more symmetric form. In that work, it was assumed that the initial state is stationary and that the dynamics followed by the system at constant perturbation h is time independent ($L_t^h \equiv L^h$). It was later realized that the MFDT near a non-equilibrium stationary state holds in fact for a general observable $A_t(c_t, h_t)$ [22] and that the method used in [20] could be extended to such an arbitrary observable [21].

3. The second route and the origin of the additive structure in MFDT

The notion of stochastic entropy [29, 30] provides significant insights into MFDTs as shown recently by several authors [13, 21–23]. This notion is defined on a given trajectory but is distinct from the Kolmogorov–Sinai entropy [31], which is also a trajectory quantity. The latter is defined as the logarithm of the probability of a partition of a given trajectory into visited configurations in a specific order. The stochastic entropy involved here is a very different object which is constructed from the full solution of the Master/Fokker Planck equation of the problem, namely $p_t(c, [h])$, which becomes a trajectory-dependent random variable when evaluated on a specific configuration $c = c_t$ taken by the system at time t when following the trajectory $[c]$. The name ‘entropy’ is justified by the property that the average of the stochastic entropy is the Shannon entropy constructed from the distribution $p_t(c, [h])$ and by the fact that it is a state function just like the standard entropy. For the MFDT, it is in fact not required to know the ‘full’ stochastic entropy $-\ln p_t(c_t, [h]) = s_t(c_t, [h])$, because only the response to linear order in perturbation is needed. Therefore, only a reduced version of it is needed, which is built from $\pi_t(c, h)$ instead of $p_t(c, [h])$, where the former quantity is obtained from the latter by freezing the time dependence of the driving. In the following, we will need only this reduced stochastic entropy denoted by $\psi_t(c_t, h) = -\ln \pi_t(c_t, h)$ instead of the real stochastic entropy. This is the function which enters the response function of the MFDT in equation (7).

The additive structure in MFDTs that is apparent in the first and second routes mentioned in the introduction follows from a decomposition of the stochastic entropy, more precisely of its reduced counterpart. Indeed, $\Delta s_t = s_t(c_t, [h]) - s_0(c_0, [h_0])$ is a system trajectory entropy production, which can be written as $\Delta s_t = -\Delta s_t^r + s_t^{\text{tot}}$, in terms of the reservoir entropy production Δs_t^r (also called medium entropy in [30]) and the total entropy production

Δs_t^{tot} . From this decomposition, the response function in equation (7) splits into

$$\begin{aligned} R(t, t') &= -\frac{d}{dt'} \langle \partial_h \Delta s_{t'}^r |_{h \rightarrow 0} A_t(c_t, h_t) \rangle \\ &= R_{\text{eq}}(t, t') - R_{\text{neq}}(t, t'), \end{aligned} \quad (8)$$

where

$$R_{\text{eq}}(t, t') = \frac{d}{dt'} \langle \partial_h \Delta s_{t'}^r |_{h \rightarrow 0} A_t(c_t, h_t) \rangle \quad (9)$$

and

$$R_{\text{neq}}(t, t') = \frac{d}{dt'} \langle \partial_h \Delta s_{t'}^{\text{tot}} |_{h \rightarrow 0} A_t(c_t, h_t) \rangle, \quad (10)$$

where the derivative of the various stochastic entropies with respect to the control parameter h should be understood as an infinitesimal shift of the protocol by a global constant. In the previous decomposition, the first term $R_{\text{eq}}(t, t')$ is analogous to (and contains in the appropriate limit) the equilibrium FDT, whereas the second term $R_{\text{neq}}(t, t')$ represents an additive correction which vanishes at equilibrium.

Following [14], one can rewrite these response functions without time derivatives, by introducing two local currents $j_{t'}$ and $v_{t'}$. These currents depend on the unperturbed solution of the master equation $\rho_t(c)$, which is the probability to be in state c at time t . The generator of the unperturbed master equation is $L_t(c', c)$ defined as in equation (1) but with unperturbed rates $w_t(c, c')$ instead of perturbed ones $w_t^h(c, c')$. The two local currents $j_{t'}$ and $v_{t'}$ are then defined as [23]

$$j_{t'}(c') = \sum_c \frac{\rho_{t'}(c)}{\rho_{t'}(c')} w_{t'}(c, c') \partial_h \ln \frac{w_{t'}^h(c, c')}{w_{t'}^h(c', c)} \Big|_{h \rightarrow 0} \quad (11)$$

and

$$v_{t'}(c') = \sum_c \frac{J_{t'}(c', c)}{\rho_{t'}(c')} \partial_h \ln w_{t'}^h(c', c) \Big|_{h \rightarrow 0}. \quad (12)$$

Both currents $v_{t'}(c')$ and $j_{t'}(c')$ have the same average, which represents a physical current

$$\langle j_{t'}(c_{t'}) \rangle = \sum_{c, c'} J_{t'}(c', c) \partial_h \ln w_{t'}^h(c', c) \Big|_{h \rightarrow 0}, \quad (13)$$

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 . In terms of the local currents, the response takes the form

$$R(t, t') = \langle (j_{t'}(c_{t'}) - v_{t'}(c_{t'})) A_t(c_t, h_t) \rangle. \quad (14)$$

This response function has the same structure as that found by Speck and Seifert [14] in their original study of the MFDT near an NESS. In that work formulated for Langevin dynamics, Speck *et al* introduced a local velocity defined as the ratio of the stationary probability current divided by the stationary probability distribution of the NESS. This framework was later extended to general discrete Markovian models [32]. The current $v_{t'}$ introduced above can be viewed as a generalization of this local velocity to arbitrary non-equilibrium states. Note that such a decomposition in terms of the local currents $j_{t'}$ and $v_{t'}$ is not unique [22]. However, what seems to be common to all the formulations of MFDT is the additive structure of the response function,

which appears here as a result of the splitting of the stochastic entropy into two parts (medium and entropy production). As we shall see in the next section, another related decomposition enters the first route.

4. The first route and the formulation of MFDT using notions of traffic and asymmetry

Following [13], and denoting the escape rate $\lambda_t^h(c') = \sum_{c \neq c'} w_t^h(c', c)$, we define the traffic by the following functional:

$$T[c] = 2 \int_0^t dt' \lambda_{t'}^{h_t}(c_{t'}). \quad (15)$$

Let us also make a particular choice of transition rates compatible with detailed balance, namely

$$w_{t'}^{h_t}(c, c') = w_{t'}(c, c') \exp\left(\beta h_{t'} \frac{O(c') - O(c)}{2}\right), \quad (16)$$

where $O(c)$ represents a physical time-independent observable. Then, the current $v_{t'}(c_{t'})$ takes the form

$$v_{t'}(c_{t'}) = \frac{1}{2} \tau_{t'}(c_{t'}) + \frac{1}{2} j_{t'}(c_{t'}), \quad (17)$$

where we have introduced

$$\tau_{t'}(c_{t'}) = \left. \frac{\delta T[c]}{\delta h_{t'}} \right|_{h \rightarrow 0}, \quad (18)$$

which is the excess of traffic introduced in [13], where excess is meant with reference with the unperturbed dynamics. In order to simplify the notation, we will omit the dependence of $A_t(c_t, h_t)$ on the protocol. With the rates given in equation (16), we have

$$\begin{aligned} R_{\text{eq}}(t, t') &= \langle j_{t'}(c_{t'}) A_t(c_t) \rangle = \beta \left\langle \frac{d}{dt'} O(c_{t'}) A_t(c_t) \right\rangle \\ &= \beta \frac{d}{dt'} \langle O(c_{t'}) A_t(c_t) \rangle. \end{aligned}$$

We have used the following definition for the derivative of an observable when it appears inside a correlation function:

$$\frac{d}{dt} O(c_t) = \sum_i \delta(t - t_i) (O_t(c_i) - O_t(c_{i-1})), \quad (19)$$

where t_i corresponds to the time of jump between the state c_{i-1} and c_i in the trajectory c_t . Note that Baiesi *et al* [13] use a different notation according to which $dO(c_t)/dt$ denotes a quantity that depends explicitly on the generator of the dynamics. We find the notation based on equation (19) simpler to use because with this choice, the derivative is independent of the generator of the dynamics and can thus pass inside the correlation function, which is not the case otherwise. In the end, the response function obtained in [13] is recovered:

$$R(t, t') = \frac{\beta}{2} \frac{d}{dt'} \langle O(c_{t'}) A_t(c_t) \rangle - \frac{1}{2} \langle \tau_{t'}(c_{t'}) A_t(c_t) \rangle. \quad (20)$$

Note that in equilibrium both terms on the rhs of equation (20) are equal and contribute to half of the response. This form of response is less general than the one of equations (7)–(14) because it relies on a particular parametrization of the

rates, namely equation (16). However, the method can be generalized to a different parametrization, as long as it is specified. The above response function can be equivalently written in terms of the so-called asymmetry [3–5], which we denote below by $\text{Asi}(t, t')$:

$$\begin{aligned} R(t, t') &= \frac{\beta}{2} \frac{d}{dt'} \langle O(c_{t'}) A_t(c_t) \rangle \\ &\quad - \frac{\beta}{2} \frac{d}{dt} \langle O(c_{t'}) A_t(c_t) \rangle + \text{Asi}(t, t'), \end{aligned} \quad (21)$$

since we have, for $t \geq t'$, that

$$\begin{aligned} \frac{d}{dt} \langle O(c_t) A_{t'}(c_{t'}) \rangle &= \sum_{c, c', c''} O(c) L_t(c'', c) \\ &\quad \times \rho(c'' t | c' t') \rho_{t'}(c') A_{t'}(c'), \\ &= \sum_{c', c''} \rho(c'' t, c' t') A_{t'}(c') \left[\sum_c (O(c) - O(c'')) L_t(c'', c) \right], \\ &= \frac{1}{\beta} \langle \tau_t(c_t) A_{t'}(c_{t'}) \rangle, \end{aligned} \quad (22)$$

where we have denoted the probability of finding the system in state c'' at time t given that it was in state c' at time t' in the unperturbed dynamics by $\rho(c'' t | c' t')$, and $L_t(c, c')$ is the generator of the unperturbed dynamics defined as in equation (1) for the perturbed dynamics. As the name suggests, the asymmetry is asymmetric in the exchange of t and t' and vanishes at equilibrium as can be checked from its expression for this case:

$$\text{Asi}(t, t') = \frac{1}{2} \langle \tau_t(c_t) A_{t'}(c_{t'}) \rangle - \frac{1}{2} \langle \tau_{t'}(c_{t'}) A_t(c_t) \rangle. \quad (23)$$

In the end, starting from the entropic form of MFDT, we have obtained the response in terms of the local currents from the decomposition of the system entropy production. Adding a hypothesis on the perturbation of the rates, we recovered the MFDT with excess of traffic and the one with asymmetry.

5. Inequalities generalizing the second law of thermodynamics

The generalized Hatano–Sasa relation of equation (5) mentioned in section 2 on the third route is also interesting in that it contains a generalization of the second law. This can be seen by substituting into this equation the particular choice $A_t(c_t, h_t) = \delta(c - c_t) p_t(c_t, h_t) / \pi_t(c_t, h_t)$. Since it is clear that in this case $\langle A(c_t, h_t) \rangle_{\pi_t} = p_t(c_t, h_t)$, one obtains

$$p_t(c, [h]) = \left\langle \frac{p_t(c_t, [h])}{\pi_t(c_t, h_t)} \delta(c - c_t) \exp(-\mathcal{Y}[c]) \right\rangle, \quad (24)$$

which can be rewritten as

$$p_t(c, [h]) = \langle \delta(c - c_t) \exp(-\mathcal{Y}[c] - \Delta s^b) \rangle \quad (25)$$

with $\Delta s^b = \ln[\pi_t(c_t, h_t) / p_t(c_t, [h])]$. This boundary term Δs^b represents the difference between the stochastic entropy production Δs_t and its reduced analogue $\Delta \psi_t = \psi_t(c_t, h_t) - \psi_0(c_0, h_0)$, and the average of this quantity with respect

to p_t represents the Kullback–Leibler divergence $D(p_t||\pi_t)$ between the two distributions p_t and π_t . Furthermore, by integrating equation (25) over c , one obtains an integral fluctuation theorem for $\mathcal{Y} + \Delta s^b$, which by Jensen’s inequality leads to

$$\langle \mathcal{Y} \rangle \geq -\langle \Delta s^b \rangle = D(p_t||\pi_t). \quad (26)$$

Note that the positivity of the Kullback–Leibler divergence implies that $\langle \mathcal{Y} \rangle \geq 0$, which also follows directly from equation (5) with the choice $A_t = 1$. In other words, the inequality of equation (26) means that the average of the work-like functional \mathcal{Y} dictates the maximum amount of lag between the two distributions p_t and π_t . This is reminiscent of a very similar relation derived for systems initially at equilibrium [33]

$$\langle W_{\text{diss}} \rangle \geq k_B T D(p_t||p_t^{\text{eq}}), \quad (27)$$

where W_{diss} is the dissipated work and p_t^{eq} represents the equilibrium distribution evaluated at the current value of the control parameter. Therefore, equation (26) should be viewed as a generalization of this result to the case when the initial distribution is time dependent.

When the final time t is sufficiently long for the relaxation of p_t toward π_t to have taken place, the present framework contains a generalization of the second law to transitions between non-stationary states corresponding to fixed values of the control parameter [24, 25]. This is analogous to the generalization of the second law of thermodynamics to transitions between NESSs, which follows from the standard Hatano–Sasa relation [12]. Note also that the inequality holds for an arbitrary protocol, and becomes an equality in the adiabatic limit of very slow driving ($\dot{h}_t \rightarrow 0$) because in this case $p_t \rightarrow \pi_t$ and $\langle \mathcal{Y} \rangle \rightarrow 0$, which follows directly from the definition of \mathcal{Y} . This property is essential for identifying this inequality as a generalization of the second law of thermodynamics to transitions between non-equilibrium states.

In [34], it was shown that remarkably the total entropy production can be split into two parts, the so-called adiabatic part (corresponding to the contribution of the entropy production which stays present in the limit of slow driving [12, 35]) and the remaining non-adiabatic part. Each part satisfies a detailed fluctuation theorem, which means that the second law can be split into these two contributions. It is natural to ask whether other decompositions of this kind are possible [36] and whether the existence of a stationary distribution is required to define the adiabatic and non-adiabatic parts of the total entropy production. With the present formalism, one can show that the special structure of the ‘three detailed fluctuation theorems’ is lost when a stationary reference is not available, although a detailed fluctuation theorem still holds separately for the adiabatic or non-adiabatic part of the dynamical action [24, 25]. The fluctuation theorem corresponding to the non-adiabatic part leads to equation (26).

6. Discussion

We have presented a general framework for systems which are prepared in a non-stationary non-equilibrium state in the absence of any perturbation and which are then further driven

by the application of a time-dependent perturbation. Typically, for applications, this perturbation is applied as a means of probing the non-equilibrium properties of the unperturbed non-equilibrium state. We can formally distinguish two different situations depending on the way the non-equilibrium state is prepared.

In the first category, the non-equilibrium state is created by some driving, and thus the perturbation that will be applied to it after some time should be viewed as a second driving. As a particular simple example of this category, one can create the initial state by a periodic driving. In these conditions, our approach predicts an MFDT near periodically driven states and a modified second law of thermodynamics for transitions between periodically driven states. Such periodically driven states are achievable in a number of experimental systems such as a vibrated granular medium, electronic circuits, manipulated colloidal systems or quantum optics for instance.

In the second category, the initial non-stationary state is a transient state produced by the choice of initial conditions. For instance, the system has been prepared by a quench of some parameter which can be the temperature or the concentration for instance, and the dynamics that follows involves relaxation or coarsening. This is typically what happens in a glassy system, where the slow relaxation following this quench leads to aging. In a recent experiment, the energy fluctuations of a Brownian particle in an aging gel have been measured and have been shown to satisfy a fluctuation theorem [37]. In this experiment, the gel in which the probe particle is embedded can be viewed as a non-equilibrium bath, which is evolving in time as a result of aging. With the same experimental setup, the deviation from the fluctuation theorem has been measured by evaluating separately the correlations and the response function [38]. In our opinion, these exciting experimental results could open the way to more systematic tests of the theoretical framework of MFDT and of the modified second law, for systems in contact with a non-equilibrium bath [25].

When implementing this program on complex realistic systems, one will encounter the difficulty already present in the standard Hatano–Sasa that the distribution $\pi_t(c, h)$ (or $p_{\text{stat}}(c, h)$ for the standard Hatano–Sasa) is difficult to determine. Indeed, this distribution can be calculated analytically only in a few simple cases, such as the case of a Langevin particle in a harmonic trap studied in the experimental references mentioned above [37, 38]. For applications to complex systems, this distribution will not be accessible analytically. However, if the system (or sub-system) of interest is of small size, the numerical determination of this distribution is possible through extensive simulations as we have shown with an example based on the Glauber–Ising model [23]. Among the various other strategies that can facilitate this numerical determination, one recent suggestion is to determine the distribution iteratively by starting from an approximate ansatz function [39]. Another potentially very promising idea is to try to determine directly the generating function of the currents through a variational approach [40].

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