

A general fluctuation-response relation for noise variations and its application to driven hydrodynamic experiments

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Abstract

The effect of a change of noise amplitudes in overdamped diffusive systems is linked to their unperturbed behavior by means of a nonequilibrium fluctuation-response relation. This formula holds also for systems with state-independent nontrivial diffusivity matrices, as we show with an application to an experiment of two trapped and hydrodynamically coupled colloids, one of which is subject to an external random forcing that mimics an effective temperature. The nonequilibrium susceptibility of the energy to a variation of this driving is an example of our formulation, which improves an earlier version, as it does not depend on the time-discretization of the stochastic dynamics. This scheme holds for generic systems with additive noise and can be easily implemented numerically, thanks to matrix operations.

Keywords: Nonequilibrium statistical mechanics; fluctuation-response relations; hydrodynamic interactions.

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Introduction

Continuous stochastic dynamics, in which a noise term randomizes the motion of each degree of freedom, is useful for modeling many physical systems. These noises can have a nontrivial structure. For example, hydrodynamic interactions between diffusing particles, due to the concerted motion of the fluid molecules, are represented by a cross-correlation of noise terms in Langevin equations, with amplitudes proportional to the bath temperature [25].

Finding the response of the system to a variation of one of the noise amplitudes is a task that encounters some mathematical difficulty. While a variation of deterministic drifts in diffusion equations may be dealt with by established tools [23, 46, 18, 56, 54, 51, 49, 55, 40, 6, 7, 2], such as the Girsanov theorem [31] and Radon-Nikodym derivatives [36, 7], it is not straightforward to compare two dynamics driven by different noise amplitudes [3, 57], due to the missing absolute continuity between the two path measures. This might have hampered the definition of a linear response theory to temperature variations or in general to noise-amplitude variations. However, there are recent results in this direction [17, 14, 3, 15, 48, 27, 26, 4]. The interest in understanding thermal response in nonequilibrium conditions is related to the definition of a steady state thermodynamics [47, 35, 52, 50, 9, 42, 38, 44], in which concepts as specific heat [14] are extended to the realm of dissipative steady states.

To circumvent issues related to the missing Girsanov theorem for stochastic processes with different noise coefficients, some attempts to define a thermal linear response theory needed to resort to a time discretization [3, 57]. Recently, with two independent proofs [27, 26], it was proposed a solution avoiding this discretization. Namely, a (continuous time) thermal response formula devoid of singular terms can be obtained either by an explicit regularization procedure based on functional methods [26] or through a coordinate rescaling which turns noise perturbations into mechanical ones [27]. This formalism was developed for uncorrelated noises and applied to an experiment of a thermally unbalanced RC circuit [19, 20], determining its nonequilibrium heat capacitance [4]. However, for example, the scheme described in [27, 26] cannot be applied to hydrodynamically coupled particles because their noises are correlated.

A recent experiment realized a minimal hydrodynamic system composed of nearby optically trapped colloids, in which one colloid is shaken by randomly displacing the optical trap position. It has been shown that this random displacement plays the role of an effective temperature for the shaken particle, whose motion in some sense is “hotter” than that of the other particle [12, 11]. The result is equivalent to a system in which heat bath as a whole is out of equilibrium. Not only does each degree of freedom experience a different temperature, but also the global structure of the stochastic equations does not meet the standard form of local detailed balance [37]. Thus, for such a system it is not straightforward to define concepts like entropy production in the environment. A thermal response in this context is possible, as shown with a theory including correlated white noise terms [57]. This approach, following the scheme presented in [3], still included a time discretization for overcoming the mathematical difficulties mentioned above, hence it was not yet written in terms only of sensible mathematical expressions such as (stochastic) integrals, but also included discrete sums of terms which are singular in the limit of continuous time.

In this paper we provide the most general thermal response theory for overdamped diffusive motions with additive correlated white noise, using a formulation based on path weights [58].

We thus merge the positive aspects of recent approaches in a single, general framework, which could be used to study how a diffusive process reacts to a variation of one or many of its noise amplitudes. This formalism is adopted to analyse the data of the experiment involving hydrodynamically coupled particles mentioned above, showing how to apply the scheme in practice. Pragmatically, a matrix formulation simplifies this last step. In this way, after the previous analysis of nonequilibrium RC circuits [4], we continue the application of a thermal response theory to experimental data. This complements earlier analysis of experiments focused on the mechanical linear response [33, 34, 32, 13].

Having computed the system’s susceptibility to a variation of the random driving, we show that there is a good agreement with another estimate obtained using Seifert and Speck’s formula [51]. This is in the class of formulas that focus on the density of states or related quantities [1, 29, 53, 18, 49, 51], and hence can be general enough to embrace also the thermal response type of problem. Note that an integrated version of the latter formula was recently connected with a statistical reweighting scheme that reappropriates data obtained from a stationary experiment as data obtained by an actual perturbation protocol [51]. Also in this case, one needs to know the steady state distribution.

The following Section introduces the experiment we will analyse. Dealing first with a real system helps in motivating the need for the new theory and in exposing the derivation of suitable fluctuation-response relations (Section 2). These are the response function to a variation of an element of the inverse diffusion matrix (Eq. (27)), and the susceptibility obtained by integrating in time a combination of these response functions, see Eq. (34), or Eq. (35) for its version in matrix notation. The susceptibility of the potential energy, either of the system or of the particle not driven, is derived from the steady state experimental data in Section 3, which is followed by conclusions and by further details in appendices.

1 Experiment

The two particles interaction is studied in the following experimental setup. A custom-built vertical optical tweezers with an oil-immersion objective (HCX PL. APO 63 \times /0.6-1.4) focuses a laser beam (wavelength 532 nm) to create a quadratic potential well where a silica bead (radius $R = 1 \mu\text{m} \pm 5\%$) can be trapped. The beam goes through an acousto-optic deflector (AOD) that allows to modify the position of the trap very rapidly (up to 1 MHz). By switching the trap at 10 kHz between two positions we create two independent traps, which allows us to hold two beads separated by a fixed distance (a schematic drawing is shown in Figure 1). The beads are dispersed in bidistilled water at low concentration to avoid interactions with multiple other beads. The solution of beads is contained in a disk-shaped cell (18 mm in diameter, 1 mm in depth). The beads are trapped at $h = 15 \mu\text{m}$ above the bottom surface of the cell. The position of the beads is tracked by a fast camera with a resolution of 115 nm per pixel, which after treatment gives the position with an accuracy better than 5 nm. The trajectories of the bead are sampled at 800 Hz. The stiffness of one trap ($\kappa_{11} = (3.37 \pm 0.01) \text{ pN}/\mu\text{m}$ for trap 1 or $\kappa_{22} = (3.33 \pm 0.01) \text{ pN}/\mu\text{m}$ for trap 2, each representing an element of a diagonal stiffness matrix κ , see next section) is

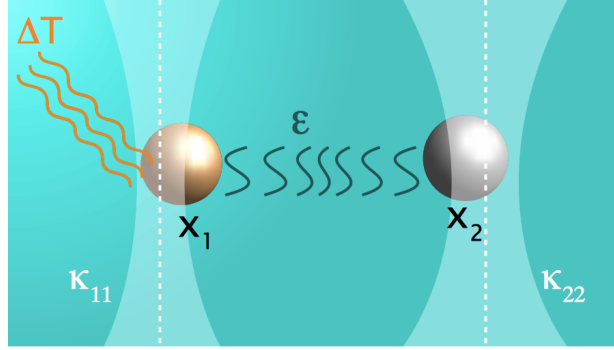


Figure 1: (Color online) Sketch of the experiment: two spherical colloidal particles in water at temperature T are confined in independent harmonic potentials (κ_{11}, κ_{22}) generated by optical traps. The particles interact hydrodynamically (ϵ), and the first particle experiences an effective temperature produced by randomly moving the position of the trap. The difference between this effective temperature and the water one (ΔT) mimics an additional thermal environment setting the system out of equilibrium.

proportional to the laser intensity¹. The two particles are trapped on a line (called “x axis”) and separated by a distance d which is tunable. The experimental results presented here have been obtained at $d = 3.9 \mu\text{m}$. At this distance the Coulombian interaction between the particle surfaces is negligible.

To create an effective temperature on one of the particles (for example on particle 1), a Gaussian white noise is sent to the AOD so that the position of the corresponding trap is moved randomly in the direction where the particles are aligned. If the amplitude of the displacement is sufficiently small to stay in the linear regime it creates a random force on the particle which does not affect the stiffness of the trap. Here the particles are over-damped and have a Lorentzian power spectrum with a typical cut-off frequency f_c of 30 Hz. The added noise is numerically created by a LABVIEW program: it is sampled at 100 kHz with a tunable amplitude A (typically of ~ 1 V) and numerically low-pass filtered at 1 kHz. It is then generated by the analog output of a NI PXIe-6366 card. The conversion factor for the displacement due to the AOD is $2.8 \mu\text{m}/\text{V}$, and the typical voltage of the noise after filtration is between ± 0.25 V. When the random force is switched on, the bead quickly reaches a stationary state with an “effective temperature” $T_{\text{eff}} = T + \Delta T$ for the randomly forced degree of freedom. Here the parameter ΔT quantifies the strength of the forcing (see Eq. (6) below) and will be used also as the tunable strength of the thermal perturbation. The properties of the effective temperature T_{eff} have been studied using power spectra [12, 10] and Fluctuations Theorems [11]. This setup allows us to create a wide range of effective temperatures for one bead, and to look at the interaction between this agitated bead and another one trapped at equilibrium at a finite distance d .

¹It can be modified by adding neutral density filters or by changing the time that the laser spend on each trap.

2 Theory

Let us consider a system with N degrees of freedom, whose state is a vector denoted by $x \equiv (x_1, x_2, \dots, x_N)$. We write the equation of motion of our overdamped system in matrix notation as

$$\dot{x} = a(x) + B\xi, \quad (1)$$

where the vector $a(x)$ is the mean drift velocity, and the $N \times N$ matrix B is the noise amplitude, which is assumed to be independent of the state x . The differential noise $\xi(t)$ is Gaussian and colorless as well as uncorrelated among different components, *i.e.*, $\langle \xi_i(t)\xi_j(s) \rangle = \delta_{ij}\delta(t-s)$. Explicit time dependences of x , ξ , and possibly a and B , are omitted for simplifying the notation, except for occasions when emphasis is appropriate. Furthermore, as has already been hinted at, we will alternate between matrix and index notations according to whichever is more intelligible in specific situations.

As an example, the drift vector of a hydrodynamic system is given in terms of the deterministic force $F(x)$ and of the mobility matrix μ ,

$$a(x) = \mu F(x). \quad (2)$$

Due to hydrodynamic interactions, the mobility matrix has nonvanishing off-diagonal entries $\mu_{ij} \neq 0$ that couple different degrees of freedom $i \neq j$. In general this would result in a state-dependent mobility, since the coupling depends on the difference of coordinates. However, we are concerned with an experiment where a state-independent mobility is a viable approximation: The hydrodynamic coupling between distant degrees of freedom is in the form of a series expansion in the ratio of particle radius to distance [25]. Hence, if the system is maintained such that the average distance is considerably larger than its fluctuations, it can be assumed to remain constant at its average value to lowest order. The optical traps in the considered experimental setup are “stiff” enough to achieve just that. The resulting mobility matrix, to the aforementioned level of approximation, for this system with $N = 2$ is given as

$$\mu = \frac{1}{\gamma} \begin{bmatrix} 1 & \varepsilon \\ \varepsilon & 1 \end{bmatrix}, \quad (3)$$

where $\varepsilon = 3R/2d$ is a small hydrodynamic coupling between spherical colloids of radius R separated by a(n average) distance d according to the Oseen approximation ($\varepsilon = 0.2766$ in the experiment we consider), and $\gamma = 16.8 \text{ pNms}/\mu\text{m}$ is the drag coefficient of the colloids in water. The distance d is set by the separation of the two harmonic optical traps, whose associated potential energy is

$$U(x) = \frac{1}{2}x^\dagger \kappa x, \quad (4)$$

when each coordinate is measured with respect to the center of its corresponding trap, where κ is a diagonal matrix of stiffness constants. We thus have a stochastic system with linear forces; see e.g. [56, 22] for some recent results on this kind of systems.

As a result of the form of the mobility matrix, the diffusivity

$$D = \frac{1}{2}BB^\dagger \quad (5)$$

is also endowed with nonvanishing off-diagonal entries that are state-independent. In fact, in the given experimental setup, the diffusivity is a sum of two contributions [57]

$$D = T\mu + \Delta T\mu \begin{bmatrix} \gamma & 0 \\ 0 & 0 \end{bmatrix} \mu = \frac{1}{\gamma} \begin{bmatrix} T + \Delta T & \varepsilon(T + \Delta T) \\ \varepsilon(T + \Delta T) & T + \varepsilon^2\Delta T \end{bmatrix}, \quad (6)$$

the first of which originates from the solvent at temperature T , and the second from the stochastic forcing on particle 1, which has a white power spectrum of magnitude $\gamma\Delta T$ [12].

2.1 Thermal perturbation and change of variables

Our aim is to investigate the response of the system to a small variation in its diffusion matrix, which is the most direct analog of temperature in this multidimensional situation. However, it is better to begin the treatment in terms of the noise amplitude, which is less physical, and revert to diffusivity at an appropriate stage.

We begin by perturbing the noise prefactor matrix as,

$$B(t) = \tilde{B} + \delta B(t), \quad (7)$$

and then follow the technique of Ref. [27], which is unfettered by terms that depend on the time discretization. To this end, we change variables by defining

$$x(t) = B(t)\tilde{B}^{-1}\tilde{x}(t), \quad (8)$$

which implies

$$\dot{x} = B\tilde{B}^{-1}\dot{\tilde{x}} + \delta\dot{B}\tilde{B}^{-1}\tilde{x}. \quad (9)$$

Plugging this into the equation of motion (1), and multiplying it by $\tilde{B}B^{-1}$ from the left, one has

$$\begin{aligned} \dot{\tilde{x}} &= -\tilde{B}B^{-1}\delta\dot{B}\tilde{B}^{-1}\tilde{x} + \tilde{B}B^{-1}a(x) + \tilde{B}\xi \\ &= -\delta\dot{B}\tilde{B}^{-1}\tilde{x} + \tilde{B}B^{-1}a(x) + \tilde{B}\xi, \end{aligned} \quad (10)$$

to first order in δB . Before proceeding with the simplification any further, one sees that the whole point of this exercise was to rid the noise process of its perturbed prefactor, and make the perturbation appear elsewhere.

Note the argument x , rather than \tilde{x} , of $a(x)$ in Eq. (10). In order to arrive at an equation of motion for the coordinate \tilde{x} , we therefore have to expand:

$$\begin{aligned} a_i(x) &= a_i(\tilde{x}) + (x_j - \tilde{x}_j) \frac{\partial a_i(\tilde{x})}{\partial \tilde{x}_j} \\ &= a_i(\tilde{x}) + \left(\delta B \tilde{B}^{-1} \tilde{x} \right)_j \frac{\partial a_i(\tilde{x})}{\partial \tilde{x}_j}, \end{aligned} \quad (11)$$

where a summation is implied over repeated indices, and the difference $x - \tilde{x}$ was given by Eq. (8). Noting also that $B^{-1} = \tilde{B}^{-1} - \tilde{B}^{-1} \delta B \tilde{B}^{-1}$ to first order, we can proceed with the simplification of Eq. (10) to find

$$\dot{\tilde{x}}_i = \left(1 - \delta B \tilde{B}^{-1}\right)_{ij} \left[a_j(\tilde{x}) + \left(\delta B \tilde{B}^{-1} \tilde{x}\right)_k \frac{\partial a_j(\tilde{x})}{\partial \tilde{x}_k} \right] - (\delta \dot{B} \tilde{B}^{-1} \tilde{x})_i + \tilde{B}_{ij} \xi_j, \quad (12)$$

or after discarding terms of order higher than 1 in δB ,

$$\dot{\tilde{x}}_i = a_i(\tilde{x}) - (\delta B \tilde{B}^{-1})_{ij} a_j(\tilde{x}) + \left(\delta B \tilde{B}^{-1} \tilde{x}\right)_k \frac{\partial a_i(\tilde{x})}{\partial \tilde{x}_k} - (\delta \dot{B} \tilde{B}^{-1} \tilde{x})_i + \tilde{B}_{ij} \xi_j. \quad (13)$$

We see that it is convenient to define

$$h(t) = \delta B(t) \tilde{B}^{-1}, \quad (14)$$

which is a dimensionless smallness parameter matrix. After the convenience of this parameter is duly exploited, it will be rewritten in terms of the diffusivity rather than the noise prefactor.

Finally, we can rearrange Eq. (13) into

$$\dot{\tilde{x}} = \tilde{a}(\tilde{x}) + \tilde{B} \xi \quad (15)$$

where

$$\tilde{a}(\tilde{x}) = a(\tilde{x}) + \alpha(\tilde{x}), \quad (16)$$

$$\alpha_i(\tilde{x}) = -h_{ik} a_k(\tilde{x}) + h_{kl} \tilde{x}_l \frac{\partial a_i(\tilde{x})}{\partial \tilde{x}_k} - \dot{h}_{ik} \tilde{x}_k, \quad (17)$$

with $\dot{h} = \delta \dot{B} \tilde{B}^{-1}$ according to Eq. (14). Thus, the perturbation of the noise prefactor in the evolution of x has been re-expressed as a perturbation of the mechanical force in the evolution of \tilde{x} . Ref. [26] discusses another perspective from which this deformation of a thermal perturbation into a deterministic perturbation can be viewed. Note that the ‘‘pseudo-force’’ $\alpha(\tilde{x}(t); t)$ depends explicitly on time through the perturbation $h(t)$.

2.2 Path weight and response

The objective of linear response is to investigate the variation of the expectation value of an observable to linear order in a small perturbation. Taking the observable to be a state observable, $\mathcal{O}(t) = \mathcal{O}(x(t))$, this variation has the form

$$\delta \langle \mathcal{O}(t) \rangle = \langle \mathcal{O}(t) \delta \ln P[x] \rangle, \quad (18)$$

due simply to the variation of the probability weights, $P[x]$, of trajectories $x(t)$. The negative logarithm of the path weight,

$$-\ln P[x] = \mathcal{A}[x], \quad (19)$$

is commonly termed the path action. However, let us remark and remind that due to technical difficulties of varying the action with respect to noise amplitude, we will utilize the coordinate \tilde{x} which, as discussed earlier, reformulates the problem in terms of a variation of the deterministic force.

According to the equation of motion (15) for $\tilde{x}(t)$ with the Gaussian noise process $\xi(t)$, the probability weight of a trajectory $\tilde{x}(t)$ has the quadratic action

$$\tilde{\mathcal{A}}[\tilde{x}] = \int \frac{ds}{4} [\dot{\tilde{x}}_i - \tilde{a}_i(\tilde{x})] \tilde{D}_{ij}^{-1} [\dot{\tilde{x}}_j - \tilde{a}_j(\tilde{x})] + \int \frac{ds}{2} \frac{\partial}{\partial \tilde{x}_i} \tilde{a}_i(\tilde{x}) \quad (20)$$

in the Stratonovich convention [58] up to a trajectory-independent additive constant, and the symmetric matrix $2\tilde{D} = \tilde{B}\tilde{B}^\dagger$.² Also note that evaluation of the integrands at time s is implied.

The variation of Eq. (20) to first order in $\alpha(\tilde{x})$ is simply

$$\delta\tilde{\mathcal{A}}[\tilde{x}] = - \int \frac{ds}{2} \left[\tilde{D}_{ij}^{-1} (\dot{\tilde{x}}_j - a_j) - \frac{\partial}{\partial \tilde{x}_i} \right] \alpha_i(\tilde{x}(s); s). \quad (21)$$

Note that due to its definition (17), $\alpha(\tilde{x})$ is of at least first order in the perturbation parameter $h(s)$. Thus, the replacement $(\tilde{x}, \tilde{D}) \rightarrow (x, D)$ incurs errors only of orders which are already truncated in this linear response scheme. That is to say, in Eq. (18) one can use $\delta \ln P[x] = -\delta\mathcal{A}[x] = -\delta\tilde{\mathcal{A}}[x]$ via $(\tilde{x}, \tilde{D}) \rightarrow (x, D)$ in Eq. (21). Using Eq. (17) to express $\alpha(x)$ explicitly, we rewrite

$$\delta\mathcal{A}[x] = - \int \frac{ds}{2} [D_{ij}^{-1} (\dot{x}_j - a_j) - \partial_i] \left(-h_{ik}a_k + h_{kl}x_l\partial_k a_i - \dot{h}_{ik}x_k \right), \quad (22)$$

where ∂_i is shorthand for $\partial/\partial x_i$. Here, one can count powers of time derivative (dots on x or h) to determine the parity of each term under time-reversal, which is relevant for thermodynamical arguments. Labeling according to the parity ($-/+$) under time-reversal, we separate $\delta\mathcal{A}[x]$ into³

$$\delta\mathcal{A}_-[x] = - \int \frac{ds}{2} \left[D_{ij}^{-1} \dot{x}_j (h_{kl}x_l\partial_k a_i - h_{ik}a_k) + \dot{h}_{ik}x_k D_{ij}^{-1} a_j + \dot{h}_{kk} \right], \quad (22a)$$

and

$$\delta\mathcal{A}_+[x] = - \int \frac{ds}{2} D_{ij}^{-1} a_j (h_{ik}a_k - h_{kl}x_l\partial_k a_i) + \int \frac{ds}{2} (h_{kl}x_l\partial_k \partial_i a_i + D_{ij}^{-1} \dot{x}_j \dot{h}_{ik}x_k), \quad (22b)$$

with $\delta\mathcal{A}[x] = \delta\mathcal{A}_+[x] + \delta\mathcal{A}_-[x]$. Via integration by parts, $\dot{h}(s)$ can be exchanged with $h(s)$.⁴ After also manipulating the indices for the convenience of isolating a common factor, $h_{kl}(s)$, one

²For brevity of notation, we denote the elements of the inverse of a matrix as in D_{ij}^{-1} rather than the unambiguous but cumbersome $(D^{-1})_{ij}$.

³Note that $\partial_i (h_{kl}x_l\partial_k a_i - h_{ik}a_k) = h_{kl}x_l\partial_k \partial_i a_i + h_{kl}\delta_{il}\partial_k a_i - h_{ik}\partial_i a_k = h_{kl}x_l\partial_k \partial_i a_i$.

⁴For this matter, we note here that we have intentionally left out integration limits in the action: The forcing $h(s)$ is assumed to be temporally localized, and the integration domain is infinite in principle. Thus, $\int ds \dot{h}g = - \int ds h\dot{g}$ for a generic function $g(s)$, and likewise $\int ds \dot{h}h_k = 0$ in Eq. (22a).

has

$$\delta\mathcal{A}_-[x] = - \int \frac{ds}{2} [D_{ij}^{-1} \dot{x}_j x_l \partial_k a_i - D_{kj}^{-1} (\dot{x}_j a_l + \dot{x}_l a_j) - D_{kj}^{-1} \dot{x}_i x_l \partial_i a_j] h_{kl}, \quad (23a)$$

$$\delta\mathcal{A}_+[x] = - \int \frac{ds}{2} [D_{kj}^{-1} a_j a_l - D_{ij}^{-1} a_j x_l \partial_k a_i - x_l \partial_k \partial_i a_i + D_{kj}^{-1} \frac{d}{ds} (\dot{x}_j x_l)] h_{kl}, \quad (23b)$$

where the identity $\dot{a}_j = \dot{x}_i \partial_i a_j$ was used in $(d/ds)(x_l a_j) = \dot{x}_l a_j + x_l \dot{a}_j$, owing to Stratonovich calculus, concerning the very last term.

Up until now, we have not elaborated about the dimensionless parameter matrix $h(s)$, which we do in Appendix B. The gist is that, rather than expressing $h(s)$ via the somewhat nonphysical noise prefactor B via Eq. (14), one can write it in terms of the variation of the inverse diffusivity, δD^{-1} , which is a more sensible physical quantity. The manifest symmetry in the contents of the resulting expression, $h = (-1/2)D\delta D^{-1}$, facilitates certain simplifications on $\delta\mathcal{A}[x]$. In particular, now that $D_{jk}^{-1} h_{kl} = (-1/2)\delta D_{jl}^{-1}$ is a symmetric matrix, the last term of Eq. (23b) can benefit from

$$D_{kj}^{-1} h_{kl} \frac{d}{ds} (\dot{x}_j x_l) = -\frac{1}{2} \delta D_{jl}^{-1} \frac{d}{ds} (\dot{x}_j x_l) = -\frac{1}{4} \delta D_{jl}^{-1} \frac{d^2}{ds^2} (x_j x_l). \quad (24)$$

Similarly, the two middle terms of Eq. (23a) combine. With these simplifications we finally have

$$\delta\mathcal{A}_-[x] = \int \frac{ds}{4} \frac{\dot{x}_i}{4} [(D_{ij}^{-1} D_{km} - \delta_{ik} \delta_{jm}) x_l \partial_k a_j - 2\delta_{im} a_l] \delta D_{ml}^{-1}, \quad (25a)$$

$$\delta\mathcal{A}_+[x] = \int \frac{ds}{4} [a_m a_l - D_{km} x_l (D_{ij}^{-1} a_j \partial_k a_i + \partial_k \partial_i a_i) + \frac{1}{2} \frac{d^2}{ds^2} (x_m x_l)] \delta D_{ml}^{-1}. \quad (25b)$$

At this stage, it would be possible to define a tensorial response function for a state observable $\mathcal{O}(t) = \mathcal{O}(x(t))$ to an impulsive variation in the inverse diffusivity matrix as

$$\mathcal{R}_{lm}(t, s) = - \left\langle \mathcal{O}(x(t)) \frac{\delta\mathcal{A}[x]}{\delta D_{lm}^{-1}(s)} \right\rangle, \quad (26)$$

which satisfies $\delta\langle\mathcal{O}(t)\rangle = -\langle\mathcal{O}(t)\delta\mathcal{A}[x]\rangle = \int ds \mathcal{R}_{lm}(t, s) \delta D_{lm}^{-1}(s)$, yielding

$$\mathcal{R}_{lm}(t, s) \equiv \mathcal{R}_{lm}^-(t, s) + \mathcal{R}_{lm}^+(t, s) \quad (27)$$

with

$$\mathcal{R}_{lm}^-(t, s) = - \left\langle \frac{\mathcal{O}(t)}{4} [(D_{ij}^{-1} D_{km} - \delta_{ik} \delta_{jm}) \dot{x}_i x_l \partial_k a_j - 2\dot{x}_m a_l](s) \right\rangle, \quad (27a)$$

$$\mathcal{R}_{lm}^+(t, s) = - \left\langle \frac{\mathcal{O}(t)}{4} [a_m a_l - D_{km} x_l (D_{ij}^{-1} a_j \partial_k a_i + \partial_k \partial_i a_i) + \frac{1}{2} \frac{d^2}{ds^2} (x_m x_l)](s) \right\rangle. \quad (27b)$$

These expressions are the generalization of those of Refs. [27, 26] to non-negligible state-independent hydrodynamic coupling. Note the ‘‘collective’’ argument (s) that applies to all the time dependence inside the square brackets preceding it.

2.3 Stationary susceptibility

The integrated response is called susceptibility. In practice, one is usually able to manipulate a single scalar parameter, call it $\lambda(s)$. It suits to write the variation $\delta D_{lm}^{-1}(s)$ in terms of the variation $\delta\lambda(s)$ of the parameter (to first order): $\delta D_{lm}^{-1}(s) = \delta\lambda(s)(\partial D_{lm}^{-1}/\partial\lambda)$. Then, an empirically accessible susceptibility to a stepwise change in λ at $t = 0$ can be expressed as

$$\chi(t) = \left. \frac{\partial \langle \mathcal{O}(t) \rangle^\lambda}{\partial \lambda} \right|_{\lambda=\lambda(0)} = \int_0^t ds H_{lm} \mathcal{R}_{lm}(t, s), \quad (28)$$

with

$$H_{lm} \equiv \left. \frac{\partial D_{lm}^{-1}}{\partial \lambda} \right|_{\lambda=\lambda(0)}. \quad (29)$$

The time integral of Eq. (28) removes one of the derivatives d/ds in the last term of Eq. (27b) and gives the boundary term,

$$- \left\langle \mathcal{O}(t) \frac{1}{8} \frac{d}{ds} [x_m(s)x_l(s)] \Big|_0^t \right\rangle. \quad (30)$$

The remaining derivative can also be removed if we specialize to the case where the initial distribution $\rho(x(0))$ in the unperturbed averages $\langle \cdot \rangle$ is stationary, since, due to invariance under time translation, the response function becomes $\mathcal{R}(t, s) = \mathcal{R}(t - s)$, and in particular

$$- \left\langle \mathcal{O}(t) \frac{1}{8} \frac{d}{ds} [x_m(s)x_l(s)] \Big|_0^t \right\rangle = \left\langle \frac{1}{8} \frac{d\mathcal{O}(t)}{dt} [x_m(s)x_l(s)] \Big|_0^t \right\rangle. \quad (31)$$

The time derivative at the later instant t can then be replaced by the backward generator of the dynamics, *i.e.*,

$$\left\langle \frac{1}{8} \frac{d\mathcal{O}(t)}{dt} [x_m(s)x_l(s)] \Big|_0^t \right\rangle = \left\langle \frac{1}{8} \mathbb{L} \mathcal{O}(t) [x_m(s)x_l(s)] \Big|_0^t \right\rangle, \quad (32)$$

where

$$\mathbb{L} = a_i \partial_i + D_{ij} \partial_i \partial_j. \quad (33)$$

This replacement is desirable, because the original expression on the left-hand side of Eq. (32) with the time derivative is much noisier, and thus requires much more statistics to converge. Since our application will be in the stationary state, we take advantage of this and continue from here on with the assumption of stationarity.

As previously done [26], we split the susceptibility into four terms

$$\chi(t) = \chi_1^-(t) + \chi_2^-(t) + \chi_1^+(t) + \chi_2^+(t), \quad (34)$$

where

$$\chi_1^-(t) = \frac{1}{2} \int_0^t ds H_{ml} \langle \mathcal{O}(t) [\dot{x}_m a_l](s) \rangle , \quad (34a)$$

$$\chi_2^-(t) = \frac{1}{4} \int_0^t ds H_{ml} \langle \mathcal{O}(t) [(\delta_{ik} \delta_{jm} - D_{ij}^{-1} D_{km}) \dot{x}_i x_l \partial_k a_j](s) \rangle , \quad (34b)$$

$$\chi_1^+(t) = \frac{1}{4} \int_0^t ds H_{ml} \langle \mathcal{O}(t) [x_l D_{km} D_{ij}^{-1} a_j \partial_k a_i - a_m a_l](s) \rangle , \quad (34c)$$

$$\chi_2^+(t) = \frac{1}{8} H_{ml} \left\langle \mathbb{L} \mathcal{O}(t) x_m(s) x_l(s) \Big|_0^t \right\rangle . \quad (34d)$$

are written in terms of Stratonovich stochastic integrals. If the heat bath is in equilibrium, the term $\chi_1^-(t)$ corresponds to the correlation between the observable and half of the entropy production in the bath. This interpretation, previously discussed for the linear response to mechanical forces [6, 7] and temperature variations [3, 27, 26], is not possible in our experiment and we remain with the fact that $\chi_1^-(t)$ as well as $\chi_2^-(t)$ correlate the observable with time-antisymmetric, *i.e.*, entropy-production-like quantities. The correlations in $\chi_1^+(t)$ and $\chi_2^+(t)$ instead involve time-symmetric, non-dissipative variables. They were named “frenetic” terms because often those time-symmetric observables quantify how agitated, or frenzied, is the exploration of the system state space [6, 7, 5].

2.4 Susceptibility in matrix notation

Despite the many indices appearing in Eq. (34), note that the highest rank tensor involved above is still of rank two. Therefore, the index contractions can be broken up into ordinary matrix operations. This should be useful, for example, for a numerical implementation in a software where matrix multiplication is defined.

We denote by ∇a^\dagger the matrix with elements $\partial_i a_j$ (where i is the row index). Thus, we deal with vectors (the positions x , velocities \dot{x} , and drifts a) and matrices. The latter are the noise variation H (defined in Eq. (29)), the diffusivity matrix D and its inverse D^{-1} , and ∇a^\dagger . By simply following the sequence of index repetitions, it is straightforward to express the terms in the susceptibility as

$$\chi_1^-(t) = \frac{1}{2} \int_0^t ds \langle \mathcal{O}(t) [\dot{x}^\dagger H a](s) \rangle , \quad (35a)$$

$$\chi_2^-(t) = \frac{1}{4} \int_0^t ds \langle \mathcal{O}(t) [\dot{x}^\dagger (\nabla a^\dagger) H x - x^\dagger H D (\nabla a^\dagger) D^{-1} \dot{x}](s) \rangle , \quad (35b)$$

$$\chi_1^+(t) = \frac{1}{4} \int_0^t ds \langle \mathcal{O}(t) [x^\dagger H D (\nabla a^\dagger) D^{-1} a - a^\dagger H a](s) \rangle , \quad (35c)$$

$$\chi_2^+(t) = \frac{1}{8} \left\langle \mathbb{L} \mathcal{O}(t) [x^\dagger H x](s) \Big|_{s=0}^{s=t} \right\rangle . \quad (35d)$$

If the stationary conditions are not satisfied, the last term should be evaluated as

$$\chi_2^+(t) = -\frac{1}{8} \frac{d}{ds} \left\langle \mathcal{O}(t) [x^\dagger H x](s) \Big|_{s=0}^{s=t} \right\rangle . \quad (35e)$$

3 Susceptibility of the colloidal system

We calculate the susceptibility of the colloidal system to a change in the effective temperature of the stochastic driving. We thus identify $\lambda = \Delta T$ and with Eq. (6) we find

$$H = \frac{\partial(D^{-1})}{\partial\Delta T} = \begin{bmatrix} \frac{-\gamma}{(T+\Delta T)^2} & 0 \\ 0 & 0 \end{bmatrix}. \quad (36)$$

Using this expression in Eq. (28) via the response functions (27a)–(27b) along with the simplifications sketched above, we find the susceptibility. Note that the term of the time-symmetric part (27b) proportional to $\partial_k \partial_i a_i$, while in general nonzero, disappears for the particular system in question, since two derivatives of the mean drift $a = \mu F = -\mu\kappa x$ vanish due to the quadratic potential (4).

Experimental data were sampled at constant rate, with a time step Δt . This means that a trajectory is a discrete collection of points $x(0), x(1), x(2), \dots, x(n)$ with $n = t/\Delta t$. For each temperature we have a long time series composed of $n_s = 10^6$ samples, out of which we extract $n_s - n + 1$ trajectories by shifting the initial point. Thus, these trajectories are partially overlapped with other ones. Since the autocorrelation time of the system is about $\gamma/\kappa_{11} \approx \gamma/\kappa_{22} \approx 5\text{ms} = 4\Delta t$, we have taken the correlation of data into account by amplifying the error bars in our results (standard deviations) by a factor of $\sqrt{4} = 2$.

Velocities are defined as $\dot{x}(i) = [x(i+1) - x(i)]/\Delta t$. The integrals in Eq. (34) or Eq. (35) follow the Stratonovich convention. In their time-discretized version, quantities (other than the \dot{x} mentioned above) in the integrals are averaged over time steps. For example, the average $[x(i+1) + x(i)]/2$ represents the positions at time step i .

To visualize the linear response of some observables, we start by considering the total energy $U = \frac{\kappa_{11}}{2}x_1^2 + \frac{\kappa_{22}}{2}x_2^2$. The susceptibility of the energy can be considered a generalized heat capacity. In Figure 2 this susceptibility χ is plotted as a function of time for four cases: $\Delta T = 0$ K, which is the equilibrium condition with no forcing on particle 1, and $\Delta T = 120$ K, 340.8 K, and $\Delta T = 967.8$ K, which become progressively farther from equilibrium. In equilibrium one can notice that twice the entropic term χ_1^- (which by definition is half of the Kubo formula for equilibrium systems), would be sufficient to obtain the susceptibility. This picture breaks down out of equilibrium, where it is the whole sum of the four components that yields the correct form of the susceptibility. For comparison, we show also the estimate $\chi^\rho(t)$ obtained with the approach by Seifert and Speck [51], through

$$\chi^\rho(t) = \left\langle \mathcal{O}(t) \left[\frac{\partial \ln \rho(x(t))}{\partial \Delta T} - \frac{\partial \ln \rho(x(0))}{\partial \Delta T} \right] \right\rangle, \quad (37)$$

which is an integrated version of their response formula. Appendix A sketches how the stationary distribution $\rho(x)$ for this system was computed. Figure 2 shows that the susceptibilities evaluated with the two approaches are compatible with each other.

As a second example, we show the susceptibility to a change of ΔT of the energy $U_2 = \frac{\kappa_{22}}{2}x_2^2$ of the second particle, namely the colloid which is not experiencing the random forcing directly. Yet, due to the hydrodynamic interaction, its energy on average increases with ΔT , as manifested

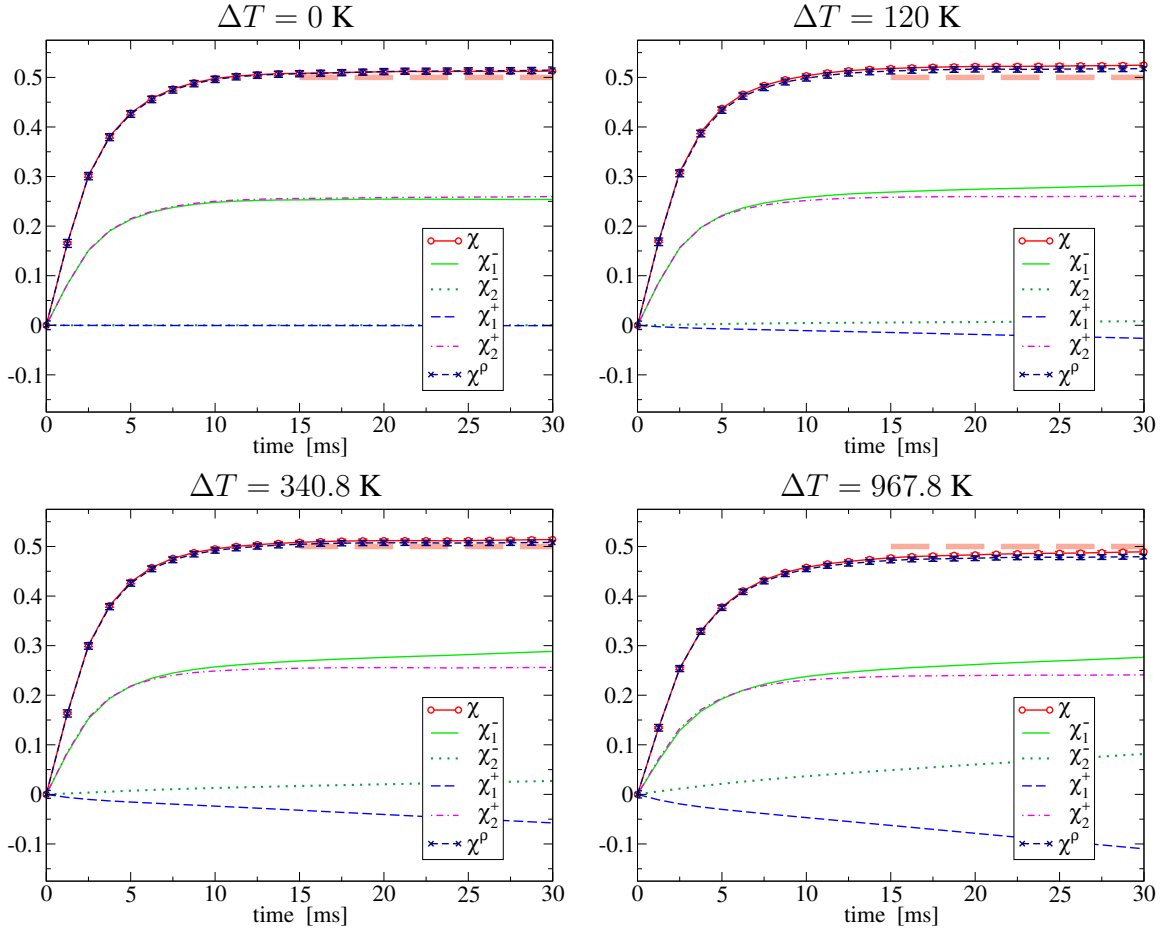


Figure 2: Response of the total energy U to a stepwise change of ΔT , for equilibrium ($\Delta T = 0$ K) and for nonequilibrium ($\Delta T = 120$ K, $\Delta T = 340.8$ K, and $\Delta T = 967.8$ K). The susceptibility χ computed with the fluctuation-response relation is shown, together with its components χ_1^- , χ_2^- , χ_1^+ , and χ_2^+ . The susceptibility χ^p computed using the response formula of Seifert and Speck [51] is also shown. The thick horizontal (pink) dashed line indicates the asymptotic value, which is determined analytically in Appendix C.

by the susceptibilities plotted in Figure 3. Again, there is a fair agreement between the two estimates. The comparison with previous numerical estimates [57], which gave very similar curves but for other values of parameters, corroborates the validity of these results.

Conclusions

We have proposed a quite general formalism for estimating the linear response of an overdamped stochastic system to a variation of its (additive) white-noise amplitude. This is a useful tool for analyzing the response of experimental data, as for the discussed system of two hydrodynamically coupled colloids in optical traps, for which we are able to compute the susceptibility to a

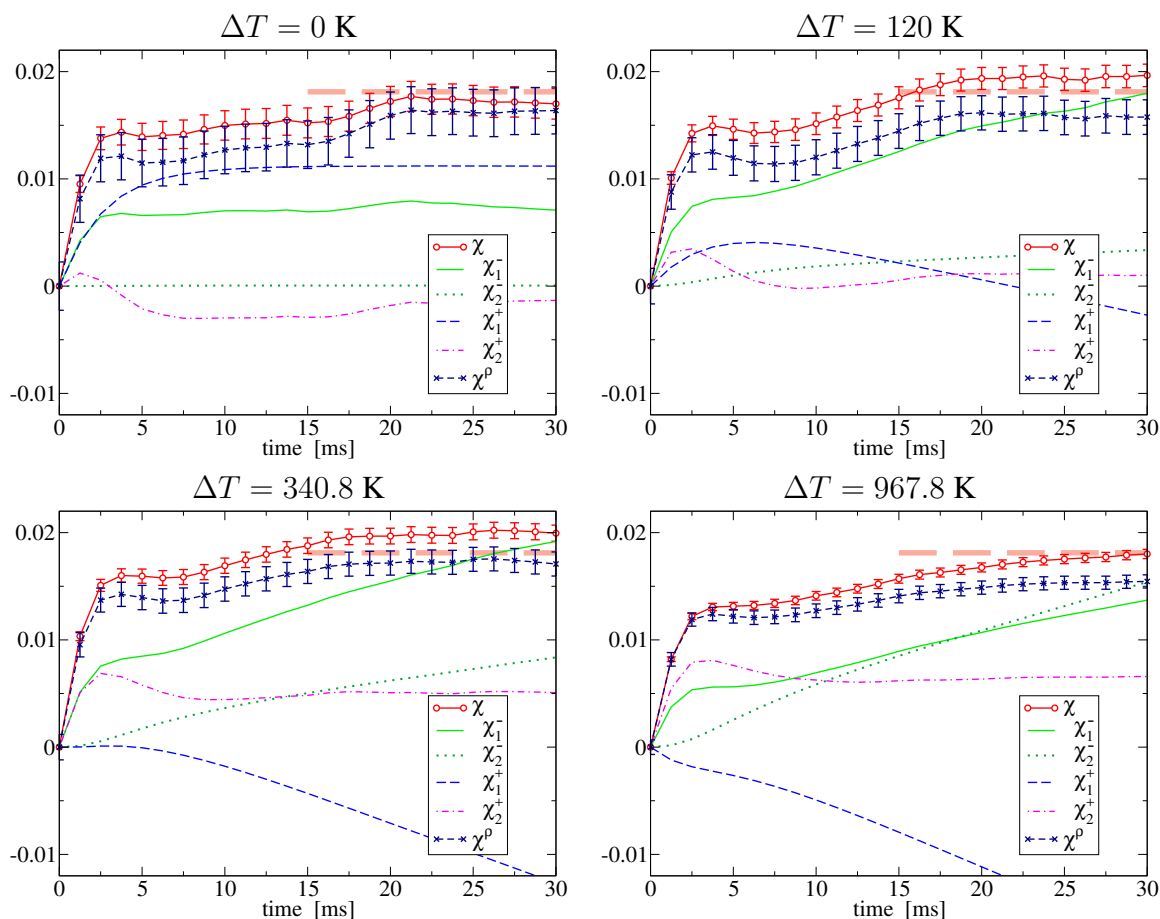


Figure 3: Response of the energy U_2 of the second particle to a stepwise change of ΔT , see the legend of Figure 2 for all the details.

change of an external random driving that reproduces a sort of higher temperature for only one of the colloids. Yet, note that our formalism is quite flexible and can be applied to more abstract stochastic diffusive dynamics with correlated noise. The next evolution of this approach should try to complete the picture by dealing with systems with multiplicative noise. Also the thermal response for nonequilibrium inertial systems should be developed without resorting to the time-discretization used in a previous work [3]. This would allow, for example, to determine nonequilibrium steady-state thermal susceptibilities of the Fermi-Pasta-Ulam model [39, 24] or other Hamiltonian models of heat conduction.

Similarly to the linear response of nonequilibrium systems to mechanical perturbations, we note that the susceptibility to temperature changes arises from dissipative as well as non-dissipative aspects of the dynamics. This differs from the simple structure of response theory for equilibrium systems, where only correlations between heat fluxes (i.e., dissipation) and observable are relevant for computing susceptibilities. With these considerations in mind, the kind of fluctuation-response relations we discussed might be used to better understand and quantify the thermal response of nonequilibrium systems such as relaxing glasses [8, 40, 16, 43, 21] and active mat-

ter [43, 41, 30, 45].

Appendices

A Stationary distribution

Here we briefly sketch the calculation of the stationary distribution of the system, for the sake of completeness.

With a drift $a(x) = -\mu\kappa x$ linear in x , the equation of motion (1) describes an Ornstein-Uhlenbeck process, which we rewrite here as

$$\dot{x} = -\Omega x + \sqrt{2D}\xi, \quad (38)$$

with $\Omega = \mu\kappa$. According to the corresponding Fokker-Planck equation, $\dot{\rho} = -\partial_i J_i$, stationarity requires the ensemble current $J = -\rho\Omega x - D\nabla\rho$ to be divergenceless, yielding

$$0 = \rho\Omega_{ii} + \Omega_{ij}x_j\partial_i\rho + D_{ij}\partial_i\partial_j\rho. \quad (39)$$

Clearly, a Gaussian distribution of the form

$$\rho(x) = Z^{-1}e^{-\frac{1}{2}x^\dagger Gx} \quad (40)$$

satisfies the zero divergence condition above. To find the inverse covariance matrix G , one substitutes the ansatz, upon which a few lines of algebra implies the condition

$$\Omega G^{-1} + G^{-1}\Omega^\dagger = 2D. \quad (41)$$

This is a system of linear equations in the elements of the covariance matrix G^{-1} , which can be solved, for instance, by rewriting it in terms of a vector composed of the columns of G^{-1} . Here, we simply quote the resulting matrix: Recalling $\Omega = \mu\kappa$ with κ diagonal and μ given in Eq. (3), and D given in Eq. (6), one finds

$$G^{-1} = \begin{bmatrix} \frac{T+(1-\varepsilon^2)\Delta T}{\kappa_{11}} + \frac{\varepsilon^2\Delta T}{\kappa_{11}+\kappa_{22}} & \frac{\varepsilon\Delta T}{\kappa_{11}+\kappa_{22}} \\ \frac{\varepsilon\Delta T}{\kappa_{11}+\kappa_{22}} & \frac{T}{\kappa_{22}} + \frac{\varepsilon^2\Delta T}{\kappa_{11}+\kappa_{22}} \end{bmatrix}. \quad (42)$$

Inversion of this matrix thus determines the stationary distribution (40).

B Form of the perturbation parameter

Even though it may sometimes be more convenient mathematically to work in terms of the noise amplitude B , it is the diffusivity matrix D which is physically relevant, since the noise amplitude is fixed only up to an orthogonal transformation. Hence the perturbation parameter, $h = \delta B\tilde{B}^{-1}$, defined in Sec. 2.1 in terms of the noise amplitude should eventually be expressed in terms of a

perturbation of the diffusivity. With $D = \tilde{D} + \delta D$ and $B = \tilde{B} + \delta B$, in line with Sec. 2.1, the definitions $2D = BB^\dagger$ and $2\tilde{D} = \tilde{B}\tilde{B}^\dagger$ imply that

$$2\delta D = \tilde{B}\delta B^\dagger + \delta B\tilde{B}^\dagger . \quad (43)$$

Note that one should not expect to solve this equation for δB uniquely given a specific δD ; many noise coefficient matrices B map into the same diffusion matrix D .

The relation above can now be used to express the perturbation parameter $h = \delta B\tilde{B}^{-1}$ in terms of \tilde{D} and δD , but not uniquely, which is not a problem. Multiplying Eq. (43) by \tilde{D}^{-1} from the left and right, one easily finds

$$\tilde{D}^{-1}\delta D\tilde{D}^{-1} = \tilde{D}^{-1}h + h^\dagger\tilde{D}^{-1} . \quad (44)$$

The left hand side of this equation is equal to $-\delta D^{-1}$ (verified easily by varying the identity $1 = DD^{-1}$) which is determined by the physical description of the perturbation. Meanwhile, the right hand side of the equation is twice the symmetric part of $\tilde{D}^{-1}h$. In other words, it is only the symmetric part of $\tilde{D}^{-1}h$ that is fixed by the physical form of the perturbation, and the antisymmetric part is left undetermined. It therefore behooves one to choose $\tilde{D}^{-1}h$ to be purely symmetric, whence one obtains $-\delta D^{-1} = 2\tilde{D}^{-1}h$, or

$$h = -\frac{1}{2}\tilde{D}\delta D^{-1} = -\frac{1}{2}D\delta D^{-1} , \quad (45)$$

where the second equality is valid due to the overarching first order approximation of linear response.

C Asymptotic values of the susceptibility

In this appendix, we sketch how the asymptotic values for the susceptibilities in Figures 2 and 3 were obtained.

One can derive a host of relations valid in the stationary regime by requiring that time derivatives of state observables vanish [28]. For the present discussion, the relevant observable is the tensor $x_i x_j$, *i.e.*,

$$0 = \frac{d}{dt} \langle x_i x_j \rangle = \langle \mathbb{L} x_i x_j \rangle , \quad (46)$$

with the backward generator $\mathbb{L} = a_i \partial_i + D_{ij} \partial_i \partial_j$. (One should keep in mind that the average is in the stationary regime, although we leave it unlabeled.) When evaluated explicitly, with $a = \mu F$, one finds the relation

$$0 = \mu \langle F x^\dagger \rangle + \langle x F^\dagger \rangle \mu + 2D . \quad (47)$$

This matrix relation entails a set of equations for the independent components of the tensor $\langle x_i F_j \rangle$, of which there are 3 in our case with 2 degrees of freedom.

We note that for our system, $U(x) = -(1/2)\text{Tr}(Fx)$. Thus, multiplying Eq. (47) from the left by μ^{-1} and taking the trace, we find the stationary average of the potential energy to be

$$\langle U(x) \rangle = -\frac{1}{2}\text{Tr} \langle Fx \rangle = \frac{1}{2}\text{Tr}(\mu^{-1}D). \quad (48)$$

Hence, the stationary (asymptotic) value for the susceptibility is found as

$$\frac{\partial \langle U \rangle}{\partial \Delta T} = \frac{1}{2}\text{Tr} \left(\mu^{-1} \frac{\partial D}{\partial \Delta T} \right) = \frac{1}{2}, \quad (49)$$

which was evaluated using Eq. (6) for the diffusion matrix. This asymptotic value for χ was indicated in Figure 2.

The susceptibility of the energy of the second particle $U_2(x_2) = -(1/2)F_2x_2$ can be extracted similarly from Eq. (47), with the exception that one has to go through the tedium of actually solving for the component $\langle F_2x_2 \rangle$. We quote only the result of this straightforward exercise:

$$\frac{\partial \langle U_2 \rangle}{\partial \Delta T} = -\frac{1}{2} \frac{\partial}{\partial \Delta T} \langle F_2x_2 \rangle = \frac{\varepsilon^2}{2 \left(1 + \frac{\kappa_{11}}{\kappa_{22}} \right)}. \quad (50)$$

This was evaluated as 0.01899 for the actual experimental values of $\kappa_{11} = 3.3745$ pN/ μm , $\kappa_{22} = 3.3285$ pN/ μm , and $\varepsilon = 0.2766$, and indicated in Figure 3 as the asymptotic value of the susceptibility χ .

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