Derivation of the nonequilibrium generalized Langevin equation from a time-dependent many-body Hamiltonian

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It has become standard practice to describe systems that remain far from equilibrium even in their steady state by Langevin equations with colored noise which is chosen independently from the friction contribution. Since these Langevin equations are typically not derived from first-principle Hamiltonian dynamics, it is not clear whether they correspond to physically realizable scenarios. By exact Mori projection in phase space we derive the nonequilibrium generalized Langevin equation (GLE) for an arbitrary phase-space dependent observable A from a generic many-body Hamiltonian with a time-dependent external force $h(t)$ acting on the same observable A. This is the same Hamiltonian from which the standard fluctuation-dissipation theorem is derived, which reflects the generality of our approach. The observable A could, for example, be the position of an atom, of a molecule or of a macroscopic object, the distance between two such entities or a more complex phase-space function such as the reaction coordinate of a chemical reaction or of the folding of a protein. The Hamiltonian could, for example, describe a fluid, a solid, a viscoelastic medium, or even a turbulent inhomogeneous environment. The GLE, which is a closed-form equation of motion for the observable A, is obtained in explicit form to all orders in $h(t)$ and without restrictions on the type of many-body Hamiltonian or the observable A. If the dynamics of the observable A corresponds to a Gaussian process, the resultant GLE has a similar form as the equilibrium Mori GLE, and in particular the friction memory kernel is given by the two-point autocorrelation function of the sum of the complementary and the external force $h(t)$. This is a nontrivial and useful result, as many observables that characterize nonequilibrium systems display Gaussian statistics. For non-Gaussian nonequilibrium observables correction terms appear in the GLE and in the relation between the force autocorrelation and the friction memory kernel, which are explicitly given in terms of cubic correlation functions of A. Interpreting the external force $h(t)$ as a stochastic process, we derive nonequilibrium corrections to the fluctuation-dissipation theorem and present methods to extract all GLE parameters from experimental or simulation time-series data, thus making our nonequilibrium GLE a practical tool to study and model general nonequilibrium systems.

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I. INTRODUCTION

The statistical mechanics foundation of nonequilibrium phenomena has occupied physicists for many decades [1–8]. More recently, new experimental techniques, such as single-molecule and optical methods, applied to nonequilibrium synthetic and biological systems [9–18] have accented the need for theories that are able to deal with nonequilibrium experiments and data. At the same time, novel theoretical approaches were developed and applied to nonequilibrium driven lattice models [19–21], interacting nonequilibrium particle systems [22–25], and nonequilibrium barrier-crossing phenomena [26,27] and used to derive nonequilibrium work and entropy relations [28–32], generalized nonequilibrium fluctuation-dissipation theorems (FDTs) [33–39], and nonequilibrium entropy-production extremal principles [40].

The generalized Langevin equation (GLE) has played a key role in the development of methods to deal with the dynamics of complex systems, as it is an exact equation of motion for an observable derived by projection from the many-body Hamiltonian; in other words, projection constitutes a method for exact coarse graining of a Hamiltonian system [41–53]. The GLE was applied to protein folding [54–57], barrier crossing dynamics [26,58–61], motion of living cells [17,18], spectroscopy [62–64], dynamical neworks [65], and data prediction [66]. While the standard GLE formulations deal with the motion of an observable in phase space described by a time-independent Hamiltonian and thus allow one to quantify the approach of a nonequilibrium state to equilibrium, they do not apply to driven nonequilibrium system as described by a time-dependent Hamiltonian. Many works dealt with generalizations of the projection framework to time-dependent and transient scenarios [67–75]. None of these works dealt with the specific time-dependent Hamiltonian system considered in this paper, where a time-dependent force couples to the observable of interest, and derived the nonequilibrium GLE in explicit form. Since models that apply the GLE framework to nonequilibrium phenomena typically choose the noise and friction terms with a certain degree of freedom [10,17,18,22–27,76], it is instructive to derive the nonequilibrium GLE from a time-dependent Hamiltonian. This enables us to check which nonequilibrium GLEs correspond to an underlying nonequilibrium Hamiltonian dynamics and which do not. This is the vantage point of this paper.

For an equilibrium system, here defined to be a system described by a time-independent Hamiltonian, the Mori GLE for the generic observable $A(t)$ reads [43]

$$\ddot{A}(t) = -K[A(t) - \langle A\rangle] - \int_{t_0}^{t} ds\, \Gamma(t-s)\dot{A}(s) + F(t), \quad (1)$$

where the stiffness of the effective harmonic potential is denoted as K, the time-dependent friction memory kernel as $\Gamma(t)$, and the complementary force as $F(t)$. The time at which the projection is done is denoted as t_0, and the friction kernel $\Gamma(t)$ is proportional to the complementary force autocorrelation function via [43]

$$\Gamma(t-s) = \frac{\langle F(s)F(t)\rangle}{\langle \dot{A}^2(t)\rangle} \quad (2)$$

[all averages are phase-space averages, and the observable $A(t)$ in Eq. (1) is in fact phase-space dependent, as will be detailed below]. The relation in Eq. (2) is often regarded as equivalent to the FDT, which is not true in general, since even for equilibrium systems it holds only for the present Mori GLE but not for a wider class of GLEs that are derived with nonlinear projection operators [52,77,78]. Note that $F(t)$ is often denoted and treated as a random force; this is an approximation since $F(t)$ is a phase-space dependent deterministic function and fulfills well-defined initial conditions at $t = t_0$, and Eq. (1) is thus deterministic and fully time reversible. In fact, despite its linear appearance, the Mori GLE in Eq. (1) is exact even for nonlinear systems, unless the complementary force $F(t)$ is approximated as Gaussian.

In the presence of an external (i.e., phase-space independent) time-dependent force $h(t)$ that couples to a phase-space dependent observable $A(t)$, a Hamiltonian system is generally out of equilibrium since the external force performs work on the system. In fact, even for constant force $h(t) = h_0 \neq 0$ such a system is out of equilibrium if the observable A is unconfined and thus driven into a steady-state motion, as will be explained below.

In this paper we derive the nonequilibrium GLE for an arbitrary phase-space dependent observable A, governed by a general many-body Hamiltonian that includes a time-dependent external force $h(t)$ acting on A, and our derivation of the GLE is nonperturbative and thus exact to all orders in $h(t)$. It is important to note that we do not impose any restriction on the observable A or the many-body Hamiltonian. Accordingly, A could represent the position of an atom, of a molecule, or of a macroscopic object and the distance between two objects or a more complex and nonlinear phase-space function, such as the reaction coordinate of a chemical reaction or of the folding of a protein. The Hamiltonian could, for example, describe a fluid, a solid, or a viscoelastic medium or even a turbulent inhomogeneous environment. The time-dependent force $h(t)$ would in the simplest scenario correspond to a force that drags a particle through a medium, as can experimentally be realized with optical tweezers [11] or by applying a time-dependent electric field. It could also correspond to a force that acts on the separation between two amino acids in a protein and thereby couples to the folding reaction coordinate, as can be experimentally realized in dual-optical-tweezer pulling experiments [79]. The specific time-dependent Hamiltonian we are employing is of high relevance, as it forms the starting point for the textbook derivation of the standard FDT [8], which is one of the corner stones of statistical mechanics. Thus, the standard FDT and the nonequilibrium GLE we derive in this paper are intimately connected since they stem from the same time-dependent Hamiltonian.

A key question we address in this paper is whether in the presence of an external time-dependent force $h(t)$ acting on A, the GLE in Eq. (1) and the relation between the friction kernel and the complementary force autocorrelation in Eq. (2) still hold. Indeed, one main result of this paper is that the simple forms of Eqs. (1) and (2) indeed remain valid if the observable A corresponds to a Gaussian process and if $F(t)$ is replaced by the sum of $F(t)$ and $h(t)$. This is a nontrivial and relevant finding, in particular since many biological nonequilibrium processes, such as the motion of cells, are Gaussian to high accuracy [17,18]. Conversely, for a non-Gaussian observable A, correction terms appear in Eqs. (1) and (2) that depend on three-point correlation functions of A. The importance of non-Gaussianity has recently been stressed in the context of nonequilibrium granular and linear model systems [80,81]. Our explicit form of the GLE allows for quantitative prediction of the nonequilibrium correction terms based on experimental or simulation time-series data. For the special case of a stochastic nonequilibrium force $h(t)$ that is defined by its second moment, we derive a generalized nonequilibrium FDT, which in the limit $h(t) \to 0$ simplifies to the standard FDT. For this stochastic scenario, we give explicit formulas for extracting the parameters of our nonequilibrium GLE from simulation or experimental time-series data, opening the route to the accurate and data-based modeling of systems that are far from equilibrium.

Section II contains the full derivation of the nonequilibrium GLE; this section can be skipped by readers not interested in technical details. In Sec. III the nonequilibrium GLE is discussed and the role of non-Gaussian observable fluctuations is explained. In Sec. IV the nonequilibrium force $h(t)$ is treated as a stochastic variable, which restores time homogeneity and simplifies the analysis of the GLE. Here the nonequilibrium response function and FDT are derived. Section V presents a short discussion and an outlook. Details of the derivations and calculations are presented in 11 Appendixes.

II. DERIVATION OF THE NONEQUILIBRIUM GENERALIZED LANGEVIN EQUATION

A. Definition of the time-dependent Hamiltonian and solution of the Liouville equation

We consider a time-dependent Hamiltonian for a system of N interacting particles or atoms in three-dimensional space with a time-dependent phase-space independent force $h(t)$ that couples to a generic phase-space dependent observable $A_S(\omega)$,

$$H(\omega, t) = H_0(\omega) - h(t)A_S(\omega), \quad (3)$$

where the subscript distinguishes this Schrödinger-like, i.e., time-independent, observable from the time-dependent Heisenberg observable that will be introduced shortly. Although not really needed for our derivation, the time-independent Hamiltonian $H_0(\omega)$ can be split into kinetic and potential contributions according to

$$H_0(\omega) = \sum_{j=1}^{3N} \frac{P_j^2}{2m_j} + V(\mathbf{R}) \tag{4}$$

with coordinate-dependent masses m_j and where the potential $V(\mathbf{R})$ contains all interactions between the particles and includes possible external potentials. As explained earlier, we impose no restrictions on the observable $A_S(\omega)$ or on the Hamiltonian $H_0(\omega)$. A point in $6N$-dimensional phase space is denoted by $\omega = (\mathbf{R}, \mathbf{P})$, which is a $6N$-dimensional vector containing the Cartesian particle positions $\mathbf{R}$ and the conjugate momenta $\mathbf{P}$ and fully specifies the microstate of the system.

Using the time-dependent Liouville operator

$$\mathcal{L}(\omega, t) = \sum_{j=1}^{3N} \left(\frac{\partial H(\omega, t)}{\partial P_j} \frac{\partial}{\partial R_j} - \frac{\partial H(\omega, t)}{\partial R_j} \frac{\partial}{\partial P_j} \right), \tag{5}$$

the $6N$-dimensional Hamilton equation of motion can be compactly written as $\dot{\omega}(t) = \mathcal{L}(\omega, t)\omega(t)$, where $\omega(t)$ is the phase-space location of the system at time t and $\dot{\omega}(t) = d\omega(t)/dt$ is the corresponding phase-space velocity. Instead of following microstate trajectories in phase space, which is the Lagrangian description of the system dynamics, it is much more convenient to switch to the Eulerian description and consider the time-dependent probability density distribution as a function of the time-independent phase-space position, $\rho(\omega, t)$, which obeys the Liouville equation

$$\dot{\rho}(\omega, t) = -\mathcal{L}(\omega, t)\rho(\omega, t). \tag{6}$$

In all of what follows we suppress the dependence of the Liouville operator on phase space. We observe that a recursive solution of Eq. (6) can be written as

$$\rho(\omega, t) = \rho(\omega, t_0) - \int_{t_0}^{t} dt_1 \mathcal{L}(t_1)\rho(\omega, t_1). \tag{7}$$

By iteration the following exact solution is obtained:

$$\rho(\omega, t) = \exp_S \left(- \int_{t_0}^{t} ds \mathcal{L}(s) \right) \rho(\omega, t_0), \tag{8}$$

which depends on the initial density distribution at time t_0 and where the time-ordered operator exponential in the Schrödinger picture has been introduced as [82,83]

$$\exp_S \left(- \int_{t_0}^{t} ds \mathcal{L}(s) \right) \equiv 1 + \sum_{n=1}^{\infty} (-1)^n \int_{t_0}^{t} dt_1 \mathcal{L}(t_1) \int_{t_0}^{t_1} dt_2 \mathcal{L}(t_2) \int_{t_0}^{t_2} dt_3 \mathcal{L}(t_3) \cdots \int_{t_0}^{t_{n-1}} dt_n \mathcal{L}(t_n). \tag{9}$$

For a time-independent Liouville operator $\mathcal{L}(t) = \mathcal{L}_0$, all nested time integrals can be trivially done, and one obtains the solution in terms of the standard operator exponential

$$\rho(\omega, t) = \exp\left(-(t - t_0)\mathcal{L}_0\right)\rho(\omega, t_0), \tag{10}$$

where the exponential of an operator is defined by its ordinary series expansion.

B. From Schrödinger to Heisenberg observables

A system observable can be generally written as a Schrödinger-type phase-space function $A_S(\omega)$. It can, for example, represent the position of one particle, the center-of-mass position of a group of particles or of a molecule, the reaction coordinate describing a chemical reaction, or the folding of a protein. To simplify the notation, we consider a scalar observable but note that our derivation can be straightforwardly extended to multidimensional observables. Using the probability density $\rho(\omega, t)$, the time-dependent expectation value (or mean) of the observable $A_S(\omega)$ can be written as

$$\begin{aligned} a(t) &\equiv \int d\omega\, A_S(\omega)\rho(\omega, t) \\ &= \int d\omega\, A_S(\omega) \exp_S \left(- \int_{t_0}^{t} ds \mathcal{L}(s) \right) \rho(\omega, t_0). \end{aligned} \tag{11}$$

Since the Liouville operator is anti-self-adjoint, it follows that [8]

$$a(t) = \int d\omega\, \rho(\omega, t_0) A(\omega, t), \tag{12}$$

where we have defined the Heisenberg observable as

$$A(\omega, t) \equiv \exp_H \left(\int_{t_0}^{t} ds \mathcal{L}(s) \right) A_S(\omega) \tag{13}$$

using the time-ordered operator exponential in the Heisenberg picture (i.e., the Heisenberg propagator),

$$\exp_H \left(\int_{t_0}^{t} ds \mathcal{L}(s) \right) \equiv 1 + \sum_{n=1}^{\infty} \int_{t_0}^{t} dt_1 \int_{t_0}^{t_1} dt_2 \cdots \int_{t_0}^{t_{n-1}} dt_n \mathcal{L}(t_n) \cdots \mathcal{L}(t_2)\mathcal{L}(t_1). \tag{14}$$

Obviously, as follows from Eqs. (13) and (14), the time derivative of the Heisenberg observable is given by

$$\dot{A}(\omega, t) = \frac{dA(\omega, t)}{dt} = \exp_H \left(\int_{t_0}^{t} ds \mathcal{L}(s) \right) \mathcal{L}(t) A_S(\omega) \tag{15}$$

and the initial condition as $A(\omega, t_0) = A_S(\omega)$. As derived in Appendix A, the Heisenberg observable also satisfies the initial differential boundary condition

$$\frac{dA(\omega, t)}{dt_0} = -\mathcal{L}(t_0) \exp_H \left(\int_{t_0}^{t} ds \mathcal{L}(s) \right) A_S(\omega), \tag{16}$$

which will be needed later to derive operator expansions.

To understand the meaning of a Heisenberg observable, we for the moment consider the initial density distribution $\rho(\omega, t_0) = \delta(\omega - \omega_0)$, which describes a system that at time t_0 is in the microstate ω_0. Inserting this into Eq. (12), we obtain $a(t) = A(\omega_0, t)$. In other words, $A(\omega_0, t)$ describes the time-dependent mean of an observable for a system that at time $t = t_0$ was in the microstate ω_0, i.e., it describes the temporal evolution of the conditional mean of the observable $A_S(\omega)$. It transpires that if we derive an equation of motion for $A(\omega, t)$, we have an equation for how this conditional mean changes in time. This is the central idea of projection and of GLEs [41–43].

Taking another time derivative of Eq. (15), we obtain for the acceleration of the observable

$$\ddot{A}(\omega, t) = \exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(t)\right)(\mathcal{L}^2(t) + \dot{\mathcal{L}}(t))A_S(\omega). \quad (17)$$

Up to now the discussion applied to a general time-dependent Hamiltonian; for the specific Hamiltonian Eq. (3), where a time-dependent external force $h(t)$ multiplies the observable $A_S(\omega)$, the Liouville operator splits into two parts

$$\mathcal{L}(t) = \mathcal{L}_0 - h(t)\Delta\mathcal{L}, \quad (18)$$

with the unperturbed Liouville operator given by

$$\mathcal{L}_0 = \sum_{j=1}^{3N}\left(\frac{\partial H_0(\omega)}{\partial P_j}\frac{\partial}{\partial R_j} - \frac{\partial H_0(\omega)}{\partial R_j}\frac{\partial}{\partial P_j}\right) \quad (19)$$

and the perturbation Liouville operator given by

$$\Delta\mathcal{L} = \sum_{j=1}^{3N}\left(\frac{\partial A_S(\omega)}{\partial P_j}\frac{\partial}{\partial R_j} - \frac{\partial A_S(\omega)}{\partial R_j}\frac{\partial}{\partial P_j}\right). \quad (20)$$

These operators have the important properties $\mathcal{L}_0 H_0(\omega) = 0$, $\Delta\mathcal{L}A_S(\omega) = 0$, and $\Delta\mathcal{L}H_0(\omega) = -\mathcal{L}_0 A_S(\omega)$, from which we derive, using Eqs. (15) and (17), the simplified expressions for the observable velocity and acceleration

$$\dot{A}(\omega, t) = \exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right)\mathcal{L}_0 A_S(\omega), \quad (21)$$

$$\ddot{A}(\omega, t) = \exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right)\mathcal{L}(t)\mathcal{L}_0 A_S(\omega). \quad (22)$$

The fact that the velocity (21) exhibits no time dependence to the right of the operator exponential is crucial: It will later allow us to use time-independent projection for the derivation of the nonequilibrium GLE in explicit and rather simple form. The reason for this massive simplification is the fact that we derive the GLE for the same observable $A_S(\omega)$ that appears in the time-dependent perturbation in the Hamiltonian (3).

C. Projection

Here we follow standard procedures [8,41–43]. We introduce a time-independent projection operator $\mathcal{P}$ that acts on a phase space function and its complementary operator $\mathcal{Q}$ via the relation $1 = \mathcal{Q} + \mathcal{P}$. Inserting this unit operator into the acceleration (22), we obtain

$$\begin{aligned}\ddot{A}(\omega, t) &= \exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right)(\mathcal{P} + \mathcal{Q})\mathcal{L}(t)\mathcal{L}_0 A_S(\omega)\\ &= \exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right)\mathcal{P}\mathcal{L}(t)\mathcal{L}_0 A_S(\omega)\\ &\quad + \exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right)\mathcal{Q}\mathcal{L}(t)\mathcal{L}_0 A_S(\omega), \end{aligned} \quad (23)$$

where we used that the Heisenberg propagator is a linear operator. The projection is performed at time t_0 at which the time propagation starts (the relevance of this will become clear later). By inserting the time-dependent Dyson operator expansion [41–43,82,83] for the Heisenberg propagator (see Appendix B for a derivation)

$$\exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right) = \exp_H\left(\mathcal{Q}\int_{t_0}^{t} ds\mathcal{L}(s)\right) + \int_{t_0}^{t} ds\exp_H\left(\int_{t_0}^{s} ds'\mathcal{L}(s')\right)\mathcal{P}\mathcal{L}(s)\exp_H\left(\mathcal{Q}\int_{s}^{t} ds'\mathcal{L}(s')\right) \quad (24)$$

into the second term on the right-hand side in Eq. (23), we obtain the GLE in general form,

$$\ddot{A}(\omega, t) = \exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right)\mathcal{P}\mathcal{L}(t)\mathcal{L}_0 A_S(\omega) + F(\omega, t_0, t) + \int_{t_0}^{t} ds\exp_H\left(\int_{t_0}^{s} ds'\mathcal{L}(s')\right)\mathcal{P}\mathcal{L}(s)F(\omega, s, t), \quad (25)$$

where the complementary force is defined as

$$F(\omega, t_0, t) \equiv \exp_H\left(\mathcal{Q}\int_{t_0}^{t} ds'\mathcal{L}(s')\right)\mathcal{Q}\mathcal{L}(t)\mathcal{L}_0 A_S(\omega). \quad (26)$$

The first term on the right-hand side in Eq. (25) will turn out to represent the conservative force from a potential, the third term represents non-Markovian friction effects, and the force $F(\omega, t_0, t)$ represents all effects that are not included in the other two terms. $F(\omega, t_0, t)$ is a function of phase space and evolves in the complementary space, i.e., it satisfies $\mathcal{P}F(\omega, t_0, t) = 0$ (as will be explained further below). While we suppress the t_0 dependence of the observable $A(\omega, t)$ and its derivatives, which can cause no confusion since this argument is invariant throughout most of the calculation, the complementary force $F(\omega, t_0, t)$ needs the t_0 argument since it varies in Eq. (25).

Clearly, the explicit form of Eq. (25) depends on the specific projection operator $\mathcal{P}$. Here we choose the Mori projection, because it is straightforward to implement and our result concerning the effect of non-Gaussian observables on the structure of the nonequilibrium GLE is accurately and transparently produced by Mori projection. We repeat in passing that the Mori GLE is exact even for non-Gaussian observables, unless one approximates the complementary force distribution. The Mori projection applied on a general Heisenberg observable $B(\omega, t)$ using the Schrödinger observable

$A_S(\omega)$ as a projection function is given by [43]

$$\begin{aligned}\mathcal{P}B(\omega,t)&\\ &= \langle B(\omega,t)\rangle + \frac{\langle B(\omega,t)\mathcal{L}_0A_S(\omega)\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}\mathcal{L}_0A_S(\omega)\\ &\quad+\frac{\langle B(\omega,t)(A_S(\omega)-\langle A_S\rangle)\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}[A_S(\omega)-\langle A_S\rangle],\end{aligned} \tag{27}$$

where we use a particularly useful form that involves threefold projection on a constant, the deviations of the observable A_S from its mean and its velocity [77,78]. Here we have defined the expectation value of an arbitrary phase-space function $X(\omega)$ with respect to a time-independent projection distribution $\rho_{\rm p}(\omega)$ as

$$\langle X(\omega)\rangle = \int d\omega X(\omega)\rho_{\rm p}(\omega), \tag{28}$$

which we take to be the equilibrium canonical distribution of the time-independent Hamiltonian

$$\rho_{\rm p}(\omega) = e^{-\beta H_0(\omega)+\beta h_p A_S(\omega)}/Z, \tag{29}$$

where Z is the normalizing partition function. The factor β has units of inverse energy and can be thought of as the inverse thermal energy characterizing the projection distribution. Note that for generality we added a linear force h_p in the projection Hamiltonian, which can be chosen as $h_p = h(t_0)$ for continuity of the Hamiltonian. Time-dependent projection has been used to derive generic nonequilibrium GLEs [67–75], but is not needed here because of the specific form of our time-dependent Hamiltonian which involves a time-dependent force $h(t)$ acting on the same observable $A_S(\omega)$ for which we derive the GLE. The time-independent Mori projection in Eq. (27) projects onto a constant, the Schrödinger observable $A(\omega,t_0) = A_S(\omega)$ and its time derivative $\dot{A}(\omega,t_0) = \mathcal{L}_0A_S(\omega)$. Thus, the projection in Eq. (27) maps any observable $B(\omega,t)$ onto the subspace of all functions linear in 1, $A_S(\omega)$ and $\mathcal{L}_0A_S(\omega)$, meaning that $\mathcal{P}1 = 1$, $\mathcal{P}A_S(\omega) = A_S(\omega)$ and $\mathcal{P}\mathcal{L}_0A_S(\omega) = \mathcal{L}_0A_S(\omega)$. From this follows immediately that $\mathcal{Q}1 = \mathcal{Q}A_S(\omega) = \mathcal{Q}\mathcal{L}_0A_S(\omega) = 0$, which are important properties that allow one to show that several expectation values involving the complementary force vanish, namely, $\langle F(\omega,t_0,t)\rangle = \langle F(\omega,t_0,t)A_S(\omega)\rangle = \langle F(\omega,t_0,t)\mathcal{L}_0A_S(\omega,)\rangle = 0$. The latter relations will be later used to extract GLE parameters from nonequilibrium time-series data.

The Mori projection is linear, i.e., for two arbitrary observables $B(\omega,t)$ and $C(\omega,t')$ it satisfies $\mathcal{P}(c_1B(\omega,t)+c_2C(\omega,t')) = c_1\mathcal{P}B(\omega,t)+c_2\mathcal{P}C(\omega,t')$, it is idempotent, i.e., $\mathcal{P}^2 = \mathcal{P}$, and it is self-adjoint, i.e., it satisfies the relation

$$\langle C(\omega,t)\mathcal{P}B(\omega,t')\rangle = \langle B(\omega,t')\mathcal{P}C(\omega,t)\rangle. \tag{30}$$

For these projection properties to hold we assume that $A_S(\omega) = A_S(\mathbf{R})$ is a function of Cartesian particle positions only. From these properties it follows that the complementary projection operator $\mathcal{Q} = 1-\mathcal{P}$ is also linear, idempotent, and self-adjoint. Thus, $\mathcal{P}$ and $\mathcal{Q}$ are orthogonal to each other, i.e., $\mathcal{P}\mathcal{Q} = 0 = \mathcal{Q}\mathcal{P}$, details are shown in Appendix C.

III. PROPERTIES OF THE NONEQUILIBRIUM LANGEVIN EQUATION

A. General properties

Using the projection (27) in the generic GLE (25), we obtain the nonequlibrium Mori GLE

$$\begin{aligned}\ddot{A}(\omega,t) =& -K(t)[A(\omega,t)-\langle A_S\rangle] - \int_{t_0}^t ds\,\Gamma(s,t)\dot{A}(\omega,s)\\ &+\int_{t_0}^t ds\,\Gamma_A(s,t)[A(\omega,s)-\langle A_S\rangle]\\ &+F(\omega,t_0,t)+[h(t)-h_p]/M;\end{aligned} \tag{31}$$

the details of the derivation are shown in Appendix D. Equation (31) is an exact and explicit equation of motion for the Heisenberg observable $A(\omega,t)$ and is time-reversible, which is a consequence of the time reversibility of the underlying Hamilton and Liouville equations. Inspection of the GLE shows that $F(\omega,t_0,t)$ is the only term [except $h(t)$] in the GLE that accounts for possible nonlinearities (i.e., non-Gaussian contributions) in $A(\omega,t)$. Thus, imposing $F(\omega,t_0,t)$ to be a Gaussian variable corresponds to a severe approximation for nonlinear systems. On the other hand, keeping the full non-Gaussian contributions of $F(\omega,t_0,t)$ makes Eq. (31) an exact description of the observable dynamics.

The first term in Eq. (31) is a force due to an effective harmonic potential with a time-dependent potential stiffness $K(t)$ given by

$$K(t) = K_0 + K_1(t) \tag{32}$$

with

$$K_0 = \frac{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}, \tag{33}$$

$$K_1(t) = \frac{-\beta[h(t)-h_p]\langle[A_S(\omega)-\langle A_S\rangle][\mathcal{L}_0A_S(\omega)]^2\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}. \tag{34}$$

The second term in Eq. (31) accounts for linear friction and depends on the memory kernel given by

$$\Gamma(s,t) = \Gamma_0(s,t) + \Gamma_1(s,t) \tag{35}$$

with

$$\Gamma_0(s,t) = \frac{\langle F(\omega,s,s)F(\omega,s,t)\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}, \tag{36}$$

$$\Gamma_1(s,t) = \frac{-\beta(h(s)-h_p)\langle F(\omega,s,t)[\mathcal{L}_0A_S(\omega)]^2\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}. \tag{37}$$

There is also a positional memory term which is not present in the equilibrium GLE in Eq. (1) and which involves the kernel function

$$\Gamma_A(s,t) = \frac{\beta(h(s)-h_p)\langle F(\omega,s,t)[A_S(\omega)-\langle A_S\rangle]\mathcal{L}_0A_S(\omega)\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}; \tag{38}$$

note that this positional memory term can be eliminated by partial integration, as done in Sec. IV D for the extraction of GLE parameters from time-series data. The last two terms in Eq. (31) are the complementary force $F(\omega,t_0,t)$ defined in Eq. (26) and the external force $h(t)$, where the mass is given by

$$M = \frac{1}{\beta\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle}. \tag{39}$$

For vanishing force $h(t) = 0$, which renders the equilibrium scenario, and choosing $h_p = 0$, we see that $K_1(t) = \Gamma_1(s,t) = \Gamma_A(s,t) = 0$, and we thus recover the standard form of the equilibrium Mori GLE in Eq. (1), where in particular the friction kernel $\Gamma_0(s,t)$ is via Eq. (2) related to the complementary force autocorrelation function [note that in Eqs. (1) and (2) we have suppressed the phase-space dependence of the observable A and of the complementary force F]. In contrast, if $h(t) - h_p \neq 0$, we see that additional terms are present in the GLE and that for $\Gamma_1(s,t) \neq 0$ Eq. (2) does not hold, i.e., the friction kernel $\Gamma(s,t)$ does not equal the complementary force autocorrelation function. We first want to discuss whether a nonzero $\Gamma_1(s,t)$ necessarily indicates the presence of nonequilibrium effects.

An insightful scenario to address this question is one where the force $h(t) = h_0$ is constant, in which case the complementary force and all memory kernels become time homogeneous and can be written as $F(\omega,s,t) = F(\omega,t-s)$, $\Gamma(s,t) = \Gamma(t-s)$, $\Gamma_A(s,t) = \Gamma_A(t-s)$. Let us first discuss an unconfined system, i.e., a system characterized by a diverging second moment, $\langle[A_S(\omega) - \langle A_S\rangle]^2\rangle = \infty$. In this case $K = 0 = \Gamma_A(t-s)$, and we must take $h_p = 0$ in order to have a bounded projection distribution $\rho_p(\omega)$ in Eq. (29). From Eq. (37) we see that $\Gamma_1(t-s)$ can in general be nonzero (as we will discuss in more detail in the next section), in which case the total friction memory kernel $\Gamma(t-s)$ does not equal the complementary force autocorrelation, reflecting that an unconfined system under the influence of a constant force dissipates energy and thus is a nonequilibrium system. In contrast, a confined system that is characterized by a finite second moment $\langle[A_S(\omega) - \langle A_S\rangle]^2\rangle$ is in the presence of a constant force $h(t) = h_0$ an equilibrium system. But we see that for $h_0 \neq h_p$ the terms $K_1(t)$, $\Gamma_1(s,t)$, and $\Gamma_A(s,t)$ do not vanish in general. In other words, unless we choose as the projection distribution in Eq. (29) the stationary distribution with $h_p = h_0$, the friction kernel in the Mori GLE $\Gamma(s,t)$ does not equal the complementary force autocorrelation. Thus, a nonzero $\Gamma_1(s,t)$ not necessarily indicates a nonequilibrium system, but can also be produced by choosing a nonstationary projection distribution $\rho_p(\omega)$ and in this case characterizes the approach of the system towards its equilibrium distribution. Conversely, and as we will derive in the next section, $\Gamma_1(s,t)$ is predicted to vanish for Gaussian nonequilibrium systems, so not all nonequilibrium systems are characterized by a nonvanishing value of $\Gamma_1(s,t)$. In summary, the condition $\Gamma(s,t) = \Gamma_0(s,t)$ with $\Gamma(s,t)$ and $\Gamma_0(s,t)$ defined in Eqs. (35) and (36), equivalent to the standard equilibrium FDT for the Mori GLE, as we will show further below, does not necessarily indicate an equilibrium system, and conversely, the condition $\Gamma(s,t) \neq \Gamma_0(s,t)$ does not necessarily indicate a nonequilibrium system. Having made this important point, we from now on put $h_p = 0$.

Note that for equilibrium systems governed by a time-independent Hamiltonian, the GLE parameters and the complementary force can be extracted from simulation or experimental time series data by various well-established techniques [45,52,53,84–88]. Similar extraction techniques for nonequilibrium time-series data will be discussed in Sec. IV for an external stochastic force $h(t)$. We mention in passing that the external force $h(t)$ in general performs work on the system, meaning that the total energy of the system will in general increase with time. Since the GLE (31) is exact, it correctly takes into account these transient energetic effects.

B. Gaussian versus non-Gaussian observables

In order to highlight the role played by non-Gaussian fluctuations of the observable $A(\omega,t)$, we slightly rewrite the friction memory kernel in Eq. (35) as

$$\Gamma(s,t) = \Gamma_0(s,t) + \beta h(s)h(t)/M + \Gamma_2(s,t) \tag{40}$$

with

$$\Gamma_2(s,t) = \frac{-\beta h(s)\langle[F(\omega,s,t) + h(t)/M][\mathcal{L}_0 A_S(\omega)]^2\rangle}{\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle}. \tag{41}$$

For the potential memory term in Eq. (38) we choose the modified equivalent form

$$\Gamma_A(s,t) = \frac{\beta h(s)\langle[F(\omega,s,t) + h(t)/M][A_S(\omega) - \langle A_S\rangle]\mathcal{L}_0 A_S(\omega)\rangle}{\langle[A_S(\omega) - \langle A_S\rangle]^2\rangle}. \tag{42}$$

From the GLE (31) we see that $F(\omega,s,t) + h(t)/M$ is linear in $A(\omega,\cdot) - \langle A_S\rangle$ defined with a projection time given by $t_0 = s$, we conclude from Eqs. (41) and (42) that $\Gamma_2(s,t)$ and $\Gamma_A(s,t)$ are proportional to correlation functions that are cubic in $A(\omega,\cdot) - \langle A_S\rangle$. In other words, for an observable $A(\omega,t) - \langle A_S\rangle$ that is Gaussian or, more generally, inversion-symmetric, the kernel functions $\Gamma_2(s,t)$ and $\Gamma_A(s,t)$ [and also the potential stiffness correction $K_1(t)$] vanish. In this case, we are thus led to the simplified GLE, valid for Gaussian observables,

$$\ddot{A}(\omega,t) = -K_0[A(\omega,t) - \langle A_S\rangle] - \int_{t_0}^{t} ds\, \Gamma_G(s,t)\dot{A}(\omega,s) + F(\omega,t_0,t) + h(t)/M, \tag{43}$$

where the Gaussian friction kernel is given by the autocorrelation of the sum of the complementary and external forces according to

$$\begin{aligned}\Gamma_G(s,t) &= \frac{\langle[F(\omega,s,s) + h(s)/M][F(\omega,s,t) + h(t)/M]\rangle}{\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle} \\ &= \frac{\langle F(\omega,s,s)F(\omega,s,t)\rangle + h(t)h(s)/M^2}{\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle} \\ &= \frac{\langle F(\omega,s,s)F(\omega,s,t)\rangle}{\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle} + \beta h(t)h(s)/M.\end{aligned} \tag{44}$$

In the derivation of Eq. (44) we used that $h(t)$ is phase-space independent. Thus, we conclude that a Gaussian nonequilibrium observable is described by a GLE with a friction memory kernel that via Eq. (44) is related to the autocorrelation of the total force $F(\omega,s,t) + h(t)$ acting on the observable. One notes that Eqs. (43) and (44) are equivalent to the standard Mori GLE formulation, Eqs. (1) and (2), provided the force in Eq. (1) is interpreted as the sum of the complementary force $F(\omega, s, t)$ and the external time-dependent force $h(t)$. It should be also noted that an observable can be Gaussian while the entire many-body system is non-Gaussian, meaning that other observables and system coordinates may very well exhibit non-Gaussian fluctuations. Thus, the class of systems exhibiting Gaussian observables is a rather large one and includes, for example, the center-of-mass motion of living cells [17,18].

Another useful equation is derived by averaging the entire GLE (31) over phase space, resulting in

$$\begin{aligned}\ddot{a}(t) &= \int d\omega\, \rho_p(\omega)\ddot{A}(\omega, t)\\ &= -K(t)[a(t) - \langle A_S\rangle] - \int_{t_0}^{t} ds\, \Gamma(s, t)\dot{a}(s)\\ &\quad + \int_{t_0}^{t} ds\, \Gamma_A(s, t)[a(s) - \langle A_S\rangle] + h(t)/M, \end{aligned} \tag{45}$$

where the phase-space averaged Heisenberg observable is denoted as $a(t) = \langle A(\omega, t)\rangle$ and $\langle F(\omega, t_0, t)\rangle = 0$ was used. This equation describes how $a(t)$ evolves in time under the influence of $h(t)$, and it therefore establishes the relation between the mean observable $a(t)$ and the external force $h(t)$. This equation is exact and valid beyond the linear-response approximation and can therefore be viewed as a generalization of the linear-response FDT, as will be explored in detail in Sec. IV B.

To derive yet another GLE, we define the deviation of the Heisenberg observable $A(\omega, t)$ around its mean as

$$\Delta A(\omega, t) = A(\omega, t) - a(t), \tag{46}$$

and by subtracting Eqs. (31) and (45) the GLE for $\Delta A(\omega, t)$ follows as

$$\begin{aligned}\Delta\ddot{A}(\omega, t) = &-K(t)\Delta A(\omega, t) - \int_{t_0}^{t} ds\, \Gamma(s, t)\Delta\dot{A}(\omega, s)\\ &+ \int_{t_0}^{t} ds\, \Gamma_A(s, t)\Delta A(\omega, s) + F(\omega, t_0, t), \end{aligned} \tag{47}$$

which does not explicitly depend on the external force $h(t)$ anymore. From Eq. (47) we see that the complementary force $F(\omega, t_0, t)$ is linear in $\Delta A(\omega, \cdot)$. Similar to our argumentation for $A(\omega, t)$, this means that if $\Delta A(\omega, t)$ is a Gaussian variable, the kernel functions $\Gamma_1(s, t)$ and $\Gamma_A(s, t)$ in Eqs. (37) and (38) vanish, and we are thus led to the simplified GLE for the Gaussian deviatory nonequilibrium variable $\Delta A(\omega, t)$

$$\begin{aligned}\Delta\ddot{A}(\omega, t) = &-K_0\Delta A(\omega, t) - \int_{t_0}^{t} ds\, \Gamma_0(s, t)\Delta\dot{A}(\omega, s)\\ &+ F(\omega, t_0, t), \end{aligned} \tag{48}$$

where the friction kernel $\Gamma_0(s, t)$ is given by the complementary force autocorrelation via Eq. (36). Thus, the deviations of a Gaussian nonequilibrium variable from its mean are described by a GLE of the form of Eq. (1) that satisfies the equilibrium relation (2).

We have demonstrated in this section that a Gaussian observable is described by a GLE that has the same form as the equilibrium GLE (1), in other words, the stochastic behavior of an observable of a nonequilibrium system differs from an equilibrium system only if the observable is non-Gaussian. This is a very important finding, since many experimental observables are Gaussian to a very good degree, and for all such observables the standard equilibrium Mori GLE in the form of Eq. (1), or, more precisely, Eqs. (43) or (48), is a valid description of the dynamics.

IV. STOCHASTIC EXTERNAL FORCE

A. Linear response function for finite (not necessarily small) external force $h(t)$

The GLE discussed so far is difficult to deal with in practice since it is inhomogeneous in time; this is utterly expected and reflects the presence of the time-dependent external force $h(t)$ in the Hamiltonian but complicates all further analysis. In many experimental scenarios, the time evolution of $h(t)$ is not known or unimportant and one is rather interested in the observable dynamics that is averaged over $h(t)$. It therefore becomes useful to interpret $h(t)$ as a stochastic variable that can be characterized by its first moments

$$\overline{1} = 1, \quad \overline{h(t)} = 0, \quad \overline{h(t)h(s)} = \sigma(t - s). \tag{49}$$

The assumption of a vanishing first moment does not restrict the generality of the model since (at least for bounded systems) we can subtract a constant from $h(t)$ and move it into the equilibrium part of the Hamiltonian in Eq. (3). Here $\sigma(t - s)$ denotes the autocorrelation or time-dependent variance of the external force; by defining only the first two moments of $h(t)$ we are not necessarily implying that the force actually is a Gaussian stochastic variable. Rather, in the explicit results we neglect the influence of higher-order correlations in $h(t)$, as will become clearer later. By averaging the kernel functions $K(t)$, $\Gamma(s, t)$, $\Gamma_A(s, t)$ in the nonequilibrium GLE Eq. (31) over $h(t)$ according to Eq. (49), we obtain the preaveraged GLE

$$\begin{aligned}\ddot{A}(\omega, t) = &-K_0[A(\omega, t) - \langle A_S\rangle] - \int_{t_0}^{t} ds\, \overline{\Gamma}(t - s)\dot{A}(\omega, s)\\ &+ \int_{t_0}^{t} ds\, \overline{\Gamma}_A(t - s)[A(\omega, s) - \langle A_S\rangle]\\ &+ F(\omega, t_0, t) + F_\epsilon(t) + h(t)/M, \end{aligned} \tag{50}$$

where we have added a generating force $F_\epsilon(t)$ by the substitution $h(t) \to h(t) + MF_\epsilon(t)$ in the Hamiltonian (3). This generating force $F_\epsilon(t)$ is independent of $h(t)$ and will be used to derive the linear response of the nonequilibrium system. Neglecting their implicit dependence on $F_\epsilon(t)$, permitted to linear order in $F_\epsilon(t)$, the force-averaged memory kernels $\overline{\Gamma}(t - s)$ and $\overline{\Gamma}_A(t - s)$ are homogeneous in time, as is shown by perturbative operator expansion to leading order in powers of the external force variance σ in Appendix E. Note that the solution $A(\omega, t)$ of the original GLE (31) is in general not the same as the solution of the preaveraged GLE in Eq. (50), which reflects the approximate nature of the preaveraging approximation. The formal derivation of the preaveraging approximation is given in Appendix F, where we show that it corresponds to the leading term in a systematic expansion in powers of the external-force autocorrelation $\sigma(t)$.

The friction kernel follows from Eq. (40) explicitly as

$$\overline{\Gamma}(t-s) = \overline{\Gamma}_0(t-s) + \beta\sigma(t-s)/M + \overline{\Gamma}_2(t-s), \quad (51)$$

where $\overline{\Gamma}_0(t-s)$ and $\overline{\Gamma}_2(t-s)$ denote the external-force averages over Eqs. (36) and (41).

By averaging Eq. (50) over the phase space variable ω and the external field $h(t)$, we obtain the GLE for the phase-space and external-force-averaged Heisenberg observable $\overline{a}(t)$ on the preaveraging approximation level,

$$\ddot{\overline{a}}(t) = -K_0[\overline{a}(t) - \langle A_S\rangle] - \int_{t_0}^{t} ds\,\overline{\Gamma}(t-s)\dot{\overline{a}}(s) + \int_{t_0}^{t} ds\,\overline{\Gamma}_A(t-s)[\overline{a}(s) - \langle A_S\rangle] + F_\epsilon(t). \quad (52)$$

By Fourier transforming Eq. (52) according to $\tilde{\overline{a}}(\nu) = \int_{-\infty}^{\infty} dt e^{-\imath t\nu}\overline{a}(t)$, we obtain the response relation to first order in $F_\epsilon(t)$ as

$$\tilde{\overline{a}}(\nu) - 2\pi\delta(\nu)\langle A_S\rangle = \tilde{\chi}(\nu)\tilde{F}_\epsilon(\nu), \quad (53)$$

where the Fourier-transformed response function is determined by

$$\begin{aligned} 1/\tilde{\chi}(\nu) &= K_0 - \nu^2 + \imath\nu\tilde{\overline{\Gamma}}^+(\nu) - \tilde{\overline{\Gamma}}_A^+(\nu) \\ &= K_0 - \nu^2 + \imath\nu(\tilde{\overline{\Gamma}}_0^+(\nu) + \beta\tilde{\sigma}^+(\nu)/M + \tilde{\overline{\Gamma}}_2^+(\nu)) \\ &\quad - \tilde{\overline{\Gamma}}_A^+(\nu). \end{aligned} \quad (54)$$

In the last step we have inserted Eq. (51). Note that the response function $\tilde{\chi}(\nu)$ accounts for the full nonlinear dependence on the nonequilibrium force $h(t)$ on the preaveraging approximation level, so it describes the linear response to an infinitesimal force $F_\epsilon(t)$ in the presence of a finite (not necessarily small) time-dependent external force $h(t)$. In deriving Eq. (54) we have shifted the projection time into the far past, $t_0 \to -\infty$, and we have introduced causal or single-sided memory kernels and correlations, i.e., $\overline{\Gamma}^+(t) = 0$, $\overline{\Gamma}_A^+(t) = 0$, $\overline{\Gamma}_0^+(t) = 0$, $\overline{\Gamma}_2^+(t) = 0$, $\sigma^+(t) = 0$ for $t < 0$. In fact, the nonequilibrium response function $\tilde{\chi}(\nu)$ can be extracted from experiments or simulations by applying a small perturbing force $F_\epsilon(t)$; alternatively, it can be extracted from an observable trajectory via a suitably defined two-point correlation function, as explained in Sec. IV C. For a Gaussian observable, in which case $\overline{\Gamma}_A(t) = 0 = \overline{\Gamma}_2(t)$ as discussed in Sec. III B, but in the presence of a nonequilibrium stochastic force, $h(t) \neq 0$, we obtain from Eq. (54) for the response function

$$1/\tilde{\chi}(\nu) = K_0 - \nu^2 + \imath\nu\tilde{\overline{\Gamma}}_0^+(\nu) + \imath\nu\beta\tilde{\sigma}_+(\nu)/M. \quad (55)$$

In the absence of an external force, which means for $\tilde{\sigma}_+(\nu) = 0$, Eq. (54) reduces to the standard equilibrium response function

$$1/\tilde{\chi}(\nu) = K_0 - \nu^2 + \imath\nu\tilde{\overline{\Gamma}}_0^+(\nu). \quad (56)$$

By comparison of Eqs. (54)–(56) we see that the presence of a stochastic external force $h(t)$ modifies the response function $\tilde{\chi}(\nu)$ significantly. Thus, for an experimental or simulation system where one is able to turn on and off the external force $h(t)$, one can determine the nonequilibrium contribution to the response function (54) $\imath\nu(\beta\tilde{\sigma}^+(\nu)/M + \tilde{\overline{\Gamma}}_2^+(\nu)) - \tilde{\overline{\Gamma}}_A^+(\nu)$ by comparison of the responses with and without external force, given by Eqs. (54) and (56). For a Gaussian observable, Eq. (55) shows that one can directly obtain the external force autocorrelation function $\sigma(t)$ by comparing the response functions for the scenarios where the external force $h(t)$ is turned on and off.

B. Nonequilibrium fluctuation-dissipation theorem for finite external force $h(t)$

In order to derive the nonequilibrium version of the FDT, we need to calculate the two-point correlation function

$$C(t-t') = \overline{\langle[A(\omega,t) - \langle A_S\rangle][A(\omega,t') - \langle A_S\rangle]\rangle} \quad (57)$$

for general times $t, t' \gg t_0$, which is obtained by simultaneously averaging over phase space and the nonequilibrium force $h(t)$ according to Eq. (49). In Appendix G we derive Eq. (57) and show that two-point correlation functions are generally given by a phase-space average over products of Heisenberg variables; for this we use the product propagation relation derived in Appendix H. In Appendix I we show that the Fourier-transformed correlation function is to first order in an expansion in powers of $\tilde{\overline{\Gamma}}_0(\nu)$ and $\tilde{\sigma}(\nu)$, the second-order moments of the complementary force $F(\omega, t_0, t)$ and the external force $h(t)$, respectively, given by

$$\tilde{C}(\nu) = \tilde{\chi}(\nu)\tilde{\chi}(-\nu)[\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle\tilde{\overline{\Gamma}}_0(\nu) + \tilde{\sigma}(\nu)/M^2], \quad (58)$$

where we note that $\tilde{\overline{\Gamma}}_0(\nu) = \tilde{\overline{\Gamma}}_0^+(\nu) + \tilde{\overline{\Gamma}}_0^+(-\nu)$ and $\tilde{\sigma}(\nu) = \tilde{\sigma}^+(\nu) + \tilde{\sigma}^+(-\nu)$ are Fourier transforms of the time-symmetrized functions. Using the expression for the response function $\tilde{\chi}(\nu)$ in Eq. (54), we can rewrite Eq. (58) as

$$\frac{\tilde{C}(\nu)}{\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle} = \frac{\tilde{\chi}(-\nu)}{\imath\nu} - \frac{\tilde{\chi}(\nu)}{\imath\nu} - \tilde{\chi}(\nu)\tilde{\chi}(-\nu)\left[\tilde{\overline{\Gamma}}_2^+(\nu) + \tilde{\overline{\Gamma}}_2^+(-\nu) - \frac{\tilde{\overline{\Gamma}}_A^+(\nu) - \tilde{\overline{\Gamma}}_A^+(-\nu)}{\imath\nu}\right]. \quad (59)$$

Since all time-domain kernel functions are real, we have $\mathrm{Re}(\tilde{\Gamma}^+(\nu)) = \mathrm{Re}(\tilde{\Gamma}^+(-\nu))$ and $\mathrm{Im}(\tilde{\Gamma}^+(\nu)) = -\mathrm{Im}(\tilde{\Gamma}^+(-\nu))$, where Re and Im denote the real and imaginary parts, respectively, of a complex number X according to $X = \mathrm{Re}X + \imath\mathrm{Im}X$. With this, we can rewrite Eq. (59) as

$$\frac{\nu\tilde{C}(\nu)}{2\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle} = -\mathrm{Im}(\tilde{\chi}(\nu)) - \tilde{\chi}(\nu)\tilde{\chi}(-\nu)[\nu\mathrm{Re}(\tilde{\overline{\Gamma}}_2^+(\nu)) - \mathrm{Im}(\tilde{\overline{\Gamma}}_A^+(\nu))], \quad (60)$$

which is the generalized FDT in the presence of a finite (not necessarily small) stochastic external force $h(t)$. As discussed in Sec. III B, if the observable $A(\omega, t)$ is Gaussian, the memory kernel contributions $\tilde{\bar{\Gamma}}_2(\nu)$ and $\tilde{\bar{\Gamma}}_A(\nu)$ vanish and we recover a relation that resembles the standard FDT [8]

$$\frac{\nu\tilde{C}(\nu)}{2\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle} = -\mathrm{Im}(\tilde{\chi}(\nu)). \tag{61}$$

However, the correlation function $\tilde{C}(\nu)$ and the response function $\tilde{\chi}(\nu)$ are modified by the presence of the external force $h(t)$ and do depend on the external-force autocorrelation $\sigma(t)$, as explicitly shown in Eqs. (54) and (58), so Eq. (61) is a nontrivial generalization of the standard FDT [obtained in the limit of infinitesimally small external force $h(t)$] to nonequilibrium Gaussian systems with finite (not necessarily small) $h(t)$. Recently, a FDT similar to Eq. (61) was derived for a specific class of dynamic perturbations introduced on the level of the Liouville equation [38]. The relation to our result based on a Hamiltonian model will be explored in the future.

In Appendix J we derive the standard FDT by leading-order perturbation analysis of the Heisenberg observable in Eq. (13). The comparison with that derivation is instructive, since it deviates slightly from the textbook derivation of the FDT and highlights the nonperturbative character of our nonequilibrium GLE. Comparison of Eqs. (60) and (61) shows that our generalized FDT for non-Gaussian nonequilibrium systems in Eq. (60) contains additional terms that are proportional to the non-Gaussian memory kernel contributions $\tilde{\bar{\Gamma}}_2(\nu)$ and $\tilde{\bar{\Gamma}}_A(\nu)$.

An alternative and particularly transparent formulation of the nonequilibrium fluctuation dissipation theorem (60) is given by

$$-\frac{\nu\tilde{C}(\nu)/\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle}{2\mathrm{Im}(\tilde{\chi}(\nu))} = \frac{\tilde{\bar{\Gamma}}_0(\nu) + \beta\tilde{\sigma}(\nu)/M}{2(\mathrm{Re}(\tilde{\bar{\Gamma}}^+(\nu)) - \mathrm{Im}(\tilde{\bar{\Gamma}}_A^+(\nu))/\nu)} \equiv 1 + \Xi(\nu), \tag{62}$$

where in the first equation we used Eq. (58) and the relation

$$\mathrm{Im}(\tilde{\chi}(\nu)) = -\tilde{\chi}(\nu)\tilde{\chi}(-\nu)(\nu\mathrm{Re}(\tilde{\bar{\Gamma}}^+(\nu)) - \mathrm{Im}(\tilde{\bar{\Gamma}}_A^+(\nu))), \tag{63}$$

which directly follows from Eq. (54). Equation (62) contains two alternative forms of our nonequilibrium FDT, the first in terms of the correlation function $\tilde{C}(\nu)$ and the imaginary part of the response function $\mathrm{Im}(\tilde{\chi}(\nu))$, which is preferred when the external-force autocorrelation $\sigma(t)$ is not known, the second in terms of the complementary and external force correlations $\tilde{\bar{\Gamma}}_0(\nu)$, $\tilde{\sigma}(\nu)$ and the memory functions $\tilde{\bar{\Gamma}}^+(\nu)$ and $\tilde{\bar{\Gamma}}_A^+(\nu)$, which is useful when $\sigma(t)$ is known, as will be elaborated on in Sec. IV D. In Eq. (62) we defined the frequency-dependent nonequilibrium correction to the standard FDT, $\Xi(\nu)$, which has been previously introduced to quantify the departure from equilibrium in biological nonequilibrium data [37,39]. Using Eq. (60), it is given by

$$\Xi(\nu) = \frac{\tilde{\chi}(\nu)\tilde{\chi}(-\nu)}{\mathrm{Im}(\tilde{\chi}(\nu))}[\mathrm{Re}(\nu\tilde{\bar{\Gamma}}_2^+(\nu)) - \mathrm{Im}(\tilde{\bar{\Gamma}}_A^+(\nu))]. \tag{64}$$

Obviously, for Gaussian observables, for which $\tilde{\bar{\Gamma}}_2^+(\nu) = 0 = \tilde{\bar{\Gamma}}_A^+(\nu)$ holds, as has been explained in Sec. III B, we have $\Xi(\nu) = 0$. The expression for $\Xi(\nu)$ in Eq. (64) factorizes into $\tilde{\chi}(\nu)\tilde{\chi}(-\nu)/\mathrm{Im}(\tilde{\chi}(\nu))$, which depends solely on the response function $\chi(\nu)$, and into the sum of the non-Gaussian memory contributions $\tilde{\bar{\Gamma}}_2^+(\nu)$ and $\tilde{\bar{\Gamma}}_A^+(\nu)$. It transpires that from the response function $\tilde{\chi}(\nu)$ and the two-point correlation function $\tilde{C}(\nu)$, which have been determined experimentally for a few different nonequilibrium biological systems [9,16] and from which via Eq. (62) the correction factor $\Xi(\nu)$ can be determined, the sum of the non-Gaussian memory contributions $\tilde{\bar{\Gamma}}_2^+(\nu)$ and $\tilde{\bar{\Gamma}}_A^+(\nu)$ can be straightforwardly extracted via Eq. (64). That means that the non-Gaussian character of an observable can be inferred without actually measuring its distribution. We note that the rescaled sum of the complementary force autocorrelation $\tilde{\bar{\Gamma}}_0(\nu)$ and the external force autocorrelation $\tilde{\sigma}(\nu)$ can also be extracted with knowledge of the response function $\tilde{\chi}(\nu)$ and the two-point correlation function $\tilde{C}(\nu)$ according to Eq. (58).

C. Response function from Volterra equation

The standard way of extracting the memory kernel from time-series data is by turning the stochastic GLE for the phase-space dependent observable $A(\omega, t)$ into a nonstochastic integro-differential equation for the two-point correlation function, which can be solved by Fourier transformation or recursively after discretization in the time domain [45,89]. Here we show that the same methodology also works for our nonequilibrium GLE. To proceed, we multiply the preaveraged GLE in Eq. (50) by $\dot{A}(\omega, t_0) = \mathcal{L}_0 A_S(\omega)$ and average over phase space ω and the external force $h(t)$ according to Eq. (49), by which we obtain the equation

$$-\dddot{C}_0(t-t_0) = K_0\dot{C}_0(t-t_0) + \int_0^{t-t_0} ds\bar{\Gamma}(s)\ddot{C}_0(t-t_0-s) - \int_0^{t-t_0} ds\bar{\Gamma}_A(s)\dot{C}_0(t-t_0-s) \tag{65}$$

for the phase-space and external-force averaged two point correlation function

$$C_0(t-t_0) = \overline{\langle[A(\omega,t_0) - \langle A_S\rangle][A(\omega,t) - \langle A_S\rangle]\rangle}, \tag{66}$$

on the preaveraging approximation level. Note that the two-point correlation function $C_0(t)$ defined here differs from the one defined in Eq. (57) in that one of the times coincides with the projection time t_0. This makes a fundamental difference, as will become clear shortly; for example, $C_0(0)$ is an equilibrium average that depends only on the canonical projection distribution $\rho_p(\omega)$ in Eq. (29), while $C(0)$ is an intrinsically nonequilibrium expectation value. When deriving Eq. (65), we used that the phase-space average over the product of $\dot{A}(\omega, t_0)$ and the forces $F(\omega, t_0, t)$ or $h(t)$ in the GLE (31) vanishes and that the resulting equation becomes homogeneous in time due to the average over the stochastic force $h(t)$, as explained in Appendix E.

Performing a single-sided Fourier transform of Eq. (65) while assuming $t \geqslant t_0$ and defining $\tilde{C}_0^+(\nu) = \int_0^\infty dt e^{-\imath t\nu} C_0(t)$, we obtain

$$\tilde{C}_0^+(\nu) = \frac{\ddot{C}_0(0)\tilde{\chi}(\nu)}{\imath\nu} + \frac{C_0(0)}{\imath\nu}, \tag{67}$$

where we used the response function $\tilde{\chi}(\nu)$ defined in Eq. (54). Symmetrizing Eq. (67) according to $\tilde{C}_0(\nu) = \tilde{C}_0^+(\nu) + \tilde{C}_0^+(-\nu)$ and using that $\ddot{C}_0(0) = \langle[\mathcal{L}_0 A_S(\omega)]^2\rangle$, it follows that $\tilde{C}_0(\nu)$ equals $\tilde{C}(\nu)$ in the Gaussian scenario, Eq. (61), but differs from $\tilde{C}(\nu)$ in the non-Gaussian scenario, Eq. (60); in other words, $C_0(t)$ and $C(t)$ differ only for non-Gaussian nonequilibrium systems.

From the time-domain correlation function $C_0(t)$ defined in Eq. (66), which can be straightforwardly obtained in experiments or simulations by turning on the nonequilibrium force at time t_0 [using that the distribution at time t_0, just prior to application of the nonequilibrium force, equals the canonical projection distribution $\rho_p(\omega)$ in Eq. (29)], the single-sided Fourier transform $\tilde{C}_0^+(\nu)$ and the values $\ddot{C}_0(0)$ and $C_0(0)$ follow, from which $\tilde{\chi}(\nu)$ is determined via Eq. (67) by direct inversion according to

$$\tilde{\chi}(\nu) = \frac{\imath\nu\tilde{C}_0^+(\nu)}{\ddot{C}_0(0)} - \frac{C_0(0)}{\ddot{C}_0(0)}. \tag{68}$$

Alternatively, instead of Fourier transformation, Eq. (65) can be recursively solved by discretization [45,52,84].

D. Extracting nonequilibrium GLE parameters from the response function and time-series data

In the previous section we showed that the correlation function $C_0(t)$, defined in Eq. (66), is the solution of the differential equation (65) and can be used to determine the response function $\tilde{\chi}(\nu)$ via Eq. (68). This is an alternative and rather straightforward method to obtain the response function $\tilde{\chi}(\nu)$ from the experimentally determined or simulated correlation function $C_0(t)$, the standard way being the application of a small perturbing force and measuring the response, as discussed in Sec. IV A.

In fact, from $\tilde{\chi}(\nu)$ the deterministic force and kernel parameters of the preaveraged GLE in Eq. (50) can be obtained. To show this, we introduce the running integral over the preaveraged memory function $\overline{G}_A(s) = \int_s^{t-t_0} ds'\, \overline{\Gamma}_A(s')$, with which Eq. (50) can after partial integration be rewritten as

$$\begin{aligned}\ddot{A}(\omega, t) = &-[K_0 - \overline{G}_A(0)][A(\omega, t) - \langle A_S\rangle] \\ &- \int_{t_0}^{t} ds\,[(\overline{\Gamma}(t-s) + \overline{G}_A(t-s)]\dot{A}(\omega, s) \\ &+ F(\omega, t_0, t) + h(t)/M. \end{aligned}\tag{69}$$

The GLE in Eq. (69) now depends on a single combined kernel function $\overline{\Gamma}(t) + \overline{G}_A(t)$. Shifting the projection time into the far past, $t_0 \to -\infty$, the single-sided Fourier transform of this kernel function can be shown to be related to the Fourier-transformed response function $\tilde{\chi}(\nu)$ given in Eq. (54) according to

$$\tilde{\overline{\Gamma}}^+(\nu) + \tilde{\overline{G}}_A^+(\nu) = \frac{1}{\imath\nu}\left(\frac{1}{\tilde{\chi}(\nu)} - \frac{1}{\tilde{\chi}(0)} + \nu^2\right). \tag{70}$$

The stiffness of the effective harmonic potential that appears in Eq. (69) is determined by the zero-frequency limit of the response function in Eq. (54) according to

$$K_0 - \overline{G}_A(0) = \frac{1}{\tilde{\chi}(0)}. \tag{71}$$

It transpires that all deterministic parameters of the GLE in Eq. (69) can be derived from the response function $\tilde{\chi}(\nu)$ according to Eqs. (70) and (71). All results derived in Secs. IV A, IV B, and IV C are equally valid for the GLE (69) if the replacement $\tilde{\overline{\Gamma}}_A^+(\nu) = \overline{G}_A^+(0) - \imath\nu\tilde{\overline{G}}_A^+(\nu)$ is used.

Furthermore, using these deterministic GLE parameters obtained from Eqs. (70) and (71), the trajectory of the sum of the complementary and external forces, $F(\omega, t_0, t) + h(t)/M$, can be calculated from a trajectory of the observable $A(\omega, t)$ via Eq. (69). This allows us to calculate the distribution and all correlations of $F(\omega, t_0, t) + h(t)/M$ and thus to obtain the complete parametrization of the GLE. In particular, after averaging over ω and $h(t)$, which amounts to an average over time, and using the definitions (36) and (39), the autocorrelation function of $F(\omega, t_0, t) + h(t)/M$ becomes proportional to the sum $\tilde{\overline{\Gamma}}_0(\nu) + \beta\tilde{\sigma}(\nu)/M$. If the external-force trajectory $h(t)$ is known, which is the case for simulations and some experimental scenarios, even the complementary force trajectory can be extracted.

In conclusion, knowledge of the response function $\tilde{\chi}(\nu)$, which can be obtained from the correlation function $C_0(t)$ defined in Eq. (66) according to Eq. (68) or from applying a perturbation force as discussed in Sec. IV A, allows us to determine the deterministic parameters of the GLE in Eq. (69). The force trajectory $F(\omega, t_0, t) + h(t)/M$ can then be determined from the observable trajectory by use of the GLE Eq. (69), which means that all parameters of the GLE are known. Thus, the exact coarse-grained model defined by the GLE can be fully parameterized based on nonequilibrium time-series data.

E. Joint observable distribution from path integrals

It remains to elucidate under which conditions the Heisenberg variable $A(\omega, t)$ can be described as a Gaussian process and thus the simplified nonequilibrium GLE (43) is valid. For this we consider the two-point joint probability distribution of $A(\omega, t)$, which is defined as

$$\rho(A_2, t_2; A_1, t_1) = \overline{\langle\delta(A_2 - A(\omega, t_2))\delta(A_1 - A(\omega, t_1))\rangle} \tag{72}$$

and involves averages over phase space ω and the external force $h(t)$ according to Eqs. (28) and (49). For the delta functions we use their Fourier representation and obtain

$$\begin{aligned}\rho(A_2, t_2; A_1, t_1) &= \int_{-\infty}^{\infty}\frac{dq_1}{2\pi}\int_{-\infty}^{\infty}\frac{dq_2}{2\pi} \\ &\times e^{\imath q_1 A_1 + \imath q_2 A_2}\overline{\langle\exp[-\imath q_1 A(\omega, t_1) - \imath q_2 A(\omega, t_2)]\rangle}. \end{aligned}\tag{73}$$

Using the time-domain version $\chi(t)$ of the response function defined in Eq. (54), the preaveraged GLE (50) can be inverted as

$$A(\omega, t) - \langle A_S\rangle = \int_{-\infty}^{\infty} ds\,\chi(s)[\overline{F}(\omega, t-s) + h(t-s)/M]. \tag{74}$$

Note that in deriving Eq. (74) we have also preaveraged the complementary force term in Eq. (50) over $h(t)$, so that it becomes time-homogeneous as well. This additional preaveraging leads to higher-order corrections in terms of the external-force autocorrelation $\sigma(t)$ that can be neglected to leading order, as explained in Appendix F. It transpires from Eq. (74) that the dependence of $A(\omega,t)$ on the phase space variable ω is solely due to the preaveraged complementary force $\overline{F}(\omega,t)$, so the phase-space average in Eq. (73) can be replaced by an average over $\overline{F}(\omega,t)$. The averages over the forces $\overline{F}(\omega,t)$ and $h(t)$ in Eq. (73) we express by Gaussian path integrals as

$$\langle X(\overline{F},h)\rangle = \int_{-\infty}^{\infty}\frac{\mathcal{D}\overline{F}(\omega,\cdot)}{\mathcal{N}_F}X(\overline{F},h)\exp\left(-\int_{-\infty}^{\infty}ds\,ds'\frac{\overline{F}(\omega,s)\overline{F}(\omega,s')\overline{\Gamma}_0^{-1}(s-s')}{2\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}\right), \tag{75}$$

$$\overline{X(\overline{F},h)} = \int_{-\infty}^{\infty}\frac{\mathcal{D}h(\cdot)}{\mathcal{N}_h}X(\overline{F},h)\exp\left(-\int_{-\infty}^{\infty}ds\,ds'\frac{h(s)h(s')\sigma^{-1}(s-s')}{2}\right), \tag{76}$$

where $\mathcal{N}_F$ and $\mathcal{N}_h$ are normalization constants and $\overline{\Gamma}_0^{-1}(s)$ and $\sigma^{-1}(s)$ are the inverse functions of the Gaussian kernels defined in Eqs. (36) and (49) according to $\int ds\overline{\Gamma}_0^{-1}(t-s)\overline{\Gamma}_0(s-t') = \delta(t'-t)$ [and similarly for $\sigma^{-1}(s)$]. Non-Gaussian fluctuations of $\overline{F}(\omega,t)$ and $h(t)$ need not be explicitly considered here as by virtue of Eqs. (36) and (49) they do not change their two-point correlations. Performing the Gaussian path integrals, the two-point distribution follows in matrix notation as

$$\rho(A_2,t_2;A_1,t_1) = \frac{\exp\left[-(A_j-\langle A_S\rangle)I_{jk}^{-1}(A_k-\langle A_S\rangle)/2\right]}{\sqrt{\det 2\pi I}}, \tag{77}$$

where the indices $j,k=1,2$ are summed over and the entries of the two-by-two matrix

$$I_{jk} = C(t_j-t_k) \tag{78}$$

are given by the two-point correlation function defined in Eq. (58); details of the derivation are given in Appendix K. From Eq. (77) we see that if the forces $\overline{F}(\omega,t)$ and $h(t)$ are described by general Gaussian processes, as assumed in Eqs. (75) and (76), then, on the preaveraging approximation level, also the observable $A(\omega,t)-\langle A_S\rangle$ is a Gaussian process determined by the correlation function $C(t)$ defined in Eq. (58). In this case, the GLE (43) is valid, which has the structure of an equilibrium GLE. Conversely, if one of the two forces $\overline{F}(\omega,t)$ or $h(t)$ is non-Gaussian, then also the observable $A(\omega,t)-\langle A_S\rangle$ is non-Gaussian and the GLE in Eq. (31) applies, which does not satisfy the simple relation between the memory kernel and the force autocorrelation in Eq. (44). To reiterate this point, if the relation in Eq. (44) is violated, this can be due to non-Gaussian contributions in the complementary force $\overline{F}(\omega,t)$ or in the nonequilibrium force $h(t)$. It will in general be difficult to tell in an experiment which of the forces generates the non-Gaussian behavior of the observable $A(\omega,t)$ [unless of course the nonequilibrium force $h(t)$ is generated by hand and explicitly known], in particular since $\overline{F}(\omega,t)$ is modified by the presence of an external force $h(t)$.

V. SUMMARY AND DISCUSSION

In this paper we have derived the nonequilibrium GLE from a many-body Hamiltonian which contains a time-dependent force $h(t)$ that acts on a general phase-space-dependent observable $A_S(\omega)$. This is the same Hamiltonian from which the standard FDT is derived; in other words, the GLE we derive is conjugate to the standard FDT with one important distinction: While the standard equilibrium FDT describes the first-order response of the time-dependent mean of the observable $A_S(\omega)$ to an infinitesimally small external force $h(t)$, we derive the nonequilibrium GLE Eq. (31) nonperturbatively, i.e., exactly to all orders in $h(t)$. While linear and nonlinear response theory describes how the mean of an observable explicitly depends on the external force $h(t)$, the GLE is an equation of motion for the fluctuating (i.e., phase-space dependent) observable, it therefore opens up a complementary field of applications and is particularly relevant for the description of the stochasticity of time-series data.

From the exact GLE Eq. (31) we infer that Gaussian nonequilibrium observables are described by the simplified GLE in Eq. (43) that takes the form of the equilibrium GLE in Eq. (1). This is an important and rather nontrivial finding, as key observables of many nonequilibrium systems are in fact Gaussian. For example, the motion of cancer cells and algae [17,18] has been shown to be described by a Gaussian process provided one looks at single-cell data. The correction terms in the nonequilibrium GLE (31) that account for non-Gaussian effects turn out to be equivalent to three-point correlation functions of the observable.

The nonequilibrium GLE in Eq. (31) breaks time homogeneity and thus is difficult to deal with in practice. Treating the external force $h(t)$ as a stochastic variable that is defined by its second moment, we preaverage the GLE over $h(t)$. This preaveraging approximation reinstalls time homogeneity of the GLE and allows us to derive the nonequilibrium FDT to first order in an expansion in terms of the external and complementary force variances. Similar to the GLE, the nonequilibrium FDT for Gaussian observables has the same form as the equilibrium FDT, only for non-Gaussian observables correction terms appear in the nonequilibrium FDT that again are related to three-point correlation functions of the observable.

We also introduce different methods for extracting the parameters of our nonequilibrium GLE from time-series data. Knowledge of just the two-point correlation function $C(t)$ allows to gain only convoluted insight, as demonstrated by Eq. (58), unless the observable is Gaussian and $C(t)$ contains complete knowledge of the system response via Eq. (61). In contrast, complete parameter extraction from data is possible even for non-Gaussian nonequilibrium systems by knowledge of the response function $\chi(t)$ and the observable trajectory. Here it is important to distinguish two scenarios: If the external force $h(t)$ can be turned on and off at will, as is the case in simulations and in experiments with explicitly applied external force, for example, by laser traps or atomic-force microscopes, $\chi(t)$ can be determined via Eq. (68) from the two-point correlation function $C_0(t)$ defined relative to the time at which $h(t)$ is turned on, as is explained in Sec. IV C. If the external force $h(t)$ cannot be abruptly turned on or off, as is the case in most biological experiments, $\chi(t)$ must be determined by explicitly applying a small additional perturbation force, as explained in Sec. IV A. From $\chi(t)$ and the observable trajectory, all GLE parameters can be extracted, as explained in Sec. IV D. Knowing all GLE parameters, one has a complete coarse-grained theory for the nonequilibrium process, from which, e.g., the nonequilibrium FDT in Eq. (60) can be predicted, as explained in Sec. IV B. For example, the response function and the two-point correlation function have been experimentally measured for active actomyosin networks [9] and living red blood cells [16], meaning that the nonequilibrium kernel functions can be extracted using the formulas given in Sec. IV B from literature experimental data.

It is a widespread misconception that the standard Mori GLE Eq. (1) is approximate and holds only for Gaussian observables. The opposite is true: Eq. (1) is exact and any nonlinear properties that the observable A might have are accurately accounted for by non-Gaussian contributions of the complementary force F. The same is also true for the nonequilibrium GLE we derive, Eq. (31), which exactly describes non-Gaussian nonequilibrium observables, provided the non-Gaussian contributions from the complementary force are correctly included.

A second widespread misconception is that the relation between the memory kernel and the complementary-force autocorrelation for equilibrium systems in Eq. (2) is a consequence of or equivalent to the fluctuation-dissipation theorem (FDT) and that deviations from the equality in Eq. (2) would signal a breakdown of the standard FDT. While this is true for the Mori projection used in this work, it does not hold for other nonlinear projection schemes that have been introduced recently [52,77,78].

One conclusion from our work is that for Gaussian observables that follow from a time-dependent Hamiltonian of the form of Eq. (3), it does not make sense to consider a GLE with a complementary force that violates the relation in Eq. (2). This nicely complements our previous finding that for Gaussian systems there is no way of detecting nonequilibrium properties from time-series data [17,18]. Our approach can also be used to derive coarse-grained nonequilibrium models of interacting active particles [22–25,40] from time-dependent Hamiltonian models, for which one would have to generalize the techniques used to derive the nonequilibrium GLE in this paper to vectorial observables. It would be highly interesting to investigate the structure of the dissipative coupling between the active particles in such a vectorial nonequilibrium GLE and how it relates to the properties of the underlying Hamiltonian model. The time-dependent Hamiltonian Eq. (3) studied by us includes only the linear coupling between the time-dependent force $h(t)$ and the observable $A_S(\omega)$; more complicated time-dependent Hamiltonians are conceivable. The motivation for studying this simple time-dependent Hamiltonian is that it leads to the standard FDT, and our nonequilibrium GLE thus is conjugate to the standard FDT. In the future, it will be interesting to derive GLEs from more complex time-dependent Hamiltonians.

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APPENDIX A: DERIVATIVE OF HEISENBERG PROPAGATOR WITH RESPECT TO INITIAL TIME

Using the Heavyside function $\theta(t)$, defined as $\theta(t) = 1$ for $t > 0$ and $\theta(t) = 0$ for $t < 0$, the time-ordered operator exponential in the Heisenberg picture (14) can be rewritten as

$$\exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right) = 1 + \sum_{n=1}^{\infty}\int_{t_0}^{t} dt_1 \int_{t_0}^{t} dt_2\theta(t_1 - t_2)\int_{t_0}^{t} dt_3\theta(t_2 - t_3)\cdots\int_{t_0}^{t} dt_n\theta(t_{n-1} - t_n)\mathcal{L}(t_n)\cdots\mathcal{L}(t_3)\mathcal{L}(t_2)\mathcal{L}(t_1). \quad \text{(A1)}$$

We now reorder the integration variables to obtain

$$\exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right) = 1 + \sum_{n=1}^{\infty}\int_{t_0}^{t} dt_n \int_{t_0}^{t} dt_{n-1}\theta(t_{n-1} - t_n)\int_{t_0}^{t} dt_{n-2}\theta(t_{n-2} - t_{n-1})\cdots\int_{t_0}^{t} dt_1\theta(t_1 - t_2)\mathcal{L}(t_n)\mathcal{L}(t_{n-1})\mathcal{L}(t_{n-2})\cdots\mathcal{L}(t_1); \quad \text{(A2)}$$

note that the Liouville operators cannot be reordered since $\mathcal{L}(t_1)$ and $\mathcal{L}(t_2)$ in general do not commute for $t_1 \neq t_2$. We now replace the Heavyside functions by the appropriate integration boundaries and obtain

$$\exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right) = 1 + \sum_{n=1}^{\infty}\int_{t_0}^{t} dt_n \int_{t_n}^{t} dt_{n-1}\int_{t_{n-1}}^{t} dt_{n-2}\cdots\int_{t_2}^{t} dt_1\mathcal{L}(t_n)\mathcal{L}(t_{n-1})\mathcal{L}(t_{n-2})\cdots\mathcal{L}(t_1). \quad \text{(A3)}$$

From this expression Eq. (16) follows directly.

APPENDIX B: DERIVATION OF THE TIME-DEPENDENT DYSON OPERATOR EXPANSION

The operator expansion for two general time-dependent operators $\mathcal{V}(t)$ and $\mathcal{W}(t)$ reads

$$\exp_H\left(\int_{t_0}^t \mathcal{V}(s)+\mathcal{W}(s)\,ds\right)=\exp_H\left(\int_{t_0}^t ds\mathcal{V}(s)\right)+\int_{t_0}^t ds\exp_H\left(\int_{t_0}^s ds'\mathcal{V}(s')\right)\mathcal{W}(s)\exp_H\left(\int_s^t \mathcal{V}(s')+\mathcal{W}(s')ds'\right). \quad \text{(B1)}$$

To prove this relation, we use Eqs. (15) and (16) to obtain from Eq. (B1)

$$\exp_H\left(\int_{t_0}^t \mathcal{V}(s)+\mathcal{W}(s)\,ds\right)=\exp_H\left(\int_{t_0}^t ds\mathcal{V}(s)\right)-\int_{t_0}^t ds\frac{d}{ds}\exp_H\left(\int_{t_0}^s ds'\mathcal{V}(s')\right)\exp_H\left(\int_s^t \mathcal{V}(s')+\mathcal{W}(s')ds'\right). \quad \text{(B2)}$$

Now the integral can be performed and the equality is obtained. An alternative operator expansion relation reads

$$\exp_H\left(\int_{t_0}^t \mathcal{V}(s)+\mathcal{W}(s)\,ds\right)=\exp_H\left(\int_{t_0}^t ds\mathcal{V}(s)\right)+\int_{t_0}^t ds\exp_H\left(\int_{t_0}^s \mathcal{V}(s)+\mathcal{W}(s)\,ds'\right)\mathcal{W}(s)\exp_H\left(\int_s^t ds'\mathcal{V}(s')\right), \quad \text{(B3)}$$

which can be proven analogously. Choosing $\mathcal{V}(s)=\mathcal{QL}(s)$ and $\mathcal{W}(s)=\mathcal{PL}(s)$ in Eq. (B3) we obtain Eq. (24).

APPENDIX C: DERIVATION OF ESSENTIAL MORI PROJECTION PROPERTIES

For the following derivations it is useful to split the Mori projection operator in Eq. (27) into three parts according to

$$\mathcal{P}B(\omega,t)=\mathcal{P}_1B(\omega,t)+\mathcal{P}_2B(\omega,t)+\mathcal{P}_3B(\omega,t) \quad \text{(C1)}$$

with

$$\mathcal{P}_1B(\omega,t)=\langle B(\omega,t)\rangle, \quad \text{(C2)}$$

$$\mathcal{P}_2B(\omega,t)=\frac{\langle B(\omega,t)\mathcal{L}_0A_S(\omega)\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}\mathcal{L}_0A_S(\omega), \quad \text{(C3)}$$

$$\mathcal{P}_3B(\omega,t)=\frac{\langle B(\omega,t)[A_S(\omega)-\langle A_S\rangle]\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}(A_S(\omega)-\langle A_S\rangle). \quad \text{(C4)}$$

The linearity of the Mori projection, i.e., the fact that for two arbitrary observables $B(\omega,t)$ and $C(\omega,t')$ the property $\mathcal{P}(c_1B(\omega,t)+c_2C(\omega,t'))=c_1\mathcal{P}B(\omega,t)+c_2\mathcal{P}C(\omega,t')$ holds, is self-evident, and $\mathcal{Q}$ is also easily seen to be linear. The idempotency of $\mathcal{P}$, i.e., the fact that $\mathcal{P}^2=\mathcal{P}$, is not self-evident and will be proven. We split the proof in three parts. First,

$$\mathcal{PP}_1B(\omega,t)=\langle\langle B(\omega,t)\rangle\rangle+\langle B(\omega,t)\rangle\frac{\langle\mathcal{L}_0A_S(\omega)\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}\mathcal{L}_0A_S(\omega)+\langle B(\omega,t)\rangle\frac{\langle[A_S(\omega)-\langle A_S\rangle]\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}[A_S(\omega)-\langle A_S\rangle]=\mathcal{P}_1B(\omega,t). \quad \text{(C5)}$$

For the first and third terms we used that $\langle\langle B(\omega,t)\rangle\rangle=\langle B(\omega,t)\rangle$, which holds since the probability distribution in Eq. (29) is normalized. For the second term we assume that A_S is a function of position only, i.e., $A_S(\omega)=A_S(\mathbf{R})$, such that $\mathcal{L}_0A_S(\omega)$ is linear in the momenta and the average $\langle\mathcal{L}_0A_S(\omega)\rangle$ vanishes.

Second,

$$\begin{aligned}\mathcal{PP}_2B(\omega,t)&=\frac{\langle B(\omega,t)\mathcal{L}_0A_S(\omega)\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}\left[\langle\mathcal{L}_0A_S(\omega)\rangle+\frac{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}\mathcal{L}_0A_S(\omega)+\frac{\langle(A_S(\omega)-\langle A_S\rangle)\mathcal{L}_0A_S(\omega)\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}(A_S(\omega)-\langle A_S\rangle)\right]\\&=\mathcal{P}_2B(\omega,t),\end{aligned} \quad \text{(C6)}$$

where again we used that $A_S(\omega)=A_S(\mathbf{R})$ such that $\mathcal{L}_0A_S(\omega)$ is linear in the momenta and the averages $\langle\mathcal{L}_0A_S(\omega)\rangle$ and $\langle[A_S(\omega)-\langle A_S\rangle]\mathcal{L}_0A_S(\omega)\rangle$ vanish.

Third,

$$\begin{aligned}\mathcal{PP}_3B(\omega,t)&=\frac{\langle B(\omega,t)[A_S(\omega)-\langle A_S\rangle]\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}\\&\quad\times\left[\langle A_S(\omega)-\langle A_S\rangle\rangle+\frac{\langle[A_S(\omega)-\langle A_S\rangle]\mathcal{L}_0A_S(\omega)\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}\mathcal{L}_0A_S(\omega)+\frac{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle}[A_S(\omega)-\langle A_S\rangle]\right]\\&=\mathcal{P}_3B(\omega,t),\end{aligned} \quad \text{(C7)}$$

where again we used that $A_S(\omega)=A_S(\mathbf{R})$. Adding Eqs. (C5)–(C7) we see that $\mathcal{P}^2=\mathcal{P}(\mathcal{P}_1+\mathcal{P}_2+\mathcal{P}_3)=\mathcal{P}_1+\mathcal{P}_2+\mathcal{P}_3=\mathcal{P}$ and thus $\mathcal{P}$ is idempotent. From the idempotency of $\mathcal{P}$ it follows that $\mathcal{Q}$ is also idempotent, to prove this one writes

$$\mathcal{Q}^2B(\omega,t)=(1-\mathcal{P})^2B(\omega,t)=(1-2\mathcal{P}+\mathcal{P}^2)B(\omega,t)=(1-\mathcal{P})B(\omega,t)=\mathcal{Q}B(\omega,t). \quad \text{(C8)}$$

The self-adjointedness of $\mathcal{P}$, Eq. (30), is straightforwardly proven by writing

$$\langle C(\omega,t)\mathcal{P}B(\omega,t')\rangle \tag{C9}$$

$$= \langle C(\omega,t)\rangle\langle B(\omega,t')\rangle + \langle C(\omega,t)\mathcal{L}_0A_S(\omega)\rangle\frac{\langle B(\omega,t')\mathcal{L}_0A_S(\omega)\rangle}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle} + \langle C(\omega,t)[A_S(\omega)-\langle A_S\rangle]\rangle\frac{\langle B(\omega,t')(A_S(\omega)-\langle A_S\rangle)\rangle}{\langle[A_S(\omega)-\langle A_S\rangle]^2\rangle} \tag{C10}$$

$$= \langle B(\omega,t')\mathcal{P}C(\omega,t)\rangle. \tag{C11}$$

Using $\mathcal{Q}=1-\mathcal{P}$ we see straightforwardly that $\mathcal{Q}$ is also self-adjoint.

Using similar arguments as above, one can show that $\mathcal{P}c=$ c, $\mathcal{P}(A_S(\omega)-\langle A_S\rangle)=[A_S(\omega)-\langle A_S\rangle]$, $\mathcal{P}\mathcal{L}_0A_S(\omega)=\mathcal{L}_0A_S(\omega)$, from which follows that also $\mathcal{P}A_S(\omega)=A_S(\omega)$. From these relations we can directly conclude that $\mathcal{Q}c=0$, $\mathcal{Q}(A_S(\omega)-\langle A_S\rangle)=0$, $\mathcal{Q}\mathcal{L}_0A_S(\omega)=0$, and also $\mathcal{Q}A_S(\omega)=0$.

From the idempotency of $\mathcal{P}$ or $\mathcal{Q}$ we follow that $\mathcal{P}\mathcal{Q}=\mathcal{P}(1-\mathcal{P})=\mathcal{P}-\mathcal{P}^2=0$ and, similarly, $\mathcal{Q}\mathcal{P}=0$, thus, the operators $\mathcal{P}$ and $\mathcal{Q}$ are orthogonal to each other.

APPENDIX D: DERIVATION OF NONEQUILIBRIUM GLE

We consider the first term in Eq. (25), which can be split into two terms and reads, apart from the propagator in front,

$$\mathcal{P}\mathcal{L}(t)\mathcal{L}_0A_S(\omega) = \mathcal{P}\mathcal{L}_0^2A_S(\omega) - h(t)\mathcal{P}\Delta\mathcal{L}\mathcal{L}_0A_S(\omega). \tag{D1}$$

We apply the projection operator (27) on the first term in Eq. (D1), which generates three contributions. The first contribution is given by

$$\left\langle\mathcal{L}_0^2A_S(\omega)\right\rangle = -\beta h_p\langle[\mathcal{L}_0A_S(\omega)]^2\rangle, \tag{D2}$$

where we used that $\mathcal{L}_0$ is anti-self-adjoint and that $\mathcal{L}_0\rho_p(\omega)=\beta h_p\rho_p(\omega)\mathcal{L}_0A_S(\omega)$. Using the same properties of $\mathcal{L}_0$, the second contribution can be written as (apart from the normalization factor)

$$\left\langle[\mathcal{L}_0A_S(\omega)]\mathcal{L}_0^2A_S(\omega)\right\rangle = -\beta h_p\langle[\mathcal{L}_0A_S(\omega)]^3\rangle/2. \tag{D3}$$

The third contribution (apart from the normalization factor) follows as

$$\left\langle[(A_S(\omega)-\langle A_S\rangle]\mathcal{L}_0^2A_S(\omega)\right\rangle$$
$$= -\langle[\mathcal{L}_0A_S(\omega)]^2\rangle - \beta h_p\langle[A_S(\omega)-\langle A_S\rangle][\mathcal{L}_0A_S(\omega)]^2\rangle. \tag{D4}$$

We now apply the projection operator Eq. (27) on the second term in Eq. (D1), which again generates three contributions. The first contribution is given by

$$-h(t)\langle\Delta\mathcal{L}\mathcal{L}_0A_S(\omega)\rangle = \beta h(t)\langle[\mathcal{L}_0A_S(\omega)]^2\rangle, \tag{D5}$$

where we used that $\Delta\mathcal{L}$ is anti-self-adjoint and that $\Delta\mathcal{L}\rho_p(\omega)=\beta\rho_p(\omega)\mathcal{L}_0A_S(\omega)$. Using the same properties of $\Delta\mathcal{L}$, the second contribution can be written as (apart from the normalization factor)

$$-h(t)\langle[\mathcal{L}_0A_S(\omega)]\Delta\mathcal{L}\mathcal{L}_0A_S(\omega)\rangle = \beta h(t)\langle[\mathcal{L}_0A_S(\omega)]^3\rangle/2. \tag{D6}$$

The third contribution (apart from the normalization factor) follows as

$$-h(t)\langle[(A_S(\omega)-\langle A_S\rangle]\Delta\mathcal{L}\mathcal{L}_0A_S(\omega)\rangle$$
$$= \beta h(t)\langle[A_S(\omega)-\langle A_S\rangle][\mathcal{L}_0A_S(\omega)]^2\rangle. \tag{D7}$$

Again assuming that the observable is a function of position only, $A_S(\omega)=A_S(\mathbf{R})$, as we did in Appendix C when we derived the idempotency of the projection operator $\mathcal{P}$, we see that Eqs. (D3) and (D6) vanish because $\langle[\mathcal{L}_0A_S(\omega)]^3\rangle$ is odd in the momenta.

Combining the results in Eqs. (D2), (D4), (D5), and (D7), the first term in Eq. (25) reads

$$\exp_H\left(\int_{t_0}^t ds\mathcal{L}(s)\right)\mathcal{P}\mathcal{L}(t)\mathcal{L}_0A_S(\omega)$$
$$= -K(t)[A(\omega,t)-\langle A_S\rangle] + [h(t)-h_p]/M, \tag{D8}$$

where $K(t)$ and M are defined in Eqs. (32) and (39).

We now consider the last term in Eq. (25), which reads, without the time integral and the propagator in front, $\mathcal{P}\mathcal{L}(s)F(\omega,s,t)$. The projection operator (27) generates three contributions. The first contribution is given by

$$\begin{aligned}\langle\mathcal{L}(s)F(\omega,s,t)\rangle &= \beta[h(s)-h_p]\langle F(\omega,s,t)\mathcal{L}_0A_S(\omega)\rangle\\ &= \beta(h(s)-h_p)\langle F(\omega,s,t)\mathcal{Q}\mathcal{L}_0A_S(\omega)\rangle\\ &= 0,\end{aligned} \tag{D9}$$

where in the first line we used that $\mathcal{L}(s)$ is anti-self-adjoint and that $\mathcal{L}(s)\rho_p(\omega)=\beta(h_p-h(s))\rho_p(\omega)\mathcal{L}_0A_S(\omega)$, in the second line that $\mathcal{Q}$ is idempotent and self-adjoint, and in the third line that $\mathcal{Q}\mathcal{L}_0A_S(\omega)=0$ (as derived in Appendix C). The second contribution can be written as (apart from the normalization factor)

$$\begin{aligned}\langle[\mathcal{L}_0A_S(\omega)]\mathcal{L}(s)F(\omega,s,t)\rangle &= -\langle F(\omega,s,t)\mathcal{L}(s)\mathcal{L}_0A_S(\omega)\rangle + \beta(h(s)-h_p)\langle[\mathcal{L}_0A_S(\omega)]^2F(\omega,s,t)\rangle\\ &= -\langle F(\omega,s,s)F(\omega,s,t)\rangle + \beta(h(s)-h_p)\langle[\mathcal{L}_0A_S(\omega)]^2F(\omega,s,t)\rangle,\end{aligned} \tag{D10}$$

where in the first equation we used the same properties of $\mathcal{L}(s)$ as before and in the second equation that $\mathcal{Q}$ is idempotent and self-adjoint and the definition of the complementary force in Eq. (26).

The third contribution can be written as (apart from the normalization factor)

$$\begin{aligned}\langle (A_S(\omega) - \langle A_S\rangle)\mathcal{L}(s)F(\omega,s,t)\rangle &= -\langle F(\omega,s,t)\mathcal{L}(s)[A_S(\omega) - \langle A_S\rangle]\rangle + \beta(h(s)-h_p)\langle F(\omega,s,t)[A_S(\omega)-\langle A_S\rangle]\mathcal{L}_0 A_S(\omega)\rangle\\ &= \beta(h(s)-h_p)\langle F(\omega,s,t)[A_S(\omega)-\langle A_S\rangle]\mathcal{L}_0 A_S(\omega)\rangle,\end{aligned} \tag{D11}$$

where in the first equation we used the same properties of $\mathcal{L}(s)$ as before and in the second equation that $\mathcal{Q}$ is idempotent and self-adjoint and that $\Delta\mathcal{L}A_S(\omega) = 0$.

Combining the results in Eqs. (D8)–(D11), the expression for the general GLE in Eq. (25) leads to the explicit GLE in Eq. (31).

APPENDIX E: DERIVATION OF THE TIME HOMOGENEITY OF MEMORY KERNELS BY AVERAGING OVER EXTERNAL FORCE $h(t)$

In this section we show that the memory kernels of the GLE in Eq. (31) become time-homogeneous if one treats the external force $h(t)$ as a stochastic force that is averaged over according to Eq. (49). The proof is done to quadratic order in powers of $h(t)$. We start with the operator expansion for two general time-dependent operators $\mathcal{V}(t)$ and $\mathcal{W}(t)$, Eq. (B1), and note that we can construct a systematic perturbative expansion in terms of the operator $\mathcal{W}(t)$ by writing

$$\begin{aligned}\exp_H\left(\int_{t_0}^t \mathcal{V}(s)+\mathcal{W}(s)\,ds\right) &= \exp_H\left(\int_{t_0}^t ds\mathcal{V}(s)\right) + \int_{t_0}^t ds\exp_H\left(\int_{t_0}^s ds'\mathcal{V}(s')\right)\mathcal{W}(s)\exp_H\left(\int_s^t \mathcal{V}(s')+\mathcal{W}(s')ds'\right)\\ &= \exp_H\left(\int_{t_0}^t ds\mathcal{V}(s)\right) + \int_{t_0}^t ds\exp_H\left(\int_{t_0}^s ds'\mathcal{V}(s')\right)\mathcal{W}(s)\exp_H\left(\int_s^t \mathcal{V}(s')\,ds'\right) + \mathcal{O}(\mathcal{W}^2).\end{aligned} \tag{E1}$$

Expressions valid to higher order in $\mathcal{W}$ can be constructed by recursively inserting the second line of Eq. (E1) into the last operator exponential in the first line.

Following this method, the complementary force can be expanded in powers of the external force $h(t)$ as

$$\begin{aligned}F(\omega,t_0,t) &\equiv \exp_H\left(\mathcal{Q}\int_{t_0}^t ds'\mathcal{L}(s')\right)\mathcal{Q}\mathcal{L}(t)\mathcal{L}_0A_S(\omega)\\ &= \exp_H\left(\mathcal{Q}\int_{t_0}^t \mathcal{L}_0 - h(s')\Delta\mathcal{L}ds'\right)\mathcal{Q}(\mathcal{L}_0 - h(t)\Delta\mathcal{L})\mathcal{L}_0A_S(\omega)\\ &= \left[e^{(t-t_0)\mathcal{Q}L_0} - \int_{t_0}^t ds' e^{(s'-t_0)\mathcal{Q}L_0}h(s')\mathcal{Q}\Delta\mathcal{L}e^{(t-s')\mathcal{Q}L_0}\right]\mathcal{Q}(\mathcal{L}_0 - h(t)\Delta\mathcal{L})\mathcal{L}_0A_S(\omega) + O(h^2),\end{aligned} \tag{E2}$$

where higher-order terms can be recursively derived. We calculate here exemplarily the force average for the nonequilibrium friction memory kernel defined in Eq. (37)

$$\overline{\Gamma}_1(s,t) = \frac{-\beta\overline{h(s)\langle F(\omega,s,t)[\mathcal{L}_0A_S(\omega)]^2\rangle}}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle} = \frac{-\beta(X_1(s,t)+X_2(s,t))}{\langle[\mathcal{L}_0A_S(\omega)]^2\rangle}, \tag{E3}$$

which we split into two terms using the expansion for $F(\omega,t_0,t)$ in Eq. (E2); note that we omit all terms linear in $h(t)$ from the start since they vanish after averaging according to Eq. (49). The first term reads

$$X_1(s,t) = -\overline{h(s)\langle[\mathcal{L}_0A_S(\omega)]^2e^{(t-s)\mathcal{Q}L_0}\mathcal{Q}h(t)\Delta\mathcal{L}\mathcal{L}_0A_S(\omega)\rangle} = -\sigma(t-s)\langle[\mathcal{L}_0A_S(\omega)]^2e^{(t-s)\mathcal{Q}L_0}\mathcal{Q}h(t)\Delta\mathcal{L}\mathcal{L}_0A_S(\omega)\rangle, \tag{E4}$$

which clearly is a function of $t-s$ only.

The second term reads

$$\begin{aligned}X_2(s,t) &= -\overline{h(s)\left\langle[\mathcal{L}_0A_S(\omega)]^2\int_s^t ds'e^{(s'-s)\mathcal{Q}L_0}h(s')\mathcal{Q}\Delta\mathcal{L}e^{(t-s')\mathcal{Q}L_0}\mathcal{Q}\mathcal{L}_0^2A_S(\omega)\right\rangle}\\ &= -\left\langle[\mathcal{L}_0A_S(\omega)]^2\int_s^t ds'e^{(s'-s)\mathcal{Q}L_0}\sigma(s'-s)\mathcal{Q}\Delta\mathcal{L}e^{(t-s')\mathcal{Q}L_0}\mathcal{Q}\mathcal{L}_0^2A_S(\omega)\right\rangle\\ &= -\left\langle[\mathcal{L}_0A_S(\omega)]^2\int_0^{t-s} d\tilde{s}'e^{\tilde{s}'\mathcal{Q}L_0}\sigma(\tilde{s}')\mathcal{Q}\Delta\mathcal{L}e^{(t-s-\tilde{s}')\mathcal{Q}L_0}\mathcal{Q}\mathcal{L}_0^2A_S(\omega)\right\rangle,\end{aligned} \tag{E5}$$

which also clearly is a function of $t-s$ only and where we have averaged over the force $h(t)$ according to Eq. (49) and have changed the integration variable according to $\tilde{s}' = s' - s$. Thus, $\overline{\Gamma}_1(t-s)$ defined in Eq. (37) is a function of $t-s$ only, the same holds for $\overline{\Gamma}_2(t-s)$ defined in Eq. (41). Using similar techniques, it can be also shown that $\overline{\Gamma}_0(t-s)$ defined in Eq. (36), $\overline{\Gamma}_A(t-s)$ defined in Eq. (38) and $\overline{F}(\omega,t-s)$ are functions of $t-s$ only, which proves the functional dependencies in Eqs. (50) and (73).

APPENDIX F: PREAVERAGING THE NONEQUILIBRIUM GLE OVER THE EXTERNAL FORCE $h(t)$

In this section we preaverage the nonequilibrium GLE (31) over the stochastic nonequilibrium force $h(t)$ and show that the preaveraging approximation corresponds to the first term in a systematic expansion in powers of the external-force autocorrelation $\sigma(t)$. To reduce the notational burden we consider an unconfined system that is characterized by a diverging second moment, $\langle [A_S(\omega) - \langle A_S\rangle]^2\rangle = \infty$, in which case $K(t) = 0 = \Gamma_A(s,t)$. Equation (31) in this scenario simplifies to

$$F(\omega, t_0, t) + h(t)/M = \ddot{A}(\omega, t) + \int_{t_0}^{t} ds\, \Gamma(s,t)\dot{A}(\omega, s). \tag{F1}$$

By adding the preaveraged memory kernel $\overline{\Gamma}(s-t)$ on both sides we obtain

$$F(\omega, t_0, t) + h(t)/M + \int_{t_0}^{t} ds\, [\overline{\Gamma}(s-t) - \Gamma(s,t)]\dot{A}(\omega, s) = \ddot{A}(\omega, t) + \int_{t_0}^{t} ds\, \overline{\Gamma}(s-t)\dot{A}(\omega, s). \tag{F2}$$

Obviously, $\overline{\Gamma}(s-t) - \Gamma(s,t)$ vanishes for $h(t) = 0$, so this term is (at least) linear in $h(t)$. Using the time-domain version $\chi^{-1}(t)$ of the inverse response function defined in Eq. (54), which is related to the response function $\chi(t)$ via $\int ds \chi^{-1}(t-s)\chi(s-t') = \delta(t'-t)$, we obtain

$$F(\omega, t_0, t) + h(t)/M + \int_{t_0}^{t} ds\, [\overline{\Gamma}(s-t) - \Gamma(s,t)]\dot{A}(\omega, s) = \int_{-\infty}^{\infty} ds\, \chi^{-1}(s-t)A(\omega, s). \tag{F3}$$

Multiplying by $\chi(t-u)$ and integrating over t we obtain

$$A(\omega, u) = \int_{-\infty}^{\infty} dt\, \chi(t-u)\left(F(\omega, t_0, t) + h(t)/M + \int_{t_0}^{t} ds\, [\overline{\Gamma}(s-t) - \Gamma(s,t)]\dot{A}(\omega, s)\right), \tag{F4}$$

which is a recursive solution for $A(\omega, u)$. Since $\overline{\Gamma}(s-t) - \Gamma(s,t)$ is at least linear in $h(t)$, Eq. (F4) can be used to construct the solution for $A(\omega, u)$ in terms of a systematic expansion in powers of $h(t)$ by iteration. Equation (F4) also demonstrates that the leading term of $A(\omega, u)$ in this expansion is linear in $F(\omega, t_0, t)$ and $h(t)/M$. Thus, the term $[\overline{\Gamma}(s-t) - \Gamma(s,t)]\dot{A}(\omega, s)$ is of second order. Therefore, to leading order in an expansion of the observable $A(\omega, u)$ in powers of $F(\omega, t_0, t)$ and $h(t)/M$, the term $\Gamma(s,t)$ in Eq. (F1) can simply be replaced by $\overline{\Gamma}(s-t)$, which corresponds to the preaveraging approximation used to derive Eq. (50). Since odd terms in $h(t)$ vanish after averaging over $h(t)$, a higher order of $h(t)$ means higher order in the external-force variance $\sigma(t)$. The same argumentation can be used to replace $F(\omega, t_0, t)$ in Eq. (F1) by $\overline{F}(\omega, t - t_0)$, as has been done in deriving Eq. (74).

APPENDIX G: DERIVATION OF TWO-POINT CORRELATION FUNCTIONS IN TERMS OF HEISENBERG OBSERVABLES

We derive the two-point correlation function of the Heisenberg observable, to reduce the notational complexity we here use a time-independent Liouville operator $\mathcal{L}_0(\omega)$. Splitting the time propagation of the density distribution in Eq. (10) into two steps we obtain

$$\rho(\omega, t) = e^{-(t-t')\mathcal{L}_0(\omega)}e^{-(t'-t_0)\mathcal{L}_0(\omega)}\rho(\omega, t_0). \tag{G1}$$

Introducing delta functions we obtain

$$\rho(\omega, t) = \int d\omega' \int d\omega_0 e^{-(t-t')\mathcal{L}_0(\omega)}\delta(\omega - \omega')e^{-(t'-t_0)\mathcal{L}_0(\omega')}\delta(\omega' - \omega_0)\rho(\omega_0, t_0) \equiv \int d\omega' \int d\omega_0 \rho(\omega, t; \omega', t'; \omega_0, t_0), \tag{G2}$$

where in the second line we defined the three-point joint distribution function $\rho(\omega, t; \omega', t'; \omega_0, t_0)$. With this distribution, the two-point correlation function defined in Eq. (57) can be written as

$$C(t, t') = \int d\omega \int d\omega' \int d\omega_0 [A_S(\omega) - \langle A_S\rangle][A_S(\omega') - \langle A_S\rangle]\rho(\omega, t; \omega', t'; \omega_0, t_0). \tag{G3}$$

Inserting the definition of $\rho(\omega, t; \omega', t'; \omega_0, t_0)$ from Eq. (G2) and using that $\mathcal{L}_0(\omega)$ is anti-self-adjoint, we obtain

$$C(t, t') = \int d\omega \rho(\omega, t_0)e^{(t'-t_0)\mathcal{L}_0(\omega)}[A_S(\omega) - \langle A_S\rangle]e^{(t-t')\mathcal{L}_0(\omega)}[A_S(\omega) - \langle A_S\rangle]. \tag{G4}$$

We now use the product propagation relation derived in Appendix H and obtain

$$C(t, t') = \int d\omega \rho(\omega, t_0)(e^{(t'-t_0)\mathcal{L}_0(\omega)}[A_S(\omega) - \langle A_S\rangle])e^{(t-t_0)\mathcal{L}_0(\omega)}[A_S(\omega) - \langle A_S\rangle], \tag{G5}$$

which can, using the definition of the Heisenberg observable, be rewritten as

$$C(t, t') = \int d\omega \rho(\omega, t_0)[A(\omega, t_0, t') - \langle A_S\rangle][A(\omega, t_0, t) - \langle A_S\rangle]. \tag{G6}$$

In this expression t_0 denotes the projection time and $t \geqslant t_0$ and $t' \geqslant t_0$ denote arbitrary later times; note that we suppress the dependence of the Heisenberg observable on the projection time throughout this paper.

APPENDIX H: DERIVATION OF PRODUCT PROPAGATION RELATION

To reduce the notational complexity, we here use a time-independent Liouville operator $\mathcal{L}_0(\omega)$ and suppress the phase-space dependence of $\mathcal{L}_0(\omega)$ and of the two general phase-space functions $A(\omega)$ and $B(\omega)$ in the following. The Liouville propagator acting on the product of two phase-space functions follows from the series definition of the exponential as

$$e^{t\mathcal{L}_0}AB = \sum_{n=0}^{\infty} \frac{t^n \mathcal{L}_0^n}{n!} AB. \tag{H1}$$

Since $\mathcal{L}_0$ is a linear differential operator the product rule applies, which reads

$$\mathcal{L}_0 AB = A\mathcal{L}_0 B + B\mathcal{L}_0 A. \tag{H2}$$

Applying the product rule recursively, we find

$$\mathcal{L}_0^n AB = \sum_{m=0}^{n} \frac{n!}{m!(n-m)!} \big(\mathcal{L}_0^m A\big)\mathcal{L}_0^{n-m} B. \tag{H3}$$

Combining Eqs. (H1) and (H3) we find

$$\begin{aligned} e^{t\mathcal{L}_0}AB &= \sum_{n=0}^{\infty}\sum_{m=0}^{n} \frac{t^n}{m!(n-m)!}\big(\mathcal{L}_0^m A\big)\mathcal{L}_0^{n-m}B \\ &= \sum_{m=0}^{\infty}\sum_{n=m}^{\infty} \frac{t^n}{m!(n-m)!}\big(\mathcal{L}_0^m A\big)\mathcal{L}_0^{n-m}B \\ &= \sum_{m=0}^{\infty}\sum_{n=0}^{\infty} \frac{t^{n+m}}{m!n!}\big(\mathcal{L}_0^m A\big)\mathcal{L}_0^{n}B. \end{aligned} \tag{H4}$$

Using Eq. (H1) we finally arrive at the product popagation relation

$$e^{t\mathcal{L}_0}AB = (e^{t\mathcal{L}_0}A)e^{t\mathcal{L}_0}B. \tag{H5}$$

APPENDIX I: DERIVATION OF THE PREAVERAGED TWO-POINT CORRELATION FUNCTION

In this section we derive the two-point correlation function defined in Eq. (57) by a preaverage over the stochastic external force $h(t)$. We start from the nonequilibrium GLE (31). To reduce the notational burden we consider an unconfined system that is characterized by a diverging second moment, $\langle [A_S(\omega) - \langle A_S\rangle]^2\rangle = \infty$, in which case $K(t) = 0 = \Gamma_A(s,t)$. Equation (31) in this scenario simplifies to

$$F(\omega, t_0, t) + h(t)/M = \ddot{A}(\omega, t) + \int_{t_0}^{t} ds\, \Gamma(s,t)\dot{A}(\omega, s). \tag{I1}$$

Averaging the product of Eq. (I1) at two different times t and t' over phase space and $h(t)$ we obtain the expression

$$\begin{aligned} \overline{\left\langle\left(F(\omega,t_0,t)+\frac{h(t)}{M}\right)\left(F(\omega,t_0,t')+\frac{h(t')}{M}\right)\right\rangle} &= \overline{\langle F(\omega,t_0,t)F(\omega,t_0,t')\rangle} + \frac{\overline{h(t)h(t')}}{M^2} \\ &= \overline{\left\langle\left(\ddot{A}(\omega,t)+\int_{t_0}^{t} ds\,\Gamma(s,t)\dot{A}(\omega,s)\right)\left(\ddot{A}(\omega,t')+\int_{t_0}^{t'} ds'\,\Gamma(s',t')\dot{A}(\omega,s')\right)\right\rangle} \\ &= \overline{\left\langle\left(\ddot{A}(\omega,t)+\int_{t_0}^{t} ds\,\overline{\Gamma}(t-s)\dot{A}(\omega,s)\right)\left(\ddot{A}(\omega,t')+\int_{t_0}^{t'} ds'\,\overline{\Gamma}(t'-s')\dot{A}(\omega,s')\right)\right\rangle}, \end{aligned} \tag{I2}$$

where in the first equation we used that $h(t)$ is independent of phase space and that $\langle F(\omega, t_0, t)\rangle = 0$ and in the last equation we used the preaveraging approximation introduced in Appendix F. Performing the average over phase space ω and $h(t)$ on the left side in Eq. (I2) and using the time-domain version of the inverse susceptibility (54) we obtain

$$\langle [\mathcal{L}_0 A_S(\omega)]^2\rangle \overline{\Gamma}_0(t-t') + \frac{\sigma(t-t')}{M^2} = \int_{-\infty}^{\infty} ds\, \chi^{-1}(s-t)\int_{-\infty}^{\infty} ds'\, \chi^{-1}(s'-t')\overline{\langle A(\omega,s)A(\omega,s')\rangle}. \tag{I3}$$

By inversion we obtain

$$\overline{\langle A(\omega,t)A(\omega,t')\rangle} = C(t-t') = \int_{-\infty}^{\infty} ds\, \chi^{-1}(s-t)\int_{-\infty}^{\infty} ds'\, \chi^{-1}(s'-t')\left(\langle [\mathcal{L}_0 A_S(\omega)]^2\rangle \overline{\Gamma}_0(s-s') + \frac{\sigma(s-s')}{M^2}\right). \tag{I4}$$

By Fourier transformation, Eq. (58) follows.

APPENDIX J: DERIVATION OF THE STANDARD FDT IN THE HEISENBERG PICTURE

Here we derive the standard (i.e., nonequilibrium) FDT by leading-order perturbation analysis of the Heisenberg observable in Eq. (13), which deviates from the textbook derivation of the standard FDT. To proceed, using the expression for the Heisenberg observable Eq. (13) and the expression for the time-dependent Liouville operator Eq. (18), we obtain

$$A(\omega,t) = \exp_H\left(\int_{t_0}^{t} ds\mathcal{L}(s)\right)A_S(\omega)$$
$$= \exp_H\left(\int_{t_0}^{t} ds(\mathcal{L}_0 - h(s)\Delta\mathcal{L})\right)A_S(\omega), \quad \text{(J1)}$$

where the Heisenberg propagator is defined in Eq. (14). Expanding the operator exponential to first order in $h(t)$, as shown in Eq. (E1), we obtain

$$A(\omega,t) = \left[e^{(t-t_0)\mathcal{L}_0} - \int_{t_0}^{t} dsh(s)e^{(s-t)\mathcal{L}_0}\Delta\mathcal{L}e^{(t-s)\mathcal{L}_0}\right]A_S(\omega). \quad \text{(J2)}$$

The mean observable follows using the definition (28) as

$$a(t) = \langle A(\omega,t)\rangle$$
$$= \langle e^{(t-t_0)\mathcal{L}_0}A_S(\omega)\rangle$$
$$- \left\langle\int_{t_0}^{t} dsh(s)e^{(s-t)\mathcal{L}_0}\Delta\mathcal{L}e^{(t-s)\mathcal{L}_0}A_S(\omega)\right\rangle, \quad \text{(J3)}$$

which can be rewritten, using the anti-self-adjointedness of $\mathcal{L}_0$ and $\Delta\mathcal{L}$, as

$$a(t) = \langle A_S(\omega)\rangle - \int_{t_0}^{t} ds\beta h(s)\langle A_S(\omega)\mathcal{L}_0 e^{(t-s)\mathcal{L}_0}A_S(\omega)\rangle$$
$$= \langle A_S(\omega)\rangle - \int_{t_0}^{t} ds\beta h(s)\frac{d}{dt}\langle A_S(\omega)e^{(t-s)\mathcal{L}_0}A_S(\omega)\rangle$$
$$= \langle A_S(\omega)\rangle + \int_{t_0}^{t} dsh(s)\chi(t-s)/M. \quad \text{(J4)}$$

Here we defined the response function as

$$\chi(t) = -\beta M\frac{d}{dt}\langle A_S(\omega)e^{t\mathcal{L}_0}A_S(\omega)\rangle = -\beta M\frac{d}{dt}C(t), \quad \text{(J5)}$$

which is the standard FDT as one finds it in textbooks, where the derivation is typically done in the Schrödinger picture as there is no advantage of the Heisenberg picture for this derivation. In this paper we use the Heisenberg picture since it is needed when deriving the GLE. Note that Eq. (J5) has no dependence on the nonequilibrium force $h(t)$ whatsoever and thus is equivalent to Eq. (61), which is the Fourier-transformed version of the FDT, in the limit $h(t) = 0$.

To make the equivalence between Eqs. (J5) and (61) obvious, we Fourier transform Eqs. (J4) and (J5). For this, we define the response function $\chi(t)$ as single-sided and choose $t_0 \to -\infty$, after which Eq. (J4) reads

$$a(t) = \langle A_S(\omega)\rangle + \int_{-\infty}^{\infty} dsh(s)\chi(t-s)/M. \quad \text{(J6)}$$

Fourier transformation yields

$$\tilde{a}(\nu) = 2\pi\delta(\nu)\langle A_S\rangle + \tilde{\chi}(\nu)\tilde{h}(\nu)/M, \quad \text{(J7)}$$

which is equivalent to Eq. (53). Accounting for the single-sidedness of $\chi(t)$, Eq. (J5) can be written as

$$\chi(t) = -\beta M\theta(t)\frac{d}{dt}C(t), \quad \text{(J8)}$$

where $\theta(t)$ denotes the Heavyside function. After Fourier transformation Eq. (J8) reads

$$\tilde{\chi}(\nu) = \beta MC(0) - \imath\nu\beta M\tilde{C}^+(\nu). \quad \text{(J9)}$$

The odd part of this equation reads

$$\tilde{\chi}(\nu) - \tilde{\chi}(-\nu) = -\imath\nu\beta M\tilde{C}(\nu), \quad \text{(J10)}$$

where we used that the Fourier transform of the symmetric correlation function is $\tilde{C}(\nu) = \tilde{C}^+(\nu) + \tilde{C}^+(-\nu)$. Since the time-domain response function $\chi(t)$ is a real function, we finally obtain

$$2\mathrm{Im}(\tilde{\chi}(\nu)) = -\nu\beta M\tilde{C}(\nu) = -\frac{\nu\tilde{C}(\nu)}{\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle}, \quad \text{(J11)}$$

where we used the definition of the mass M in Eq. (39) and which is equivalent to Eq. (61), provided we take the equilibrium limit $h(t) = 0$ in Eq. (61).

APPENDIX K: DERIVATION OF JOINT DISTRIBUTION FUNCTION VIA PATH INTEGRALS

Inserting Eq. (74) into Eq. (73) and after Fourier transformation we obtain

$$\rho(A_2,t_2;A_1,t_1) = \int_{-\infty}^{\infty}\frac{dq_1}{2\pi}\int_{-\infty}^{\infty}\frac{dq_2}{2\pi}e^{\imath q_1 A_1 - \langle A_S\rangle) + \imath q_2(A_2 - \langle A_S\rangle)]}\overline{\left\langle\exp\left[-\imath\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}(\tilde{\tilde{F}}(\omega,\nu) + \tilde{h}(\nu)/M)\tilde{g}(\nu)\right]\right\rangle}, \quad \text{(K1)}$$

where we have defined

$$\tilde{g}(\nu) \equiv \tilde{\chi}(\nu)(q_1 e^{\imath\nu t_1} + q_2 e^{\imath\nu t_2}). \quad \text{(K2)}$$

We diagonalize the path integrals in Eqs. (75) and (76) by Fourier transformation and obtain

$$\langle X(\tilde{\tilde{F}},\tilde{h})\rangle = \int_{-\infty}^{\infty}\frac{\mathcal{D}\tilde{\tilde{F}}(\omega,\cdot)}{\tilde{\mathcal{N}}_F}X(\tilde{\tilde{F}},\tilde{h})\exp\left(-\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}\frac{\tilde{\tilde{F}}(\omega,\nu)\tilde{\tilde{F}}(\omega,-\nu)}{2\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle\tilde{\tilde{\Gamma}}_0(\nu)}\right), \quad \text{(K3)}$$

$$\overline{X(\tilde{\tilde{F}},\tilde{h})} = \int_{-\infty}^{\infty}\frac{\mathcal{D}\tilde{h}(\cdot)}{\tilde{\mathcal{N}}_h}X(\tilde{\tilde{F}},\tilde{h})\exp\left(-\int_{-\infty}^{\infty}\frac{d\nu}{2\pi}\frac{\tilde{h}(\nu)\tilde{h}(-\nu)}{2\tilde{\sigma}(\nu)}\right), \quad \text{(K4)}$$

where $\tilde{\mathcal{N}}_F$ and $\tilde{\mathcal{N}}_h$ are normalization constants. Now the averages over $\tilde{\tilde{F}}(\omega, \nu)$ and $\tilde{h}(\nu)$ in Eq. (K1) can be done by explicitly performing the path integrals, after which we obtain

$$\rho(A_2, t_2; A_1, t_1) = \int_{-\infty}^{\infty} \frac{dq_1}{2\pi} \int_{-\infty}^{\infty} \frac{dq_2}{2\pi} e^{\imath q_1[A_1 - \langle A_S \rangle) + \imath q_2(A_2 - \langle A_S \rangle]} \exp\left(-\int_{-\infty}^{\infty} \frac{d\nu}{4\pi} \tilde{g}(\nu)\tilde{g}(-\nu)(\langle[\mathcal{L}_0 A_S(\omega)]^2\rangle \tilde{\tilde{\Gamma}}_0(\nu) + \tilde{\sigma}(\nu)/M^2)\right). \tag{K5}$$

Inserting the definition of $\tilde{g}(\nu)$ from Eq. (K2) we obtain

$$\rho(A_2, t_2; A_1, t_1) = \int_{-\infty}^{\infty} \frac{dq_1}{2\pi} \int_{-\infty}^{\infty} \frac{dq_2}{2\pi} e^{\imath q_1[A_1 - \langle A_S \rangle) + \imath q_2(A_2 - \langle A_S \rangle]} \exp\left(-\int_{-\infty}^{\infty} \frac{d\nu}{4\pi} \tilde{C}(\nu) \sum_{j,k=1}^{2} q_j q_k e^{\imath\nu(t_j - t_k)}\right), \tag{K6}$$

where we used the explicit expression for the Fourier-transformed correlation function $\tilde{C}(\nu)$ from Eq. (58). Now the Fourier integral in the exponent can be done, and we obtain

$$\rho(A_2, t_2; A_1, t_1) = \int_{-\infty}^{\infty} \frac{dq_1}{2\pi} \int_{-\infty}^{\infty} \frac{dq_2}{2\pi} e^{\imath q_1[A_1 - \langle A_S \rangle) + \imath q_2(A_2 - \langle A_S \rangle]} \exp\left(-\frac{1}{2} \sum_{j,k=1}^{2} q_j q_k C(t_k - t_j)\right), \tag{K7}$$

which can be slightly rewritten as

$$\rho(A_2, t_2; A_1, t_1) = \int_{-\infty}^{\infty} \frac{dq_1}{2\pi} \int_{-\infty}^{\infty} \frac{dq_2}{2\pi} \exp\left(-\frac{1}{2} \sum_{j,k=1}^{2} q_j q_k C(t_k - t_j) + \imath \sum_{j=1}^{2} q_j(A_j - \langle A_S \rangle)\right). \tag{K8}$$

Now we use the definition (78), $I_{jk} = C(t_j - t_k)$, and obtain

$$\rho(A_2, t_2; A_1, t_1) = \int_{-\infty}^{\infty} \frac{dq_1}{2\pi} \int_{-\infty}^{\infty} \frac{dq_2}{2\pi} \exp\left(-\frac{1}{2} \sum_{j,k=1}^{2} q_j q_k I_{jk} + \imath \sum_{j=1}^{2} q_j(A_j - \langle A_S \rangle)\right). \tag{K9}$$

The Gaussian integrals over q_1 and q_2 can be done, after which we obtain the final result reported in Eq. (77),

$$\rho(A_2, t_2; A_1, t_1) = \frac{\exp\left(-\sum_{j,k=1}^{2}(A_j - \langle A_S \rangle) I_{jk}^{-1}(A_k - \langle A_S \rangle)/2\right)}{\sqrt{\det 2\pi I}}, \tag{K10}$$

which is the two-point distribution of a general Gaussian process. Note that in Eq. (77) the Einstein summation convention is used.

As a final step, we show how to marginalize the two-point distribution. This is most easily done on the level of Eq. (K9) where the positions A_1 and A_2 appear linearly. By integration over one observable value we obtain

$$\rho(A_1, t_1) = \int_{-\infty}^{\infty} dA_2 \rho(A_2, t_2; A_1, t_1) = \int_{-\infty}^{\infty} \frac{dq_1}{2\pi} \exp\left(-\frac{q_1^2 C(0)}{2} + q_1(A_1 - \langle A_S \rangle)\right) = \frac{\exp\left(-\frac{(A_1 - \langle A_S \rangle)^2}{2C(0)}\right)}{\sqrt{2\pi C(0)}}, \tag{K11}$$

which, expectedly, is a Gaussian distribution with a variance corresponding to the equal-time correlation $C(0)$.

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