

Identities for nonlinear memory kernels

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Perturbing a system far away from equilibrium via a time-dependent protocol can formally be described by a nonlinear Volterra series expansion. Here we derive identities for the nonlinear memory kernels arising in such nonlinear expansion, including the possibility of a nonlinear coupling between perturbation and system. These identities rely on local detailed balance, and they include the fluctuation dissipation theorem as the lowest-order identity. We test them in simulations for driven over- and underdamped Brownian particles. These identities for memory kernels can be recast in a series relation for the nonequilibrium cumulants of the observable conjugate to the driving and the observable described by the Volterra series. We discuss the relation to previously found identities.

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I. INTRODUCTION

When a system is driven away from equilibrium by a perturbation protocol, its nonequilibrium evolution is generally expected to be a convolution of the protocol with linear or nonlinear memory kernels [1–5]. Such memory kernels have been found via projection operator techniques [6–9], in which a set of degrees of freedom are projected out, or in mode-coupling theory (MCT) [10,11]. Such formulations then give rise to non-Markovian stochastic descriptions, including non-Markovian Langevin equations, exemplified for the subdiffusive plateau in mean squared displacements [12] observed experimentally [13–16]. For linear systems, such as for a probe particle in a bath of harmonic oscillators, the famous Caldeira-Leggett model [8,17], explicit forms for the appearing kernels are found.

Driving nonlinear systems more strongly will bring them far away from equilibrium causing nonlinear response and nonlinear fluctuations with less well known properties. Examples of behaviors that have been found for strongly driven systems are particle oscillations [18,19], shear-thinning for macro- or microshear [20–22], or nonlinear behavior of Brownian particles immersed in micellar fluids [23–26]. Also superdiffusive behavior [22,27,28] can occur under strong driving. Nontrivial fluctuations of systems far away from equilibrium have been rationalized in terms of temperatures that are different from ambient temperature, so called effective temperatures [29–32].

It is challenging to describe systems driven far away from equilibrium theoretically. One successful route extends the above-mentioned projection operator techniques

to stronger driving [4,21,33–42], including the integration through transient technique [4]. A number of nonlinear fluctuation dissipation relations [43–47] and fluctuation theorems [46,48–51] have been found. Also approaches in field theory [19,30] have been extended to systems far away from equilibrium. Dynamical density functional theory [41,52–55], including power functional theory, provides powerful tools. Lattice models [28,56,57] allow for analytical solutions in certain cases. Nonlinear response theory [58–60] within path integral formalism [51,61–63] forms another promising route. Using such nonlinear response theory, Volterra series expansions for the mean response as well as for fluctuations have been formulated recently [64].

In this manuscript, we present a general Volterra series expansion for a system driven by a time dependent protocol, based on the methods of Refs. [59,60,64]. The Volterra series contains nonlinear memory kernels for the cumulants involving the observable conjugate to the driving protocol, and the observable the series is designed for. In contrast to previous work [64], we lift the restriction to overdamped Brownian particles, so that in this manuscript the perturbation in general enters nonlinearly in path weights. The main result in this manuscript are however identities for the nonlinear memory kernels occurring in this series. While found independently here, we discuss their similarity to other nonlinear fluctuation dissipation relations [43–47]. We demonstrate that these identities rely on local detailed balance, and the lowest order identity is the fluctuation dissipation theorem. We show how they yield a series identity for nonequilibrium cumulants. The latter is shown to agree with an expansion of a fluctuation theorem [46,50]. These identities are tested and analyzed in simulations of driven over or underdamped Brownian particles with nonlinear interactions [26,65]. We also show how they can be used to quantify the error in the fluctuation dissipation theorem for stronger driving.

II. SYSTEM AND SUMMARY OF IDENTITIES

Consider a system of N stochastic degrees of freedom $y^{(i)}$ ($i = 1, \dots, N$), abbreviated $\mathbf{y}$, in contact with a thermal bath

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at temperature T , coupled to a deterministic control parameter x_s with time index s , via the energy function $U(\mathbf{y}_s, x_s)$. The system is prepared in equilibrium at time $s = t_0$ with x taking the value x_{t_0} , i.e., for $s \leq t_0$, the distribution of $\mathbf{y}$ is the Boltzmann distribution for $U(\mathbf{y}, x_{t_0})$. x_s follows the protocol $\mathcal{X} = (x_s, t_0 \leq s \leq t)$, driving the system away from equilibrium for $s > t_0$. The derivative of U with respect to x_s defines the observable conjugate to the protocol, $F_s \equiv F(\mathbf{y}_s, x_s) \equiv \partial_{x_s} U(\mathbf{y}_s, x_s)$. In case x_s is a position, F_s is (minus) the force acting on the control parameter. x_s may however, depending on the form of U , represent other types of perturbation, e.g., external fields or forces.

In the state out of equilibrium, the cumulants of F_s and its correlations with another state observable $B_t = B(\mathbf{y}_t, x_t)$ acquire nonequilibrium forms. We allow B to explicitly depend on x_t to include the important case of $B_t \equiv F_t$. We assume that the cumulants $\langle B_t; F_{t_1}; \dots; F_{t_m} \rangle$ can be expanded in a Volterra series in $\dot{x}_s$, i.e., Ref. [64], in which the following expansion (memory) kernels appear [66]:

$$\Gamma_{t-s_1, \dots, t-s_n; t-t_1, \dots, t-t_m}^{(m,n)} := \frac{\beta^{m+1}}{n!} \frac{\delta^n}{\delta \dot{x}_{s_1} \dots \delta \dot{x}_{s_n}} \langle B_t; F_{t_1}; \dots; F_{t_m} \rangle \Big|_{\{x_s\}=x_t}. \quad (1)$$

Here, $m+1$ denotes the order of cumulant, and n marks the order of $\dot{x}$. The joint cumulant of order n is defined as [67]

$$\langle A_1; \dots; A_n \rangle := \sum_{\pi} (|\pi| - 1)! (-1)^{|\pi|-1} \prod_{B \in \pi} \left\langle \prod_{i \in B} A_i \right\rangle. \quad (2)$$

We will in this manuscript derive identities for these kernels. The first identity reads

$$\Gamma_t^{(0,1)} = \Gamma_t^{(1,0)}. \quad (3)$$

Equation (3) connects the linear response of the mean ($m=0$, $n=1$) with the zeroth order (i.e., equilibrium) of the covariance ($m=1$, $n=0$). It is the familiar fluctuation dissipation theorem [68,69]. The second reads

$$\Gamma_{s_1, s_2}^{(0,2)} = \frac{1}{2} \sum_{\pi \in S_2} \Gamma_{s_{\pi(1)}, s_{\pi(2)}}^{(1,1)} - \frac{1}{2} \Gamma_{s_1, s_2}^{(2,0)}. \quad (4)$$

Here, the sum runs over all elements of the permutation group S_2 . Equation (4) is nonlinear, as it connects the second-order response of the mean ($m=0$, $n=2$) with the first-order response of the covariance ($m=1$, $n=1$) and the zeroth order of the third cumulant ($m=2$, $n=0$). The appearance of the third cumulant is remarkable, as, this way, Eq. (4) connects nonlinear response to non-Gaussian fluctuations. The next identity reads

$$\begin{aligned} \Gamma_{s_1, s_2, s_3}^{(0,3)} &= \frac{1}{6} \sum_{\pi \in S_3} \Gamma_{s_{\pi(1)}, s_{\pi(2)}, s_{\pi(3)}}^{(1,2)} - \frac{1}{12} \sum_{\pi \in S_3} \Gamma_{s_{\pi(1)}, s_{\pi(2)}, s_{\pi(3)}}^{(2,1)} \\ &\quad + \frac{1}{6} \Gamma_{s_1, s_2, s_3}^{(3,0)}, \end{aligned} \quad (5)$$

with the sum over all elements in the permutation group S_3 . Equation (5) connects the third order response of mean ($m=0$, $n=3$), with the second-order response of the covariance ($m=1$, $n=2$), the first-order response of the third cumulant ($m=2$, $n=1$) and the zeroth order of the fourth

cumulant ($m=3$, $n=0$). While Eqs. (3)–(5) suggest a continuation to higher orders, we have only explicitly verified the three identities given above. We speculate about the form of higher orders in the conclusion section.

These identities rely on local detailed balance [70] as will be detailed below. It is worth noticing that F and B may range from micro- to macroscopic scales, as they can, e.g., be the positions of molecular particles, or macroscopic observables such as the macroscopic magnetization. The mentioned identities thus appear to be applicable on various length scales.

To our knowledge, the forms of Eqs. (4) and (5) have not been given in literature. We show in Appendix C that they can, for the respective moments, be found from the fluctuation theorem of Refs. [46,50]. While we expect that also the form in terms of cumulants can be found from that relation, the explicit calculation remains open. We note that some relations given in Refs. [43–47] are similar, but not identical to the ones given here.

III. DERIVATION OF IDENTITIES

We will lay out the derivation of the above identities in this section. Notably, as the energy function U contains the protocol x_s in arbitrary nonlinear order, this includes an explicit expansion of the conjugate observable F in $\dot{x}_s$, and the corresponding expansion of the path weight.

A. Thermodynamic considerations

Due to the time dependence of x_s , the energy function U depends explicitly on time for $s \geq t_0$. It is advantageous to split the energy function U into the unperturbed part given at time $s \leq t_0$ and the perturbation,

$$U(\mathbf{y}_s, x_s) = U(\mathbf{y}_s, x_{t_0}) + \underbrace{U(\mathbf{y}_s, x_s) - U(\mathbf{y}_s, x_{t_0})}_{H_1(\mathbf{y}_s, x_s, x_{t_0})}. \quad (6)$$

The perturbed, protocol-dependent part of the energy function is denoted by H_1 , it plays the role of the perturbation Hamiltonian; It yields the change of $U(\mathbf{y}_s, x_s)$ with respect to the reference equilibrium energy function, where the protocol is fixed at x_{t_0} .

To determine the action of the perturbed process, we consider the change ΔU between times t_0 and t . From the first law of thermodynamics [51,63],

$$\Delta U = U(\mathbf{y}_t, x_t) - U(\mathbf{y}_{t_0}, x_{t_0}) = W + Q. \quad (7)$$

Q denotes the heat, i.e., the amount of energy transferred to the system from the bath in contact with the degrees $\mathbf{y}$. W refers to the work applied,

$$W = \int_{t_0}^t ds \dot{x}_s \frac{\partial U}{\partial x_s} = \int_{t_0}^t ds \dot{x}_s F(\mathbf{y}_s, x_s). \quad (8)$$

The entropy change of the bath, $\mathcal{S} = -\beta Q$ [49] thus reads

$$\begin{aligned} \mathcal{S} &= \beta(-\Delta U + W) \\ &= \beta \left[U(\mathbf{y}_{t_0}, x_{t_0}) - U(\mathbf{y}_t, x_t) + \int_{t_0}^t ds \dot{x}_s F(\mathbf{y}_s, x_s) \right]. \end{aligned} \quad (9)$$

We will below use the assumption of local detailed balance, which relates $\mathcal{S}$ to the log ratio of path probabilities [49].

For this purpose, it is beneficial to note that S contains a part from the unperturbed energy function $U(\mathbf{y}_s, x_{t_0})$ and a part from $H_1(\mathbf{y}_s, x_s, x_{t_0})$ in Eq. (6). It is useful to isolate the latter, denoted S ,

$$\begin{aligned} S &\equiv \mathcal{S} - \mathcal{S}|_{\{x_s\}=x_{t_0}} \\ &= \beta(-\Delta H_1 + W) \\ &= \beta \left[U(\mathbf{y}_t, x_{t_0}) - U(\mathbf{y}_t, x_t) + \int_{t_0}^t ds \dot{x}_s F(\mathbf{y}_s, x_s) \right]. \end{aligned} \quad (10)$$

B. Volterra series expansion

We aim to expand the cumulants of observables $B(\mathbf{y}_t, x_t)$ and $F(\mathbf{y}_t, x_t)$ in a Volterra series in powers of the control parameter, around the equilibrium state with the control parameter being fixed at x_{t_0} [64]. We will see that this expansion takes a power series in $\dot{x}_s$, i.e., the time derivative of x_s . We consider a path integral representation, where $\omega = (\mathbf{y}_s, t_0 \leq s \leq t)$ denotes the path of the stochastic degrees of freedom $\mathbf{y}$ over the time interval $[t_0, t]$. Recall the protocol (path) $\mathcal{X} = (x_s, t_0 \leq s \leq t)$. The expectation value of an observable $O(\mathcal{X}, \omega)$ is given by

$$\langle O \rangle = \int \mathcal{D}\omega e^{-\mathcal{A}(\mathcal{X}, \omega)} \mathcal{P}_{\text{eq}}(x_{t_0}, \omega) O(\mathcal{X}, \omega). \quad (11)$$

$\mathcal{P}_{\text{eq}}(x_{t_0}, \omega)$ denotes the path weight of the corresponding equilibrium state, and $\mathcal{A}$ is the action quantifying the change due to the perturbation. O will be the corresponding products of B_t and F_t , to obtain the sought cumulants in Eq. (1).

We decompose $\mathcal{A}$ into a time-symmetric and a time-antisymmetric component [63,71]. The basic assumption employed concerns the connection of the antisymmetric part with S of Eq. (10), i.e.,

$$S = \mathcal{A}(\theta\mathcal{X}, \theta\omega) - \mathcal{A}(\mathcal{X}, \omega), \quad (12)$$

with time reversal operator θ , i.e., $(\theta\mathcal{X}, \theta\omega)_s = (\pi x_{t+t_0-s}, \pi \mathbf{y}_{t+t_0-s})$, where π denotes the kinematic time-reversal flipping the sign of velocities. The assumption formulated in Eq. (12) is known as the condition of *local detailed balance* [70,72,73]. In Appendix B we provide an example of underdamped Langevin dynamics for which the equality of the expressions in Eqs. (10) and (12) is found from the Onsager-Machlup action. We denote $D = \frac{1}{2}(\mathcal{A}(\theta\mathcal{X}, \theta\omega) + \mathcal{A}(\mathcal{X}, \omega))$, the symmetric part.

The expansion is cumbersome due to (i) the boundary terms in Eq. (10), (ii) the nonlinear dependence of S on x_s , and (iii) the dependence of F on x_s . A simplification occurs by taking the limit of $t_0 \rightarrow -\infty$ and setting the initial value of the control parameter equal to the final one, $x_{t_0} = x_t$ [59]. As x_{t_0} is the value in the infinite past, this choice has no influence on the system at time t . One has however to keep in mind that the expansion is performed around the equilibrium state corresponding to x_t , and this defines the kernels in Eq. (1). With this choice, the first two terms in Eq. (10) cancel, and S simplifies to

$$S = \beta W = \frac{1}{k_B T} \int_{-\infty}^t ds \dot{x}_s F(\mathbf{y}_s, x_s). \quad (13)$$

As mentioned, the stochastic conjugate observable F explicitly depends on x_s , rendering S nonlinear in $\dot{x}$. This is seen by expanding F_s in powers of $(x_s - x_{t_0})$ recalling that $x_{t_0} = x_t$,

$$\begin{aligned} F(\mathbf{y}_s, x_s) &= F(\mathbf{y}_s, x_t) + (x_s - x_t) F'(\mathbf{y}_s, x_t) \\ &\quad + \frac{1}{2} (x_s - x_t)^2 F''(\mathbf{y}_s, x_t) + \dots \end{aligned} \quad (14)$$

The primes denote derivatives with respect to x_s , which are evaluated at $x_s = x_t$. The resulting expansion of $S = S' + S''/2 + S'''/6 + \dots$ contains the terms

$$S' = \frac{1}{k_B T} \int_{-\infty}^t ds \dot{x}_s F(\mathbf{y}_s, x_t), \quad (15a)$$

$$S'' = -\frac{2}{k_B T} \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} F'(\mathbf{y}_s, x_t) \Theta(s' - s), \quad (15b)$$

$$\begin{aligned} S''' &= \frac{3}{k_B T} \int_{-\infty}^t ds_1 \int_{-\infty}^t ds_2 \int_{-\infty}^t ds_3 \dot{x}_{s_1} \dot{x}_{s_2} \dot{x}_{s_3} \\ &\quad \times F''(\mathbf{y}_{s_1}, x_t) \Theta(s_2 - s_1) \Theta(s_3 - s_1). \\ &\vdots \end{aligned} \quad (15c)$$

We used the identity $x_s - x_t = -\int_{-\infty}^t ds' \dot{x}_{s'} \Theta(s' - s)$ with the Heaviside function $\Theta(t)$ to make the dependence on $\dot{x}$ explicit. In the often considered case that U is linear in x_s [62,74], only S' in the above is finite, with $S'', S''', \dots$ vanishing.

The time-symmetric part D of the action in general takes no such simple form as it depends on dynamical details. This however poses no problem, as the form of D is not needed to find the derived identities. To get there, we introduce notation $D = D' + D''/2 + \dots$ with

$$D' = \int_{-\infty}^t ds \dot{x}_s \mathcal{D}_s, \quad (16)$$

$$\begin{aligned} D'' &= \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \mathcal{D}_{s,s'}, \\ &\vdots \end{aligned} \quad (17)$$

and with $\mathcal{D}_s, \mathcal{D}_{s,s'}, \dots$, as mentioned, generally unknown coefficients.

By expanding also the exponential in Eq. (11) (compare calculation steps in Ref. [64]), we find for the response of any path observable $O = O(\mathcal{X}, \omega)$,

$$\begin{aligned} \langle O \rangle &= \langle O \rangle_{\text{eq}} + \frac{1}{2} \langle S'; O \rangle_{\text{eq}} - \langle D'; O \rangle_{\text{eq}} \\ &\quad + \frac{1}{2} [\langle D'; D'; O \rangle_{\text{eq}} - \langle D'; S'; O \rangle_{\text{eq}}] - \frac{1}{2} \langle D''; O \rangle_{\text{eq}} \\ &\quad + \frac{1}{4} \langle S''; O \rangle_{\text{eq}} + \frac{1}{8} \langle S'; S'; O \rangle_{\text{eq}} + \dots \end{aligned} \quad (18)$$

$\langle \cdot \rangle_{\text{eq}}$ denote equilibrium averages with the protocol fixed at x_t ,

$$\langle O \rangle_{\text{eq}} = \int \mathcal{D}\omega \mathcal{P}_{\text{eq}}(x_{t_0}, \omega) O(\mathcal{X}, \omega). \quad (19)$$

However, the path observables in Eq. (18) depend explicitly on the nonequilibrium protocol x_s . To arrive at an explicit Volterra series expansion of the response in $\dot{x}$ the terms in Eq. (18) are additionally expanded to obtain the desired orders in $\dot{x}$.

The response of a state observable $O_t = O(x_t, \mathbf{y}_t)$, in turn, allows for simplifications. We define the expectation value of

a state observable O_t under the reversed path and protocol (compare Refs. [62,64]),

$$\langle(O\theta)_t\rangle \equiv \int \mathcal{D}\omega e^{-\mathcal{A}(\theta\mathcal{X},\theta\omega)} \mathcal{P}_{\text{eq}}(x_{t_0}, \theta\omega) O(x_t, \mathbf{y}_t), \quad (20)$$

and consider the difference of Eq. (18) and a respective expansion of Eq. (20). Because $\mathbf{y}_t$ correspond to the end of paths, which, in the reversed weights, corresponds to the distribution of equilibrium, one finds

$$\langle(O\theta)_t\rangle = \frac{1}{Z} \int d\mathbf{y} e^{-\beta U(\mathbf{y}, x_{t_0})} O(x_t, \mathbf{y}) = \langle O_t \rangle_{\text{eq}}, \quad (21)$$

with partition sum Z . In other words, $\langle(O\theta)_t\rangle$ evaluates to an equilibrium average, and we find

$$\begin{aligned} \langle O_t \rangle &= \langle O_t \rangle_{\text{eq}} + \langle S'; O_t \rangle_{\text{eq}} + \frac{1}{2} \langle S''; O_t \rangle_{\text{eq}} - \langle D'; S'; O_t \rangle_{\text{eq}} \\ &\quad - \frac{1}{2} [\langle D'; S''; O_t \rangle_{\text{eq}} + \langle D''; S'; O_t \rangle_{\text{eq}}] \\ &\quad + \frac{1}{2} \langle D'; D'; S'; O_t \rangle_{\text{eq}} + \frac{1}{24} \langle S'; S'; S'; O_t \rangle_{\text{eq}} \\ &\quad + \frac{1}{6} \langle S'''; O_t \rangle_{\text{eq}} + \dots \end{aligned} \quad (22)$$

These simplify further as $x_t = x_{t_0}$ is used. The presented form of Eqs. (18) and (22) in terms of connected correlation functions follows from the list of identities (these are modified compared to the ones given in Ref. [64] due to the higher order derivatives of S):

$$\langle S' \rangle_{\text{eq}} = \langle D' \rangle_{\text{eq}} = 0, \quad (23)$$

$$\langle S''/2 - D'S' \rangle_{\text{eq}} = 0, \quad (24)$$

$$\langle D^2 S' - D'S'' - D''S' + S'^3/12 + S'''/3 \rangle_{\text{eq}} = 0, \quad (25)$$

$$\langle D'' - D'^2 - S'^2/4 \rangle_{\text{eq}} = 0. \quad (26)$$

We will apply Eqs. (18) and (22) to calculate the cumulants of the observables $B(\mathbf{y}_t, x_t)$ and $F(\mathbf{y}_t, x_t)$. The mean is calculated using Eq. (22), while higher-order cumulants are calculated using Eq. (18). We find the following form of the Volterra series:

$$\begin{aligned} \beta \langle B_t \rangle &= \Gamma^{(0,0)} + \int_{-\infty}^t ds \dot{x}_s \Gamma_{t-s}^{(0,1)} \\ &\quad + \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \Gamma_{t-s, t-s'}^{(0,2)} \\ &\quad + \int_{-\infty}^t ds \int_{-\infty}^t ds' \int_{-\infty}^t ds'' \dot{x}_s \dot{x}_{s'} \dot{x}_{s''} \\ &\quad \times \Gamma_{t-s, t-s', t-s''}^{(0,3)} + \dots, \end{aligned} \quad (27a)$$

$$\begin{aligned} \beta^2 \langle B_t; F_{t'} \rangle &= \Gamma_{t-t'}^{(1,0)} + \int_{-\infty}^t ds \dot{x}_s \Gamma_{t-s, t-t'}^{(1,1)} + \int_{-\infty}^t ds \\ &\quad \times \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \Gamma_{t-s, t-s', t-t'}^{(1,2)} + \dots, \end{aligned} \quad (27b)$$

$$\begin{aligned} \beta^3 \langle B_t; F_{t'}; F_{t''} \rangle &= \Gamma_{t-t', t-t''}^{(2,0)} + \int_{-\infty}^t ds \dot{x}_s \Gamma_{t-s, t-t', t-t''}^{(2,1)} \\ &\quad + \dots, \end{aligned} \quad (27c)$$

$$\begin{aligned} \beta^4 \langle B_t; F_{t'}; F_{t''}; F_{t'''} \rangle &= \Gamma_{t-t', t-t'', t-t'''}^{(3,0)} + \dots \\ &\vdots \end{aligned} \quad (27d)$$

The memory kernels in these expansions are in accordance with the definition in Eq. (1). The explicit microscopic expressions in terms of S and D are given in Appendix A. The kernels $\Gamma_{t-s_1, \dots, t-s_n; t-t_1, \dots, t-t_m}^{(m,n)}$ are by construction symmetric in time arguments $s_1, \dots, s_n$ and symmetric in time arguments $t_1, \dots, t_m$. In the identities below, we furthermore symmetrize them by hand with respect to interchanges of time arguments s_n and t_m . This symmetrization is needed for the identities to hold. It poses no problem in experimental use of the identities, and the symmetrization also occurs naturally when representing the identities in integral form (see Eqs. (28), (29), and (30) below). We repeat the identities of Eqs. (3)–(5) for convenience, the derivation follows from the kernel expressions given in Appendix A:

$$\Gamma_t^{(0,1)} = \Gamma_t^{(1,0)}, \quad (3)$$

$$\Gamma_{s_1, s_2}^{(0,2)} = \frac{1}{2} \sum_{\pi \in S_2} \Gamma_{s_{\pi(1)}, s_{\pi(2)}}^{(1,1)} - \frac{1}{2} \Gamma_{s_1, s_2}^{(2,0)0}, \quad (4)$$

$$\begin{aligned} \Gamma_{s_1, s_2, s_3}^{(0,3)} &= \frac{1}{6} \sum_{\pi \in S_3} \Gamma_{s_{\pi(1)}, s_{\pi(2)}, s_{\pi(3)}}^{(1,2)} \\ &\quad + \frac{1}{6} \Gamma_{s_1, s_2, s_3}^{(3,0)} \\ &\quad - \frac{1}{12} \sum_{\pi \in S_3} \Gamma_{s_{\pi(1)}, s_{\pi(2)}, s_{\pi(3)}}^{(2,1)} \\ &\vdots \end{aligned} \quad (5)$$

We repeat that one may expect the series of identities to extend to higher orders, but, due to the cumbersome nature of derivation, we have only verified them up to the order given. We note some similarity of these expressions to the relations found in Refs. [43,44,46,47,74].

We also repeat that B and F can be macroscopic, such as, e.g., the force acting on the wall of a piston, the force acting on a colloidal particle. F and B can also be microscopic, e.g., one of the degrees $\mathbf{y}_t$.

Notably, each identity connects kernels with the same sum $m+n$.

IV. INTERPRETATION OF THE IDENTITIES

The identities of Eqs. (3)–(5) can be formulated in a relation between the cumulants of Eq. (27),

$$\begin{aligned} \beta \langle B_t \rangle &= \beta \langle B \rangle_{\text{eq}} + \beta^2 \int_{-\infty}^t ds \dot{x}_s \langle B_t; F_s \rangle \\ &\quad - \frac{\beta^3}{2} \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \langle B_t; F_s; F_{s'} \rangle \\ &\quad + \frac{\beta^4}{6} \int_{-\infty}^t ds \int_{-\infty}^t ds' \int_{-\infty}^t ds'' \dot{x}_s \dot{x}_{s'} \dot{x}_{s''} \\ &\quad \times \langle B_t; F_s; F_{s'}; F_{s''} \rangle + \mathcal{O}(\dot{x}^4). \end{aligned} \quad (28)$$

Equation (28) contains the nonequilibrium cumulants. Equation (28), as written, is valid up to order $\mathcal{O}(\dot{x}^4)$; this means that only the equilibrium part of the fourth cumulant enters, while the third cumulant enters up to linear order, and so on. We remind that Eq. (28) is derived under the choice $x_{t_0} = x_t$, so that, e.g., $\beta\langle B \rangle_{\text{eq}}$ is evaluated in the equilibrium ensemble with protocol value x_t . While we have formally expanded in powers of $\dot{x}$, the structure of Eq. (28) suggests that the small dimensionless expansion parameter can be estimated

to $\beta F \int_{t-\tau}^t ds \dot{x}_s$ with relaxation time τ of the cumulants. It should also be noted that on the level of moments, Eq. (28) agrees with an expansion of a fluctuation theorem [46,50], see Appendix C.

Equation (28) shows that the identities allow to express any cumulant purely in terms of other cumulants. For example, the first cumulant can be expanded in terms of the following expansion coefficients (memory kernels):

$$\begin{aligned} \beta\langle B_t \rangle &= \langle B \rangle_{\text{eq}} + \int_{-\infty}^t ds \dot{x}_s \Gamma_{t-s}^{(1,0)} + \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \left(\Gamma_{t-s,t-s'}^{(1,1)} - \frac{1}{2} \Gamma_{t-s,t-s'}^{(2,0)} \right) \\ &+ \int_{-\infty}^t ds \int_{-\infty}^t ds' \int_{-\infty}^t ds'' \dot{x}_s \dot{x}_{s'} \dot{x}_{s''} \left(\Gamma_{t-s,t-s',t-s''}^{(1,2)} + \frac{1}{6} \Gamma_{t-s,t-s',t-s''}^{(3,0)} - \frac{1}{2} \Gamma_{t-s;t-s',t-s''}^{(2,1)} \right) + \mathcal{O}(\dot{x}^4). \end{aligned} \quad (29)$$

In Eq. (29), the kernels of second, third, and fourth order cumulants appear on the right-hand side. Analogously, the expansion of the second cumulant is given by

$$\begin{aligned} \beta^2 \int_{-\infty}^t ds \dot{x}_s \langle B_t; F_s \rangle &= \int_{-\infty}^t ds \dot{x}_s \Gamma_{t-s}^{(0,1)} + \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \left(\Gamma_{t-s,t-s'}^{(0,2)} + \frac{1}{2} \Gamma_{t-s,t-s'}^{(2,0)} \right) \\ &+ \int_{-\infty}^t ds \int_{-\infty}^t ds' \int_{-\infty}^t ds'' \dot{x}_s \dot{x}_{s'} \dot{x}_{s''} \left(\Gamma_{t-s,t-s',t-s''}^{(0,3)} - \frac{1}{6} \Gamma_{t-s,t-s',t-s''}^{(3,0)} + \frac{1}{2} \Gamma_{t-s;t-s',t-s''}^{(2,1)} \right) + \mathcal{O}(\dot{x}^4). \end{aligned} \quad (30)$$

Equation (30) quantifies nonequilibrium fluctuations of B and F . It is expressed in terms of mean of B , and third and fourth cumulants.

Equations (29) and (30) also allow to determine the difference of the mean of B and the covariance of B and F ,

$$\begin{aligned} \beta\langle B_t \rangle - \beta^2 \int_{-\infty}^t ds \dot{x}_s \langle B_t; F_s \rangle &= \Gamma^{(0,0)} - \frac{1}{2} \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \Gamma_{t-s,t-s'}^{(2,0)} + \frac{1}{2} \int_{-\infty}^t ds \int_{-\infty}^t ds' \int_{-\infty}^t ds'' \dot{x}_s \dot{x}_{s'} \dot{x}_{s''} \\ &\times \left(\frac{1}{3} \Gamma_{t-s,t-s',t-s''}^{(3,0)} - \Gamma_{t-s;t-s',t-s''}^{(2,1)} \right) + \mathcal{O}(\dot{x}^4) \end{aligned} \quad (31)$$

$$\begin{aligned} &= \beta\langle B \rangle_{\text{eq}} - \frac{\beta^3}{2} \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \langle B_t; F_s; F_{s'} \rangle \\ &+ \frac{\beta^4}{6} \int_{-\infty}^t ds \int_{-\infty}^t ds' \int_{-\infty}^t ds'' \dot{x}_s \dot{x}_{s'} \dot{x}_{s''} \langle B_t; F_s; F_{s'}; F_{s''} \rangle + \mathcal{O}(\dot{x}^4). \end{aligned} \quad (32)$$

The left-hand side vanishes identically up to first order, as it resembles FDT. The right-hand side thus quantifies the deviation from FDT, starting at second order. The first term on the right, notably, is the equilibrium third cumulant.

Notably, the right-hand side of Eq. (31) vanishes for Gaussian observables F and B , as then the higher cumulants vanish. In such systems, the mean thus equals the integrated covariance to the given order. It will be interesting to investigate this for higher orders than the ones given here.

V. TEST: DRIVEN PROBE PARTICLE IN NONLINEAR BATH

In this section we will test the identities for $B \equiv F$ in simulations of a model of a driven probe particle in a nonlinear surrounding.

A. Model

We consider two coupled Brownian particles. Particle 1, the probe, with position $y^{(1)}$ and a second particle, making

up a nonlinear surrounding, with position $y^{(2)}$. The particles carry friction coefficients γ_1 and γ_2 , respectively, and masses m_1 and m_2 . Varying the latter allows to test overdamped and underdamped cases. The particles follow the Langevin equations

$$m_i \ddot{y}_t^{(i)} + \gamma_i \dot{y}_t^{(i)} = -\partial_{y_t^{(i)}} U(\{y_t^{(i)}\}, x_t) + \xi_t^{(i)}, \quad (33)$$

with Gaussian white noises $\xi_t^{(i)}$,

$$\langle \xi_t^{(i)} \rangle = 0 \quad \text{and} \quad \langle \xi_t^{(i)} \xi_{t'}^{(j)} \rangle = 2k_B T \gamma_i \delta_{ij} \delta(t - t'). \quad (34)$$

The potential U contains two distinct terms, it reads

$$U(\mathbf{y}, x) = U_{\text{int}}(y^{(2)} - y^{(1)}) + U_{\text{ext}}(y^{(1)} - x), \quad (35)$$

which encompasses an interaction potential U_{int} as well as a coupling of the probe to the protocol via an external potential U_{ext} . We have in mind that this model mimics the case of a Brownian particle in a laser trap and interacting with a viscoelastic surrounding.

The interaction potential is nonlinear to allow for nonlinear effects, and it is specifically a periodic potential [65]

$$U_{\text{int}}(y) = -2U_0 \cos\left(\frac{\pi y}{d_1}\right) \sin\left(\frac{\pi y}{d_2}\right), \quad (36)$$

with amplitude U_0 and length scales d_1 and d_2 . This interaction potential is nonbinding, which allows, e.g., for pronounced nonlinear effects such as shear thinning [26,65]. For $d_1 = d_2$, we have $U_{\text{int}}(y) = -U_0 \cos(\frac{2\pi y}{d_1} - \frac{\pi}{2})$, which is the symmetric potential used earlier [64,65]. For $d_1 \neq d_2$, the potential is asymmetric. Such asymmetric case will be used to explore the identities involving even order responses. Even order responses vanish for the symmetric potential case.

The external potential is chosen quadratic, mimicking, e.g., the potential of a harmonic trap,

$$U_{\text{ext}}(y) = \frac{1}{2}\kappa y^2, \quad (37)$$

with stiffness κ . The protocol x_t thus presents the center of the trap, and changing its position as a function of time presents the perturbation. The force

$$F_t = \partial_{x_t} U(\mathbf{y}_t, x_t) = -\kappa(y_t^{(1)} - x_t) \quad (38)$$

is proportional to the distance between probe and the center of the trap, which is measurable in typical experimental setups.

We use the following parameter values. $\gamma_2 = 10\gamma_1$, i.e., particle 2 has a large friction compared to particle 1, yielding pronounced viscoelastic effects. For the symmetric system $d_1 = d_2 \equiv d$ and $U_0 = 2k_B T$, the latter giving rise to large nonlinear effects. For the asymmetric system $d_2 = 3d_1 = 1.5d$ and $U_0 = k_B T$. For the overdamped case, $m_1 = m_2 = 0$, while $m_1 = m_2 = \gamma_1^2 d^2 / k_B T$ is used. The graphs are expressed in terms of γ_1 , d , and $k_B T$. Simulations are performed using the Euler-Maruyama method and involve typically hundreds of trajectories, each with $\sim 10^7$ steps and step size $\Delta t \sim 10^{-3} d^2 \gamma_1 / k_B T$, for details see Appendix D.

B. Testing identities, overdamped case

We start with the symmetric overdamped model, i.e., $d_1 = d_2$ and $m_1 = m_2 = 0$. In this case, the kernels are displayed in Fig. 1, as a function of the curvature κ of the trapping potential. We evaluate the kernels with all time arguments integrated over, i.e., we evaluate them at $z = 0$ for all arguments, with Laplace transform defined as $\hat{f}_z = \int_0^\infty dt e^{-zt} f_t$. The kernels with $m + n = 1$ are related in Eq. (3). This relation, being of linear order, resembles FDT, and it is displayed in Fig. 1 for completeness. The identity for $m + n = 2$, Eq. (4), has no content for the symmetric system, as all kernels vanish. Indeed, for reasons of symmetry, kernels with even sum $m + n$ vanish.

The kernels with $m + n = 3$ are finite and also shown in Fig. 1. In the graph, we show these kernels extracted in two ways. The first way is the “experimental” way, where we extract the force cumulants in simulations and from them the kernels (square symbols). The second way uses, as a double check, the explicit (microscopic) expressions for the kernels, which can be found for overdamped Brownian particles. These are given in Eqs. (23)–(30) of Refs. [64,75], with the time-symmetric components explicitly given in Eqs. (10) and (11) of Ref. [64]. They thus require correlations to be

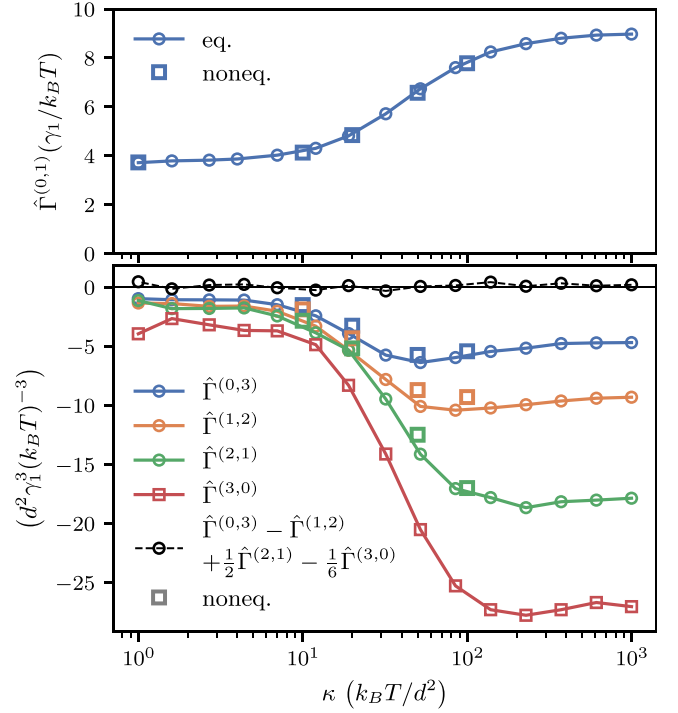


FIG. 1. Kernels for the symmetric overdamped model with all time arguments integrated over, as a function of stiffness κ of the harmonic trap. The top panel shows the first-order kernels $\hat{\Gamma}^{(0,1)} = \hat{\Gamma}^{(1,0)}$ and the bottom panel shows the third order kernels. Circles connected by lines are obtained from evaluation of the explicit forms for the kernels (Eqs. (23)–(30) in Ref. [64]), squares are extracted from (non)equilibrium force cumulants. $U_0 = 2k_B T$.

taken in equilibrium simulations, and the results are given as circles in Fig. 1. The graph displays agreement between the methods, and also shows that the kernels indeed fulfill Eq. (5).

Figure 2 shows the kernels for an asymmetric potential, i.e., for $d_1 \neq d_2$. In this case, also the kernels with $m + n = 2$ are finite, and shown in the graphs. They are also evaluated via

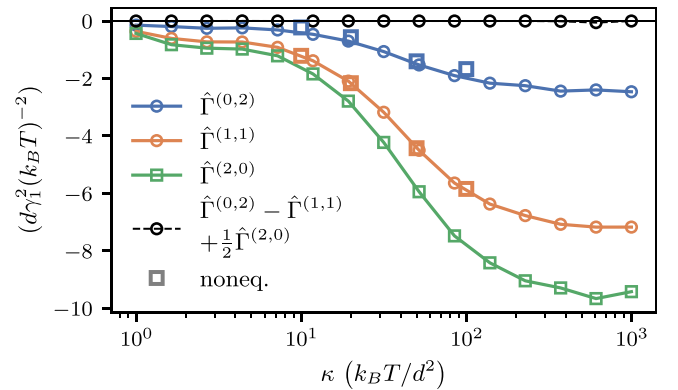


FIG. 2. Second-order kernels with all time arguments integrated over for the asymmetric overdamped model with $d_2 = 3d_1$ and $U_0 = k_B T$, for different stiffness κ . Circles with lines are obtained from direct evaluation kernel expressions (Eqs. (24), (27), and (29) in Ref. [64]), while squares are extracted from (non)equilibrium force cumulants.

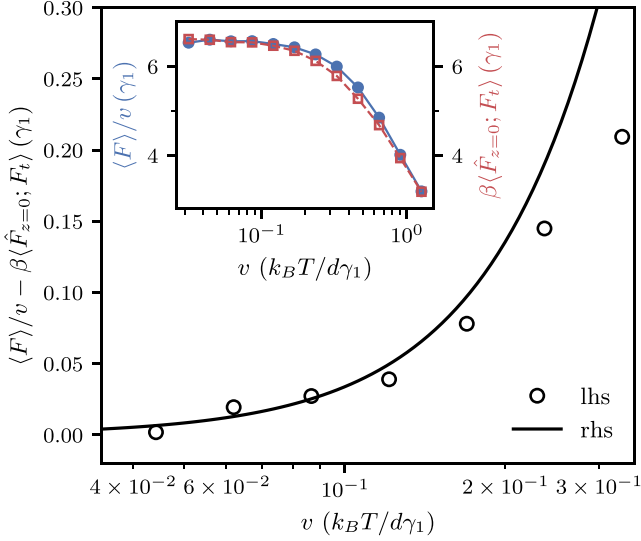


FIG. 3. Main graph: Difference of mean force and force covariance for the symmetric overdamped model (symbols), together with the prediction to second order given by Eq. (39) (line). Inset: Mean force (blue) and force covariance (red) for a large range of driving velocities v . $U_0 = 2 k_B T$ and $\kappa = 50 k_B T / d^2$.

the two mentioned ways, using the kernel expressions, and extracted from the force cumulants. We find identity Eq. (4) fulfilled.

C. Quantifying error of FDT form

In this subsection we evaluate the identities in a different manner, namely by use of Eqs. (28) and (31). For these relations, the protocol must be specified, and we use a constant velocity protocol, i.e., $\dot{x} = v$. For this case, Eq. (31) can be simplified to

$$\begin{aligned} \frac{\beta \langle F \rangle}{v} - \beta^2 \langle \hat{F}_{z=0}; F_t \rangle &= -\frac{v}{2} \hat{\Gamma}_{z=0, z=0}^{(2,0)} + \frac{v^2}{6} \hat{\Gamma}_{z=0, z=0, z=0}^{(3,0)} \\ &\quad - \frac{v^2}{2} \hat{\Gamma}_{z=0, z=0, z=0}^{(2,1)} + \mathcal{O}(v^3) \\ &= -\frac{\beta^3 v}{2} \langle \hat{F}_{z=0}; \hat{F}_{z=0}; F_t \rangle \\ &\quad + \frac{\beta^4 v^2}{6} \langle \hat{F}_{z=0}; \hat{F}_{z=0}; \hat{F}_{z=0}; F_t \rangle + \mathcal{O}(v^3). \end{aligned} \quad (39)$$

The first equality in Eq. (39) thereby is expressed in terms of the kernels, while the second equality is expressed in terms of force cumulants, providing two possibilities for determination of the right-hand side in practice.

We recall that the left-hand side of Eq. (39) vanishes in linear response, by virtue of the fluctuation dissipation theorem. The right-hand side of Eq. (31) thus quantifies the error of FDT in higher orders.

We start with the symmetric case, for which the right-hand side of Eq. (31) is of order v^2 . The inset of Fig. 3 shows the two terms on the left-hand side of Eq. (39), i.e., mean force and force covariance as a function of protocol velocity v . For small v , the two agree, as mentioned, by reason of the

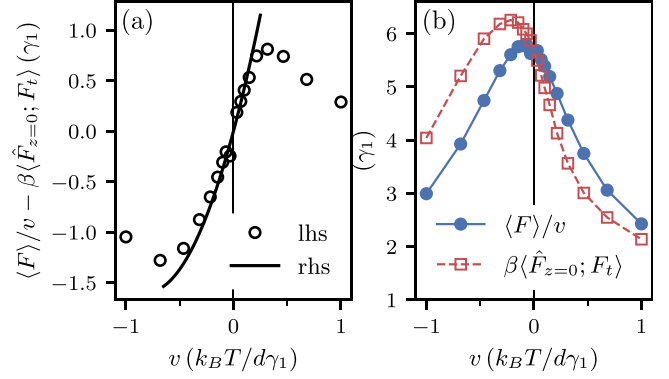


FIG. 4. (a) Difference of mean force and force covariance for the asymmetric overdamped model with $d_2 = 3 d_1$ (symbols), with the predictions to second order given by Eq. (39). (b) Mean force (blue) and force covariance (red) for positive and negative driving velocities v . $U_0 = k_B T$ and $\kappa = 100 k_B T / d^2$.

fluctuation dissipation theorem. For larger v , they deviate. The deviation is shown in the main graph, together with the right-hand side of Eq. (39). This difference is indeed of order v^2 , and it agrees with Eq. (39). Notably, the deviation between the terms on the left-hand side is quite small, for all velocities shown. This might be coincidental.

Figure 4 shows the asymmetric case, i.e., $d_1 \neq d_2$, where the right-hand side of Eq. (39) is of order v . Figure 4(b) shows the two terms on the left-hand side of Eq. (39), i.e., mean force and force covariance as a function of protocol velocity v . Here we extend to negative v , as the asymmetric model shows an asymmetric response curve. Both terms on the left-hand side of Eq. (39) thus show asymmetric behavior as a function of v , and as expected, they agree for small v , and deviate for larger v . The deviation is shown in Fig. 4(a), together with the right-hand side of Eq. (39). We show the sum of first and second-order terms of the right-hand side of Eq. (39), which yield the slope and the curvature at the origin, with good agreement with the data points.

It is interesting to note that the response $\langle F \rangle$ is almost symmetric, while the covariance, i.e., the fluctuations, are strongly asymmetric.

D. Underdamped case

As a final illustration and test, we employ the underdamped version of the asymmetric model, i.e., we use finite particle masses $m_1 = m_2 = \gamma_1^2 d^2 / k_B T$. Figure 5(a) shows the deviation of mean force and force covariance as a function of protocol velocity v . The right-hand side of Eq. (39) is shown up to second order and is extracted using the second identity in Eq. (39). It shows good agreement with the slope and curvature of the data points at the origin. Figure 5(b) shows the two terms on the left-hand side of Eq. (39) with clear asymmetry and expected agreement for small v .

Comparing Figs. 4 and 5 displays that a finite mass seems to enhance the observed effects. Both the mean response as well as the variance are larger absolutely, and the asymmetry is also more pronounced. This hints that underdamped modes are especially sensitive to asymmetry.

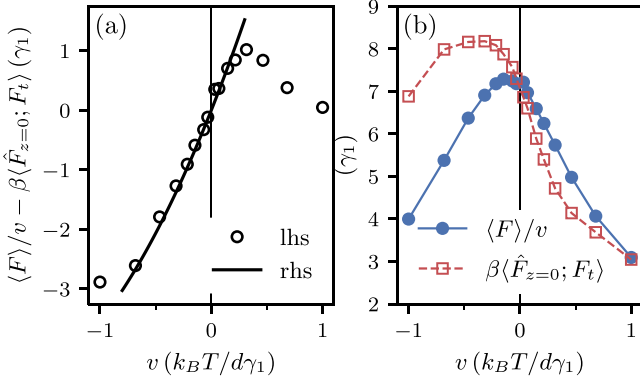


FIG. 5. (a) Difference of mean force and force covariance for the asymmetric underdamped model with $d_2 = 3d_1$ and $m_1 = m_2 = 1$, with the predictions to second order given by Eq. (39). (b) Mean force (blue) and force covariance (red) for positive and negative driving velocities v . $U_0 = k_B T$ and $\kappa = 100 k_B T / d^2$.

VI. CONCLUSION

We derived identities for nonequilibrium cumulants—specifically for their memory kernels—and tested them in various ways. These identities rely on local detailed balance, and can otherwise be applied to perturbations of the form $U(\mathbf{y}_s, x_s)$, with a protocol x_s . The identities range from micro- to macroscopic scales.

Future work can investigate these identities in higher dimensions, e.g., using multidimensional protocols. Types of protocols other than the examples investigated here are also of interest, for example time dependent particle interactions, or systems compressed in pistons.

Another task for future work concerns the derivation of identities for higher orders. While we have not checked these, the forms of Eqs. (3)–(5) show a pattern that suggests a form of identity of order n ,

$$\Gamma_{s_1, \dots, s_n}^{(0,n)} = \frac{1}{n!} \sum_{\pi \in S_n} \sum_{m=1}^n \frac{(-1)^{m+1}}{m!} \Gamma_{s_{\pi(1)}, \dots, s_{\pi(n-m)}; s_{\pi(n-m+1)}, \dots, s_{\pi(n)}}^{(m, n-m)}, \quad (40)$$

compare also Ref. [74] and the expansion of a fluctuation theorem [46,50].

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX A: MEMORY KERNELS

Recalling that all averages $\langle \cdot \rangle_{\text{eq}}$ are evaluated in the equilibrium ensemble with protocol value x_t , the memory kernels entering the mean ($m = 0$) read

$$\Gamma^{(0,0)} = \beta \langle B \rangle_{\text{eq}}, \quad (A1)$$

$$\Gamma_s^{(0,1)} = \beta^2 \langle F_s; B_0 \rangle_{\text{eq}}, \quad (A2)$$

$$\Gamma_{s_1, s_2}^{(0,2)} = \frac{\beta^2}{2} \sum_{\pi \in S_2} (\langle \mathcal{D}_{s_{\pi(1)}}; F_{s_{\pi(2)}}; B_0 \rangle_{\text{eq}} + \langle F'_{s_{\pi(1)}}; B_0 \rangle_{\text{eq}} \Theta(s_{\pi(1)} - s_{\pi(2)})), \quad (A3)$$

$$\begin{aligned} \Gamma_{s_1, s_2, s_3}^{(0,3)} = \frac{1}{6} \sum_{\pi \in S_3} & \left(\frac{\beta^2}{2} [\langle \mathcal{D}_{s_{\pi(1)}}; \mathcal{D}_{s_{\pi(2)}}; F_{s_{\pi(3)}}; B_0 \rangle_{\text{eq}} - \langle \mathcal{D}_{s_{\pi(1)}, s_{\pi(2)}}; F_{s_{\pi(3)}}; B_0 \rangle_{\text{eq}}] + \frac{\beta^4}{24} \langle F_{s_{\pi(1)}}; F_{s_{\pi(2)}}; F_{s_{\pi(3)}}; B_0 \rangle_{\text{eq}} \right. \\ & \left. + \frac{\beta^2}{2} \langle F''_{s_{\pi(1)}}; B_0 \rangle_{\text{eq}} \Theta(s_{\pi(1)} - s_{\pi(2)}) \Theta(s_{\pi(1)} - s_{\pi(3)}) + \beta^2 \langle \mathcal{D}_{s_{\pi(1)}}; F'_{s_{\pi(2)}}; B_0 \rangle_{\text{eq}} \Theta(s_{\pi(2)} - s_{\pi(3)}) \right). \quad \vdots \quad (A4) \end{aligned}$$

In these expressions, the state observables B, F, F' and so on are evaluated with the explicit dependence on x_s set to x_t . Note that $\Gamma^{(0,0)}$ does not depend on the time instance of $\mathbf{y}$ which is why we omit the time index of B . The sums in $\Gamma^{(0,2)}$ and $\Gamma^{(0,3)}$ go over all elements of the permutation group S_2 and S_3 , respectively. The first-order kernels entering the covariance ($m = 1$) read

$$\Gamma_{t_1}^{(1,0)} = \beta^2 \langle F_{t_1}; B_0 \rangle_{\text{eq}}, \quad (A5)$$

$$\Gamma_{s, t_1}^{(1,1)} = \beta^2 \left\langle \left(\mathcal{D}_s + \frac{\beta F_s}{2} \right); F_{t_1}; B_0 \right\rangle_{\text{eq}} + \beta^2 \langle F'_{t_1}; B_0 \rangle_{\text{eq}} \Theta(t_1 - s), \quad (A6)$$

$$\begin{aligned}
\Gamma_{s_1, s_2; t_1}^{(1,2)} &= \frac{\beta^2}{2} \sum_{\pi \in S_2} \left[\frac{1}{2} \langle \mathcal{D}_{s_{\pi(1)}}; \mathcal{D}_{s_{\pi(2)}}; F_{t_1}; B_0 \rangle_{\text{eq}} - \frac{1}{2} \langle \mathcal{D}_{s_{\pi(1)}, s_{\pi(2)}}; F_{t_1}; B_0 \rangle_{\text{eq}} + \frac{\beta}{2} \langle \mathcal{D}_{s_{\pi(1)}}; F_{s_{\pi(2)}}; F_{t_1}; B_0 \rangle_{\text{eq}} \right. \\
&\quad + \frac{\beta^2}{8} \langle F_{s_{\pi(1)}}; F_{s_{\pi(2)}}; F_{t_1}; B_0 \rangle_{\text{eq}} + \left(\frac{\beta}{2} \langle F_{s_{\pi(1)}}; F_{t_1}'; B_0 \rangle_{\text{eq}} + \langle \mathcal{D}_{s_{\pi(1)}}; F_{t_1}'; B_0 \rangle_{\text{eq}} \right) \Theta(t_1 - s_{\pi(2)}) \\
&\quad \left. + \frac{1}{2} \langle F_{t_1}''; B_0 \rangle_{\text{eq}} \Theta(t_1 - s_{\pi(1)}) \Theta(t_1 - s_{\pi(2)}) + \frac{\beta}{2} \langle F_{s_{\pi(1)}}'; F_{t_1}; B_0 \rangle_{\text{eq}} \Theta(s_{\pi(1)} - s_{\pi(2)}) \right]. \\
&\vdots
\end{aligned} \tag{A7}$$

The kernels entering the third cumulant ($m = 2$) are given by

$$\Gamma_{t_1, t_2}^{(2,0)} = \beta^3 \langle F_{t_1}; F_{t_2}; B_0 \rangle_{\text{eq}}, \tag{A8}$$

$$\begin{aligned}
\Gamma_{s; t_1, t_2}^{(2,1)} &= \beta^3 \left\langle \left(\mathcal{D}_s + \frac{\beta F_s}{2} \right); F_{t_1}; F_{t_2}; B_0 \right\rangle_{\text{eq}} + \beta^3 \langle F_{t_1}'; F_{t_2}; B_0 \rangle_{\text{eq}} \Theta(t_1 - s) + \beta^3 \langle F_{t_1}; F_{t_2}'; B_0 \rangle_{\text{eq}} \Theta(t_2 - s), \\
&\vdots
\end{aligned} \tag{A9}$$

and the zeroth-order kernel of the fourth cumulant ($m = 3$) reads

$$\begin{aligned}
\Gamma_{t_1, t_2, t_3}^{(3,0)} &= \beta^4 \langle F_{t_1}; F_{t_2}; F_{t_3}; B_0 \rangle_{\text{eq}}. \\
&\vdots
\end{aligned} \tag{A10}$$

APPENDIX B: EXAMPLE: ENTROPIC COMPONENT FOR UNDERDAMPED LANGEVIN DYNAMICS

We consider a setup of N underdamped Brownian degrees of freedom $\mathbf{y}$ with components $y^{(i)}$ ($i = 1, \dots, N$) which are subject to a potential $U(\mathbf{y}_t, x_t)$. x_t is an external control parameter which couples to the Brownian degrees of freedom. The setup is similar to that used in Ref. [64], but we consider underdamped dynamics and do not assume that the potential is translation invariant. The corresponding Langevin equation for the degree of freedom i is

$$m_i \ddot{y}_t^{(i)} = -\gamma_i \dot{y}_t^{(i)} - \partial_{y_t^{(i)}} U(\mathbf{y}_t, x_t) + \xi_t^{(i)}, \tag{B1}$$

with Gaussian white noise $\xi_t^{(i)}$,

$$\langle \xi_t^{(i)} \rangle = 0 \quad \text{and} \quad \langle \xi_t^{(i)} \xi_{t'}^{(j)} \rangle = 2k_B T \gamma_i \delta_{ij} \delta(t - t'). \tag{B2}$$

m_i and γ_i denote the mass and the bare friction coefficient of the degree of freedom i , respectively. The system is prepared in equilibrium at time $t = t_0$ with the protocol being fixed at x_{t_0} . Under the protocol $\mathcal{X} = (x_s, t_0 \leq s \leq t)$ the system evolves to non-equilibrium, which is characterized by a perturbation action $\mathcal{A}$, compare Eq. (11). It is given by the difference of the Onsager-Machlup action functional [76] for Eq. (B1) for x_t and $x_t = x_{t_0}$,

$$\mathcal{A} = \sum_i \frac{\gamma_i}{4k_B T} \int_{t_0}^t ds \left[\left(\dot{y}_s^{(i)} + \frac{m_i}{\gamma_i} \ddot{y}_s^{(i)} + \frac{\partial_{y_s^{(i)}} U(\mathbf{y}_s, x_s)}{\gamma_i} \right)^2 - \left(\dot{y}_s^{(i)} + \frac{m_i}{\gamma_i} \ddot{y}_s^{(i)} + \frac{\partial_{y_s^{(i)}} U(\mathbf{y}_s, x_{t_0})}{\gamma_i} \right)^2 \right]. \tag{B3}$$

Equation (B3) simplifies to

$$\mathcal{A} = \sum_i \frac{1}{4k_B T} \int_{t_0}^t ds \left\{ 2 \left(\dot{y}_s^{(i)} + \frac{m_i}{\gamma_i} \ddot{y}_s^{(i)} \right) [\partial_{y_s^{(i)}} U(\mathbf{y}_s, x_s) - \partial_{y_s^{(i)}} U(\mathbf{y}_s, x_{t_0})] + \frac{1}{\gamma_i} [(\partial_{y_s^{(i)}} U(\mathbf{y}_s, x_s))^2 - (\partial_{y_s^{(i)}} U(\mathbf{y}_s, x_{t_0}))^2] \right\}. \tag{B4}$$

Using the total derivative of the potential,

$$\frac{d}{dt} U(\mathbf{y}_t, x_t) = \sum_i \dot{y}_t^{(i)} \frac{\partial U(\mathbf{y}_t, x_t)}{\partial y_t^{(i)}} + \dot{x}_t F(\mathbf{y}_t, x_t), \tag{B5}$$

with the conjugate observable $F(\mathbf{y}_t, x_t) = \partial_{x_t} U(\mathbf{y}_t, x_t)$, we finally obtain for the action,

$$\begin{aligned}
\mathcal{A} &= \frac{1}{2k_B T} \left[U(\mathbf{y}_t, x_t) - U(\mathbf{y}_t, x_{t_0}) - \int_{t_0}^t ds \dot{x}_s F(\mathbf{y}_s, x_s) \right] + \sum_i \frac{m_i}{2\gamma_i k_B T} \int_{t_0}^t ds \ddot{y}_s^{(i)} [\partial_{y_s^{(i)}} U(\mathbf{y}_s, x_s) - \partial_{y_s^{(i)}} U(\mathbf{y}_s, x_{t_0})] \\
&\quad + \sum_i \frac{1}{4\gamma_i k_B T} \int_{t_0}^t ds [(\partial_{y_s^{(i)}} U(\mathbf{y}_s, x_s))^2 - (\partial_{y_s^{(i)}} U(\mathbf{y}_s, x_{t_0}))^2].
\end{aligned} \tag{B6}$$

The last three lines are symmetric under time reversal; they form the nonthermodynamic component D of the action. The first line of Eq. (B6) is anti-symmetric under time reversal and gives the entropic component S . Applying the definition $\mathcal{A} = D - S/2$, we find for the entropic component,

$$S = \frac{1}{k_B T} \left[U(\mathbf{y}_t, x_{t_0}) - U(\mathbf{y}_t, x_t) + \int_{t_0}^t ds \dot{x}_s F(\mathbf{y}_s, x_s) \right]. \quad (\text{B7})$$

The result is consistent with Eq. (10) derived from thermodynamic considerations.

APPENDIX C: EXPANSION OF FLUCTUATION THEOREM OF REFS. [46,50]

From Andrieux and Gaspard's work relation [50], for zero free-energy difference between initial equilibrium states of forward and backward process and in the absence of an external field, one finds

$$\langle B_t e^{-S} \rangle = \langle B_t e^{-\beta \int_{-\infty}^t ds \dot{x}_s F(\mathbf{y}_s, x_s)} \rangle = \langle B \rangle_{\text{eq}}, \quad (\text{C1})$$

compare also Ref. [46]. The notation is adapted to our system, with $B_t = B(\mathbf{y}_t, x_t)$ a state observable and S of Eq. (10) in the limit of $t_0 \rightarrow -\infty$, i.e., with $x_{t_0} = x_t$.

In this Appendix, we show that a relation between the *moments* of B_t and F_s of the form of Eq. (28) can be derived from Eq. (C1). Note that we have not been able to derive Eq. (28) in terms of *cumulants*, noting that Bochkov and Kuzovlev [45] state that the relations should be identical in form.

The expansion is performed in $\dot{x}$, around $\{x_s\} = x_t$,

$$\begin{aligned} \langle B_t e^{-S} \rangle &= \langle B_t \rangle_{\text{eq}} + \int_{-\infty}^t ds \dot{x}_s \frac{\delta}{\delta \dot{x}_s} \langle B_t e^{-S} \rangle \Big|_{\{x_s\}=x_t} + \frac{1}{2} \int_{-\infty}^t ds \\ &\quad \times \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \frac{\delta^2}{\delta \dot{x}_s \delta \dot{x}_{s'}} \langle B_t e^{-S} \rangle \Big|_{\{x_s\}=x_t} \\ &\quad + \mathcal{O}(\dot{x}^3) \\ &\stackrel{!}{=} \langle B \rangle_{\text{eq}}. \end{aligned} \quad (\text{C2})$$

We consider the expansion coefficients order by order and express the expansion of the action as

$$\mathcal{A} = \int_{-\infty}^t ds \dot{x}_s A_s + \frac{1}{2} \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} A_{s,s'} + \mathcal{O}(\dot{x}^3), \quad (\text{C3})$$

with coefficients $A_s \equiv \mathcal{D}_s - \beta F_s/2$, and so on [compare Eqs. (15)–(17)]. For the first-order coefficient of Eq. (C2) we find

$$\begin{aligned} \frac{\delta}{\delta \dot{x}_s} \langle B_t e^{-S} \rangle \Big|_{\{x_s\}=x_t} &= \frac{\delta}{\delta \dot{x}_s} \int \mathcal{D}\omega e^{-\int ds \dot{x}_s A_s + \dots} \\ &\quad \times e^{-\beta \int ds \dot{x}_s F_s} \mathcal{P}_{\text{eq}}(x_{t_0}, \omega) B_t \Big|_{\{x_s\}=x_t} \end{aligned} \quad (\text{C4})$$

$$= -\langle (A_s + \beta F_s) B_t \rangle_{\text{eq}} \quad (\text{C5})$$

$$= \frac{\delta}{\delta \dot{x}_s} \langle B_t \rangle \Big|_{\{x_s\}=x_t} - \beta \langle F_s B_t \rangle_{\text{eq}}. \quad (\text{C6})$$

Note that here we do not expand $F_s = F(\mathbf{y}_s, x_s)$ in $\dot{x}$, but leave the expansion implicit. The last identity, Eq. (C6), follows

TABLE I. Simulation parameters used to generate the data shown in Fig. 3 for the symmetric overdamped model with $U_0 = 2 k_B T$ and $\kappa = 50 k_B T/d^2$.

	No. of traj.	Steps	$t_{\text{eq}} (d^2 \gamma_1 / k_B T)$	$\Delta t (d^2 \gamma_1 / k_B T)$
$\langle F \rangle / v$	1000	2×10^7	5	$10^{-3.2}$
$\beta \langle \hat{F}_{z=0}; F_t \rangle$	200	2×10^7	5	$10^{-3.2}$
$\hat{\Gamma}^{(2,1)}, \hat{\Gamma}^{(3,0)}$	500	8×10^7	5	$10^{-3.2}$

from an analogous expansion of $\langle B_t \rangle$. For the second-order coefficient we find

$$\begin{aligned} &\frac{\delta^2}{\delta \dot{x}_s \delta \dot{x}_{s'}} \langle B_t e^{-S} \rangle \Big|_{\{x_s\}=x_t} \\ &= \langle (A_s A_{s'} - A_{s,s'}) B_t \rangle_{\text{eq}} \\ &\quad + \beta \langle (F_s F_{s'} + F_s A_{s'} + A_s F_{s'}) B_t \rangle_{\text{eq}} \end{aligned} \quad (\text{C7})$$

$$\begin{aligned} &= \frac{\delta^2}{\delta \dot{x}_s \delta \dot{x}_{s'}} \langle B_t \rangle \Big|_{\{x_s\}=x_t} + \beta \langle F_s F_{s'} B_t \rangle_{\text{eq}} \\ &\quad - \sum_{\pi \in S_2} \frac{\delta}{\delta \dot{x}_{s_{\pi(1)}}} \langle F_{s_{\pi(2)}} B_t \rangle \Big|_{\{x_s\}=x_t}. \end{aligned} \quad (\text{C8})$$

Inserting Eqs. (C6) and (C8) back into Eq. (C2), we arrive at

$$\begin{aligned} \langle B \rangle_{\text{eq}} &= \langle B_t \rangle - \beta \int_{-\infty}^t ds \dot{x}_s \langle B_t F_s \rangle \\ &\quad + \frac{\beta^2}{2} \int_{-\infty}^t ds \int_{-\infty}^t ds' \dot{x}_s \dot{x}_{s'} \langle B_t F_s F_{s'} \rangle + \mathcal{O}(\dot{x}^3), \end{aligned} \quad (\text{C9})$$

a relation between the moments of B_t and F_s of the form of Eq. (28).

APPENDIX D: SIMULATION PARAMETERS

All simulations are performed using the Euler-Maruyama method. Simulation parameters differ, depending on stiffness κ , lengths d_2 , masses m_1 and m_2 , as well as measured quantities, as detailed below.

For the data shown in Fig. 1, i.e., the symmetric overdamped model with $U_0 = 2 k_B T$ (Fig. 1) we chose an equilibration time $t_{\text{eq}} = \{100, \dots, 1\} d^2 \gamma_1 / k_B T$, $\Delta t = \{10^{-2.5}, \dots, 10^{-3.7}\} d^2 \gamma_1 / k_B T$ for $\kappa = \{1, \dots, 100\} k_B T / d^2$. Equilibrium simulations are performed with ~ 100 trajectories, each with 10^8 steps; nonequilibrium simulations are performed with ~ 100 – 1000 trajectories, each with 2×10^7 steps.

For the data shown in Fig. 2, i.e., the overdamped model with $d_2 = 3 d_1$ and $U_0 = k_B T$ we chose $t_{\text{eq}} = \{200, \dots, 2\} d_1^2 \gamma_1 / k_B T$, $\Delta t = \{10^{-2.5}, \dots, 10^{-4}\} d^2 \gamma_1 / k_B T$ for $\kappa = \{1, \dots, 100\} k_B T / d^2$ and simulated ~ 500 trajectories, each with 2×10^7 steps.

Similarly, the simulation parameters used to generate the data in Figs. 3–5 are listed in Tables I–III below.

TABLE II. Simulation parameters used to generate the data shown in Fig. 4 for the asymmetric overdamped model with $d_2 = 3 d_1$, $U_0 = k_B T$ and $\kappa = 100 k_B T / d^2$.

	No. of traj.	Steps	$t_{\text{eq}} (d^2 \gamma_1 / k_B T)$	$\Delta t (d^2 \gamma_1 / k_B T)$
$\langle F \rangle / v$	600	2×10^7	10	$10^{-3.1}$
$\beta \langle \hat{F}_{z=0}; F_t \rangle$	600	2×10^7	10	$10^{-3.1}$
$\hat{\Gamma}^{(2,0)}$	500	2×10^7	10	$10^{-3.1}$
$\hat{\Gamma}^{(2,1)}, \hat{\Gamma}^{(3,0)}$	500	4×10^7	10	$10^{-3.1}$

TABLE III. Simulation parameters used to generate the data shown in Fig. 5 for the asymmetric underdamped model with $d_2 = 3 d_1$, $m_1 = m_2 = 1$, $U_0 = k_B T$ and $\kappa = 100 k_B T / d^2$.

	No. of traj.	Steps	$t_{\text{eq}} (d^2 \gamma_1 / k_B T)$	$\Delta t (d^2 \gamma_1 / k_B T)$
$\langle F \rangle / v$	100	2×10^7	10	$10^{-3.5}$
$\beta \langle \hat{F}_{z=0}; F_t \rangle$	100	2×10^7	10	$10^{-3.5}$
$\hat{\Gamma}^{(2,0)}, \hat{\Gamma}^{(3,0)}$	60×10^3	2×10^7	10	$10^{-3.1}$
$\hat{\Gamma}^{(2,1)}$	1500	2×10^7	10	$10^{-3.5}$

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