

Time-symmetric fluctuations in nonequilibrium systems

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Time-Symmetric Fluctuations in Nonequilibrium Systems

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For nonequilibrium steady states, we identify observables whose fluctuations satisfy a general symmetry and for which a new reciprocity relation can be shown. Unlike the situation in recently discussed fluctuation theorems, these observables are time-reversal symmetric. That is essential for exploiting the fluctuation symmetry beyond linear response theory. In addition to time reversal, a crucial role is played by the reversal of the driving fields that further resolves the space-time action. In particular, the time-symmetric part in the space-time action determines the second order effects of the nonequilibrium driving.

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Nonequilibrium statistical mechanics is still very much under construction. Its very rich phenomenology reaching from some of the great unsolved problems of classical physics such as turbulence, to the interdisciplinary fields of biophysics and complex systems, has not yet found a sufficiently general and powerful theoretical foundation. Compared with equilibrium statistical mechanics, we lack a global fluctuation theory of far from equilibrium systems, we do not have a controlled perturbation theory, and even many conceptual debates have not found their generally accepted and logical conclusions. In contrast, the Gibbs formalism of equilibrium statistical mechanics yields general identities and inequalities connecting thermodynamic quantities and correlation functions. They relate the response of a system to an external action with the fluctuations in the system of the corresponding variable. That is partially inherited by irreversible thermodynamics, as in the fluctuation-dissipation theorem, [1], that works close to equilibrium but is mostly limited to the so-called linear response regime; away from equilibrium no such general relations exist.

In recent years, however, [2–8] a broader range of thermodynamic and fluctuation relations have become available also further away from equilibrium and there has been a general sentiment that these are important steps in the construction of nonequilibrium statistical mechanics. In what follows, we limit ourselves to the steady state. A fluctuation symmetry for a dissipation function S has been proposed and studied in various contexts. Schematically, that fluctuation relation goes somewhat like

$$\text{Prob}[S = \sigma] = \text{Prob}[S = -\sigma] \exp(\sigma), \quad (1)$$

where the probability distribution is in the steady state and S denotes in general a path-dependent variable whose average can often be interpreted as the total change (in dimensionless units) of entropy over a long time interval $[0, \tau]$. Depending on the particular realization, the function S is better called entropy current, dissipated work, or

entropy production. Subject to the scale of description $S = S_\varphi(\omega)$ is a function of the path or history ω and of the driving fields φ . We will call S the dissipation function.

Symmetries such as (1) first appeared in [2,4] in the context of thermostated and smooth dynamical systems where the phase space contraction is identified with entropy production. A slightly stronger statement is that the steady state averages satisfy

$$\langle f\theta \rangle = \langle f e^{-S} \rangle, \quad (2)$$

where $f = f(\omega)$ is a function of the history $\omega = (\omega_t)_0^\tau$ of the system over the time interval $[0, \tau]$ and θ is the time reversal, $\theta\omega = (\omega_{\tau-t})_0^\tau$, so that $f\theta(\omega) = f(\theta\omega)$.

While that symmetry (2) is very generally valid and while the symmetry-identity (1) is nonperturbative (i.e., not restricted to close to equilibrium situations), so far its further theoretical consequences have only been made visible in the derivation of linear response relations such as those of Green and Kubo, see, e.g., [9,10]. The bad news is that while relations such as (1) or (2) are very general, their consequences for expansions around equilibrium are necessarily limited to first order, the linear regime, as we show next.

Expansion of the fluctuation symmetry for the dissipation function.—As a central assumption, here as in irreversible thermodynamics [1] we take the dissipation function S as given by

$$S = S_\varphi(\omega) = \sum_\alpha \varphi_\alpha J_\alpha(\omega) = \varphi \cdot J, \quad (3)$$

where the J_α 's are the currents describing the displacement of a quantity of type α (matter, charge, energy, ...); the φ_α 's indicate the various accompanying (driving) fields or affinities ($\varphi = 0$ is set to correspond to equilibrium). This assumption implies that we ignore temporal boundary terms of the form $U(\omega_\tau) - U(\omega_0)$, as they usually play no further role in the fluctuation theory for $\tau \uparrow +\infty$; see, however, [11]. We write $\langle \cdot \rangle_\varphi$ for the steady state expecta-

tion and $\langle \cdot \rangle_0 = \langle \cdot \rangle_{\text{eq}}$ stands for the equilibrium expectation. No systematic expansion around equilibrium is at all possible from the relations (1) or (2). That can be seen as follows.

If f^+ is time symmetric, $f^+\theta = f^+$, then (2) gives to linear order in φ ,

$$\langle f^+ \rangle_\varphi = \langle f^+ e^{-S} \rangle_\varphi \Rightarrow \langle f^+ \rangle_\varphi = \langle f^+ \rangle_\varphi - \langle f^+ \varphi \cdot J \rangle_0 \quad (4)$$

and indeed, the product Jf^+ is antisymmetric under time reversal, hence vanishes under equilibrium expectation: (4) results in the identity $0 = 0$ to first order in φ and (2) gives no first order term around equilibrium for time-symmetric observables. If f^- is antisymmetric, $f^-\theta = -f^-$, then the second order term of $\langle f^- \rangle_\varphi$ requires information about the first order terms $\langle f^- J_\alpha \rangle_\varphi$ but that information is lacking because of the arguments above applied to the time-symmetric observable $f^+ = f^- J_\alpha$.

Lagrangian setup.—From the above remarks, it is clear what is missing. We need information about the generation of time-symmetric observables. That is made most visible in a Lagrangian setup. It amounts to the realization of the steady state space-time distribution as a Gibbs distribution. That can be done under a wide variety of contexts, see [2,5,6,8,9,12,13], but most easily and explicitly for stochastic dynamics.

Quite generally we can construct the probability distribution P_φ on space-time configurations ω for a nonequilibrium steady state parametrized by some vector $\varphi = (\varphi_\alpha)$, in terms of the corresponding equilibrium distribution P_0 :

$$P_\varphi(\omega) = \exp[-\mathcal{L}_\varphi(\omega)]P_0(\omega) \quad (5)$$

denoting the Lagrangian action by \mathcal{L}_φ to avoid confusion with the dissipation function S . One cannot go too lightly over the choice of the equilibrium distribution P_0 but we momentarily choose to ignore these issues.

Relation (2) teaches that the time-antisymmetric part is given by the dissipation function:

$$S_\varphi(\omega) = \mathcal{L}_\varphi(\theta\omega) - \mathcal{L}_\varphi(\omega). \quad (6)$$

Indeed, with (6) and (2), is equivalent with

$$P_\varphi(\omega) = e^{S_\varphi(\omega)}P_0(\theta\omega), \quad (7)$$

where we used that in equilibrium, $P_0\theta = P_0$; that relation implies (1). In other words, the source of time-reversal breaking is (essentially) given by the dissipation function. That has been confirmed by many physically motivated examples and models and was also derived in much greater generality [8,14]. It was also argued there how (7) can be interpreted as a unification of existing fluctuation theorems, ranging from the Jarzynski equality [3] to the Gallavotti-Cohen theorem for chaotic dynamical systems [2].

To go beyond linear order in the nonequilibrium response-functions, it appears that we must obtain extra information about the time-symmetric part in \mathcal{L}_φ . It is

important to understand here that there is no reason to think that the nonequilibrium driving would not generate an extra term in \mathcal{L}_φ which is symmetric under time reversal. Adding a nonequilibrium driving changes the time-symmetric part in the space-time action, which is the reason why the response functions cannot be generated by the expression for the dissipation function (or entropy production) alone.

Time-symmetric part.—For characterizing nonequilibrium one has to realize that time-symmetry breaking is not spontaneous and is itself very much linked with, if not caused by, breaking of spatial or even internal symmetries. A simple case is steady heat conduction made possible by the spatial arrangement of different heat baths in contact with the system. Therefore, to resolve further the nonequilibrium state, one can exploit also directly the (anti-)symmetries associated to the driving fields. It is straightforward to introduce the notion of field reversal as $\varphi_\alpha \rightarrow -\varphi_\alpha$, which could, e.g., mean to exchange the two heat reservoirs in a one-dimensional heat conduction problem or to reverse the direction of an externally given driving field. We utilize field reversal as our basic transformation on the driving fields and with (3)

$$S_{-\varphi} = -S_\varphi, \quad \text{and hence} \quad S_\varphi(\theta\omega) = S_{-\varphi}(\omega). \quad (8)$$

This is different for the time-symmetric part in the Lagrangian $A_\varphi \equiv \mathcal{L}_\varphi + \mathcal{L}_\varphi\theta$, for which in general $A_\varphi \neq \pm A_{-\varphi}$. For $Y_\varphi \equiv \mathcal{L}_{-\varphi} - \mathcal{L}_\varphi$ we have the identity $A_{-\varphi} - A_\varphi = Y_\varphi\theta + Y_\varphi$ and we may derive that $A_\varphi = A_{-\varphi}$ iff $Y_\varphi = S_\varphi$ iff $\mathcal{L}_{-\varphi} = \mathcal{L}_\varphi\theta$. We do not assume that the system is homogeneous and there is no reason in general that A_φ would be symmetric under field reversal.

The reason why the time-symmetric term A_φ is unseen in the linear response to currents is because, to linear order, field reversal can be replaced with time reversal: for small φ ,

$$\langle J \rangle_{-\varphi} \simeq -\langle J \rangle_\varphi = \langle J\theta \rangle_\varphi.$$

This is not generally true beyond order φ , which is, for example, all important for the construction of so-called rectifiers; see, for instance, [15]. Hence, while for small driving (small φ , close to equilibrium), field reversal can be implemented by time reversal, the time-symmetric A_φ is expected to be important further away from equilibrium. Consider, for example, the flow of a viscous fluid through a tube under influence of a pressure difference $\varphi = \Delta p$. For small φ one observes a laminar flow and hence, according to Poiseuille's equation the flow rate is proportional to the pressure drop and one cannot distinguish between reversal of time and reversal of the field or pressure difference. At higher pressure differences, however, or above a critical velocity, the laminar pattern breaks up and the flow becomes turbulent. Then time reversal of the flow pattern definitely differs from the typical pattern obtained via field reversal, and hence \mathcal{L}_φ must have a time-symmetric part. Ultimately, $A_\varphi \neq A_{-\varphi}$ ($\Leftrightarrow \mathcal{L}_{-\varphi} \neq \mathcal{L}_\varphi\theta$) is intimately re-

lated to nonlinear response. The *field*-antisymmetric part $A_{-\varphi} - A_{\varphi} = Y_{\varphi}\theta + Y_{\varphi}$ rather than the *time*-antisymmetric part (the dissipation function S_{φ}) is also the pivotal quantity to observe, when one attempts higher order expansions.

New fluctuation symmetry.—Remember that $Y_{\varphi} = \mathcal{L}_{-\varphi} - \mathcal{L}_{\varphi}$. Applying field reversal we have, by construction,

$$\langle f \rangle_{-\varphi} = \langle f e^{-Y_{\varphi}} \rangle_{\varphi}. \quad (9)$$

Combining it with time reversal,

$$\langle f\theta \rangle_{-\varphi} = \langle f e^{-(1/2)(Y_{\varphi}\theta + Y_{\varphi})} \rangle_{\varphi}, \quad (10)$$

where we have used that $S_{\varphi} = (Y_{\varphi} - Y_{\varphi}\theta)/2$. As a consequence, for $R_{\varphi} \equiv (Y_{\varphi} + Y_{\varphi}\theta)/2$ we have $\text{Prob}_{\varphi}[R_{\varphi} = r] = \text{Prob}_{-\varphi}[R_{\varphi} = r]e^r$. It often happens that the field reversal can be implemented as a map on the histories. In that case, there is an involution Γ on paths ω so that $\mathcal{L}_{-\varphi} = \mathcal{L}_{\varphi}\Gamma$. The transformation Γ could, for example, simply spatially mirror all the internal degrees of freedom; cf. the examples below. Then, from (10) and similar to (2),

$$\langle f\theta\Gamma \rangle_{\varphi} = \langle f e^{-(1/2)(Y_{\varphi}\theta + Y_{\varphi})} \rangle_{\varphi}. \quad (11)$$

As $R = R_{\varphi}$ satisfies $R\theta\Gamma = -R$, we then have the fluctuation symmetry (1) not only for the dissipation function $S = (Y_{\varphi} - Y_{\varphi}\theta)/2$ but we now also get it for $R \equiv (Y_{\varphi} + Y_{\varphi}\theta)/2 = (A_{-\varphi} - A_{\varphi})/2$:

$$\text{Prob}[R = r] = \text{Prob}[R = -r]e^r, \quad (12)$$

where the probability distribution is in the steady state and the fluctuation observable R denotes the variable time-symmetric and field-reversal-antisymmetric part in the Lagrangian action over a long time interval $[0, \tau]$. This fluctuation symmetry is again generally valid, nonperturbatively and away from equilibrium. This time, however, it has implications for the response of time-symmetric observables. Indeed, from (9) or (10), when $f^+ = f^+\theta$,

$$\begin{aligned} \frac{1}{2\varphi} (\langle f^+ \rangle_{\varphi} - \langle f^+ \rangle_{-\varphi}) &= \frac{1}{2\varphi} \langle f^+ (1 - e^{-R}) \rangle_{\varphi} \frac{\partial}{\partial \varphi_{\alpha}} \langle f^+ \rangle_{\varphi=0} \\ &= \frac{1}{2} \left\langle f^+ \frac{\partial}{\partial \varphi_{\alpha}} R_{\varphi} \right\rangle_{\varphi=0}. \end{aligned} \quad (13)$$

As a result, we have an Onsager reciprocity for the observables

$$V_{\alpha} \equiv \frac{\partial}{\partial \varphi_{\alpha}} R_{\varphi} |_{\varphi=0} \quad (14)$$

in the sense that to first order $\langle V_{\alpha} \rangle_{\varphi} = \langle V_{\alpha} \rangle_0 + \sum_{\gamma} M_{\alpha\gamma} \varphi_{\gamma}$ with symmetric linear response coefficients $M_{\alpha\gamma} = M_{\gamma\alpha}$; this can be seen by taking $f^+ = V_{\gamma}$ in (13). The application of (13) obviously leads to a higher order expansion of the currents J_{α} around equilibrium when taking $f^+ = J_{\alpha} J_{\gamma}$ in (13).

Examples.—We consider here a classical model of heat conduction, [16,17]. At each site $i = 1, \dots, N$ there is an oscillator characterized by a scalar position q_i and momentum p_i . The dynamics is Hamiltonian except at the boundary $\{1, N\}$ where the interaction with the reservoirs has the form of Langevin forces as expressed by the Itô stochastic differential equations

$$\begin{aligned} dq_i &= p_i dt, & i = 1, \dots, N \\ dp_i &= -\frac{\partial U}{\partial q_i}(q) dt, & i = 2, \dots, N-1 \\ dp_i &= -\frac{\partial U}{\partial q_i}(q) dt - \gamma p_i + \sqrt{\frac{2\gamma}{\beta_i}} dW_i(t), & i = 1, N. \end{aligned}$$

The β_1, β_N are the inverse temperatures of the heat baths coupled to the boundary sites $i = 1, N$; $dW_1(t), dW_N(t)$ are mutually independent standard white noise. Appropriate growth and locality conditions on the potential U (which need not be homogeneous) allow the existence of a corresponding smooth Markov diffusion process with a unique stationary distribution. When $\beta_1 = \beta_N = \beta$, the Gibbs measure $\sim \exp[-\beta H]$, $H = \sum p_i^2/2 + U$ is time reversible for the process with kinematical time reversal π given by the involution $\pi f(q, p) = f(q, -p)$. Let $\omega = ([q(t), p(t)], t \in [0, \tau])$ denote the evolution of the system in the period $[0, \tau]$. The natural definition of time reversal θ is thus $(\theta\omega)_t = \pi(\omega_{\tau-t}) = [q(\tau-t), -p(\tau-t)]$. For determining the action in (5) we take for P_0 the equilibrium process where the inverse temperatures of both heat reservoirs are equal to $\beta = (\beta_1 + \beta_N)/2$. There is one driving field $\varphi = \beta_1 - \beta_N$. To compute the antisymmetric part under time reversal, see (6), in [16] standard stochastic calculus yields $S_{\varphi} = (\beta_1 - \beta_N)J$ where $J = J(\omega)$ is the (path-dependent) heat dissipated in the reservoir at site $i = 1$ in time τ . The steady state average of S_{φ} is the heat dissipation and is easily seen to be non-negative by integrating (7) over all ω and by using the Jensen inequality; see further in [16–18]. But we can also compute the symmetric part under time reversal; it equals

$$A_{\varphi}(\omega) = \frac{\beta_1^2 - \beta^2}{\beta} \int_0^{\tau} p_1^2(t) dt + \frac{\beta_N^2 - \beta^2}{\beta} \int_0^{\tau} p_N^2(t) dt,$$

which involves the time integral of the (path-dependent) kinetic temperatures at the ends of the chain. Field reversal corresponds to exchanging β_1 with β_N and hence

$$R = (\beta_N - \beta_1) \left[\int_0^{\tau} p_1^2(t) dt - \int_0^{\tau} p_N^2(t) dt \right]$$

satisfies the fluctuation symmetry (12). Furthermore, for (14), we find

$$V = \int_0^{\tau} p_N^2(t) dt - \int_0^{\tau} p_1^2(t) dt$$

from which we get that the left-right difference of time-integrated kinetic temperatures satisfies a Green-Kubo type relation (13), but now for the time-symmetric observable

$$V = \int (p_N^2 - p_1^2) dt,$$

$$\frac{\partial}{\partial \varphi} \langle V \rangle_{\varphi=0} = \frac{1}{2} \langle V^2 \rangle_{\varphi=0}.$$

A second class of examples is given by the more general Langevin-type equation

$$dx(t) = -F(x(t))dt + \sqrt{2}dW(t).$$

The drift is given by the force $-F$, that, in nonequilibrium situations, cannot be derived from a potential. To parametrize the driving, we write $F = \nabla U - \varphi G$ where U is a potential function and G is an external force. E.g., we imagine a Brownian particle in a 1D, periodic but asymmetric sawtooth potential U , where φG is a constant external field. The asymmetry then makes field reversal nontrivial. For driving field $\varphi = 0$ the process is reversible in the stationary distribution $\sim \exp[-U]$. The path-space measure (5) is formally given, [19], by

$$P_\varphi(\omega) = \exp\left(-\frac{1}{4} \int_0^\tau dt [(\dot{x}(t) + F(x(t)))^2 \nabla F(x(t))]\right).$$

The antisymmetric part under time reversal in the action $\log P_\varphi$ indeed gives the dissipated power in terms of a stochastic Stratonovich integral of the “force” $G(x(t))$ times the “velocity” $\dot{x}(t)$: $S = \varphi \int G(x(t))\dot{x}(t)dt$. On the other hand, the part in \mathcal{L}_φ that is symmetric under time reversal and is antisymmetric under field reversal is given by

$$R = \varphi \int_0^\tau \nabla U(x(t))G(x(t))dt + \frac{\varphi}{2} \int_0^\tau \nabla G(x(t))dt. \quad (15)$$

It satisfies the symmetry (12). When there is a constant external field G , the second term vanishes and the first term picks up the total force exerted on the particle on its trajectory $x(t)$, $t \in [0, \tau]$. The same can be repeated when $x(t) = \rho(r, t)$ is a field on some bounded domain, $r \in V$, that is subject to dissipation and forcing at its “left” and “right” boundary. The expression of R is likewise of the form $\int_{\partial_\ell V} dr dt U'(\rho(r, t)) - \int_{\partial_r V} dr dt U'(\rho(r, t))$, where the spatial integration is over the respective boundaries $\partial_\ell V$, $\partial_r V$ of V ; the integrand $U'(\rho(r, t))$ could, for example, indicate the local chemical potential as defined in the system. In that sense, the external force couples with the local density of the particles. In the case of the previous example (heat conduction) one has the external force coupling with the local kinetic energy.

Conclusions.—For an irreversible thermodynamics as a linear response theory for time-antisymmetric observables like fluxes or currents, mostly only the entropy production and its fluctuations are needed. Beyond linear order, nonequilibrium physics must come to terms also with the fluctuations of time-symmetric observables. The symmetry that we know is present in the fluctuations of the dissipa-

tion function is also found to be valid for a time-symmetric observable (R). That enables us to continue the perturbation expansion around equilibrium beyond linear order in the driving fields. As a consequence, a Green-Kubo-type relation and corresponding Onsager reciprocity are obtained for time-symmetric functions (V). While some examples were shown to give specific physical meanings to our R and V , still, nonequilibrium effects beyond linear order reside, at least for theory, in a vast terra incognita and it will be mainly through experimental work that the usefulness of our new and general relations will be checked.

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