Letter

Nonequilibrium solvent response force: What happens if you push a Brownian particle

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In this Letter we discuss how to add forces to the Langevin equation. We derive an exact generalized Langevin equation for the dynamics of one particle subject to an external force embedded in a system of many interacting particles. The external force may depend on time and/or on the phase-space coordinates of the system. We construct a projection operator such that the drift coefficient, the memory kernel, and the fluctuating force of the generalized Langevin equation are the same as for the system without external driving. We show that next to the external force another force term occurs that is caused by the nonequilibrium response of the solvent. The first contribution to the ensemble average of this force stems from third or higher order terms of the external force and from sixth or higher order terms of time. We also analyze the additional force term numerically for an exemplary system.

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Langevin's description of the motion of a Brownian particle is a typical example of a coarse-grained model [1]. When modeling Brownian motion, the effect of the solvent degrees of freedom is not computed explicitly but replaced by two effective, coarse-grained quantities: the friction coefficient γ and the random force $\boldsymbol{f}(t)$. The velocity of a Brownian particle of mass m then obeys the equation

$$m\frac{d\boldsymbol{v}(t)}{dt} = -\gamma\boldsymbol{v}(t) + \boldsymbol{f}(t). \tag{1}$$

The components of $\boldsymbol{f}(t)$ are related to γ by the fluctuation dissipation theorem (FDT), $\langle f_i(t)f_j(s)\rangle = 2\gamma k_B T\delta_{i,j}\delta(t-s)$, where k_B is Boltzmann's constant and T is the temperature.

If the Brownian particle is subjected to an external force $\boldsymbol{F}_{\text{ext}}$, the force often is simply added to Eq. (1). This is common practice when molecular dynamics (MD) simulations are carried out with a Langevin thermostat [2], i.e., it is done in a large number of MD simulation and Brownian Dynamics (BD) simulation studies of colloidal systems, biomolecular systems, and polymeric systems.

Two seminal pieces of work are often cited to justify this approach: the article by Kubo on the fluctuation dissipation theorem [3] and the article by Zwanzig on the nonlinear generalized Langevin equation [4]. However, in neither of these articles is such a statement derived. Kubo explicitly wrote that he only considered linear effects in the external force, and Zwanzig considered particles in a bath, i.e., the case in which the degrees of freedom which are integrated out do not interact with each other. Other derivations like the one by Batchelor [5] or the one by Hauge and Martin-Löf [6] already start out from hydrodynamics, thus they contain assumptions about local thermal averages and do not give a justification based on the full microscopic description.

In contrast, there is a piece of work in the literature that proves that forces cannot simply be added: a very readable article from 1972 by Kim and Oppenheim [7], which unfortunately has hardly ever been cited. In this article, the method famously used by Mazur and Oppenheim to derive the time-local Langevin equation [8] is extended to the case of an applied external force. One result of the derivation by Kim and Oppenheim is that the FDT does not hold if the dynamics are non-Markovian and there is an external force.

In this Letter we recall why forces cannot simply be added on the level of the Langevin equation. Then we introduce a version of the projection operator method [9] (different from the one used in Ref. [7]) which allows to recover all terms in Eq. (1) including the FDT and which does produce an additive force term, however, this comes at the cost of one additional term in the resulting Langevin equation. We then discuss the impact of this additional term on the resulting effective dynamics.

We start by briefly recalling Mori's work [10]: We consider the Hamiltonian equations of motion of N interacting particles and integrate out all degrees of freedom apart from one component of the momentum of one particle. Under stationary conditions, the linear projection operator by Mori can be used for this task and one obtains the linear generalized Langevin equation:

$$\frac{dp_z(t;\Gamma)}{dt} = -\int_0^t d\tau K(t-\tau)p_z(\tau;\Gamma) + \eta(t;\Gamma). \tag{2}$$

Here $p_z(t;\Gamma)$ denotes the momentum in the z direction at time t given that the entire system was initialized at the phase-space point Γ at time zero. $K(\tau)$ is the so-called memory kernel and $\eta(t;\Gamma)$ is the (deterministic) fluctuating force.

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The memory kernel and fluctuating force fulfill a fluctuation-kernel theorem

$$\langle \eta(t;\Gamma)\eta(t';\Gamma)\rangle = K(t-t')\langle p_z^2\rangle. \tag{3}$$

[This type of equation is usually called a second fluctuation-dissipation theorem. However, as the structure holds generally for linear versions of the generalized Langevin equation irrespective of whether the kernel can be related to dissipation, we prefer to call it a fluctuation-kernel theorem (FKT).]

Now we address the following question: If one considers the same system but with an additional external force acting on the particle of interest, could the motion be described by an equation of the form

$$\frac{dp_z(t;\Gamma)}{dt} = -\int_0^t d\tau K(t-\tau)p_z(\tau;\Gamma) + \eta(t;\Gamma) + F_{\rm ext}(t;\Gamma)? \tag{4}$$

In the case of time-scale separation between p_z and the other degrees of freedom, Eq. (1) with an additive force would follow from Eq. (4). However, Mori's derivation only holds under stationary conditions, and it is not obvious that the effect of the external force can simply be added on the level of the coarse-grained description.

We first tackle the problem in more general terms and then treat Eq. (4) as a special case. Given a system which is described by two Liouvillians, a time-independent Liouvillian $\mathcal{L}_0$ for the interactions within the system and $\mathcal{L}_1(t)$ for the time-dependent external force, the time evolution of an arbitrary observable $A(\Gamma)$ is given by [11,12]

$$\frac{dA(t;\Gamma)}{dt} = \mathcal{U}(t,t_0)\mathcal{L}_{\rm tot}(t)A(\Gamma), \tag{5}$$

where $\mathcal{L}_{\rm tot}(t) = \mathcal{L}_0 + \mathcal{L}_1(t)$. Here, $\mathcal{U}(t,t_0)$ is the time-evolution operator that can be expressed as a negatively time-ordered exponential

$$\mathcal{U}(t,t_0) = \exp_-\left(\int_{t_0}^t d\tau \mathcal{L}_{\rm tot}(\tau)\right), \tag{6}$$

and Γ denotes the initial point in phase space at time t_0. Introducing a projection operator $\mathcal{P}$ and its orthogonal complement $\mathcal{Q} := 1 - \mathcal{P}$, Eq. (5) can be rewritten as

$$\frac{dA(t;\Gamma)}{dt} = \mathcal{U}(t,t_0)\mathcal{P}\mathcal{L}_0A(\Gamma) + \mathcal{U}(t,t_0)\mathcal{Q}\mathcal{L}_0A(\Gamma) + \mathcal{U}(t,t_0)\mathcal{L}_1(t)A(\Gamma). \tag{7}$$

We now rewrite second term on the right-hand side using a modified Dyson decomposition. To this end, we define

$$\mathcal{Z}(t,t_0) := \mathcal{U}(t,t_0)\mathcal{Q} \tag{8}$$

and calculate the time derivative of $\mathcal{Z}(t,t_0)$ in order to obtain the differential equations

$$\frac{\partial Z(t,t_0)}{\partial t} = \mathcal{U}(t,t_0)\mathcal{L}_{\rm tot}(t)\mathcal{Q} \tag{9}$$

$$= \mathcal{Z}(t,t_0)\mathcal{L}_{\rm tot}(t)\mathcal{Q} + \mathcal{U}(t,t_0)\mathcal{P}\mathcal{L}_{\rm tot}(t)\mathcal{Q} \tag{10}$$

$$= \mathcal{Z}(t,t_0)\mathcal{L}_0\mathcal{Q} + \mathcal{U}(t,t_0)(\mathcal{P}\mathcal{L}_0 + \mathcal{L}_1(t))\mathcal{Q}. \tag{11}$$

In more general cases where the time-dependent part of the Hamiltonian/Liouvillian cannot be separated from the time-independent part as easily as here, Eq. (10) is usually taken as the starting point to derive a generalized Langevin equation/Dyson decomposition [12,13]. However, as the homogeneous part of the differential equation,

$$\frac{\partial Z_{\rm hom}(t,t_0)}{\partial t} = \mathcal{Z}_{\rm hom}(t,t_0)\mathcal{L}_{\rm tot}(t)\mathcal{Q}, \tag{12}$$

already contains the external force through $\mathcal{L}_{\rm tot}(t)$, it is not suitable to derive a generalized Langevin equation that has the same memory kernel as the stationary process as well as a fluctuation-kernel theorem. Instead we use Eq. (11), where the $\mathcal{L}_1(t)$ contribution is shifted from the homogeneous part of the differential equation to the inhomogeneity. In this case the solution to the homogeneous differential equation reads

$$\mathcal{G}(t-t_0) := \exp\left((t-t_0)\mathcal{L}_0\mathcal{Q}\right), \tag{13}$$

which can be used to find the special solution to the inhomogeneous equation

$$\mathcal{Z}(t,t_0) = \mathcal{U}(t',t_0)\mathcal{Q}\mathcal{G}(t-t') + \int_{t'}^t d\tau \mathcal{U}(\tau,t_0)(\mathcal{P}\mathcal{L}_0 + \mathcal{L}_1(\tau))\mathcal{Q}\mathcal{G}(t-\tau). \tag{14}$$

It is easy to check that this expression solves the differential equation (11) together with the boundary condition $\mathcal{Z}(t',t_0) = \mathcal{U}(t',t_0)\mathcal{Q}$ (cf. Refs. [11,14]) and, hence, one can substitute $\mathcal{U}(t,t_0)\mathcal{Q}$ in Eq. (7) with the new expression. One obtains the following equation of motion:

$$\frac{dA(t;\Gamma)}{dt} = \mathcal{U}(t,t_0)\mathcal{P}\mathcal{L}_0A(\Gamma) + \int_{t'}^t d\tau\, \mathcal{U}(\tau,t_0)\mathcal{P}\mathcal{L}_0\mathcal{Q}\mathcal{G}(t-\tau)\mathcal{L}_0A(\Gamma) + \mathcal{U}(t',t_0)\mathcal{Q}\mathcal{G}(t-t')\mathcal{L}_0A(\Gamma) + \mathcal{U}(t,t_0)\mathcal{L}_1(t)A(\Gamma) + \int_{t'}^t d\tau\, \mathcal{U}(\tau,t_0)\mathcal{L}_1(\tau)\mathcal{Q}\mathcal{G}(t-\tau)\mathcal{L}_0A(\Gamma). \tag{15}$$

To make use of this expression one needs to find a suitable projection operator. We use a linear Mori projection operator of the form

$$\mathcal{P}X(\Gamma) := \frac{(X,A)}{(A,A)}A(\Gamma), \tag{16}$$

with

$$(X,Y) := \int d\Gamma\, \rho_0(\Gamma)X(\Gamma)Y(\Gamma). \tag{17}$$

Here, $\rho_0(\Gamma)$ is the phase-space distribution at time t_0, which needs to be stationary under the dynamics $\mathcal{L}_0$, but which is

not stationary under $\mathcal{L}_{\rm tot}(t)$. Using this projection operator, Eq. (15) becomes

$$\frac{dA(t;\Gamma)}{dt} = cA(t;\Gamma) - \int_{t'}^{t} d\tau\, K(t-\tau)A(\tau;\Gamma) + \eta(t,t';\Gamma) + \mathcal{U}(t,t_0)\mathcal{L}_1(t)A(\Gamma) + F_{\rm NER}(t,t';\Gamma), \tag{18}$$

with

$$c := \frac{(\mathcal{L}_0 A, A)}{(A,A)}, \tag{19}$$

$$K(t) := -\frac{(\mathcal{L}_0 \mathcal{Q}\mathcal{G}(t)\mathcal{L}_0 A, A)}{(A,A)}, \tag{20}$$

$$\eta(t,t';\Gamma) := \mathcal{U}(t',t_0)\mathcal{Q}\mathcal{G}(t-t')\mathcal{L}_0 A(\Gamma), \tag{21}$$

$$F_{\rm NER}(t,t';\Gamma) := \int_{t'}^{t} d\tau\, \mathcal{U}(\tau,t_0)\mathcal{L}_1(\tau)\mathcal{Q}\mathcal{G}(t-\tau)\mathcal{L}_0 A(\Gamma). \tag{22}$$

Note that the drift c does not contain $\mathcal{L}_1(t)$. If we set $t' = t_0$ [and hence $\mathcal{U}(t',t_0) = 1$], the memory kernel $K(t)$ and the fluctuating force $\eta(t,t_0;\Gamma)$ do not depend on $\mathcal{L}_1(t)$, either. Thus, they are defined exactly as in the case of stationary dynamics under $\mathcal{L}_0$. The memory kernel can then be written in the form

$$K(t) = \frac{(\mathcal{Q}\mathcal{G}(t)\mathcal{L}_0 A, \mathcal{Q}\mathcal{L}_0 A)}{(A,A)} \tag{23}$$

$$= \frac{\langle \eta(t,0;\Gamma)\eta(0,0;\Gamma)\rangle}{(A,A)}, \tag{24}$$

i.e., the FKT still holds true.

If now the observable of interest is a component of the momentum of one particle, $A(\Gamma) = p_z$, and the Liouvillian $\mathcal{L}_1(t)$ is an external force which acts on this degree of freedom, e.g., $\mathcal{L}_1(t) = F_{\rm ext}(t,z)\partial_{p_z}$, Eq. (18) turns into

$$\frac{dp_z(t;\Gamma)}{dt} = -\int_0^t d\tau\, K(t-\tau)p_z(\tau;\Gamma) + \eta(t,0;\Gamma) + F_{\rm ext}(t,z(t)) + F_{\rm NER}(t,0;\Gamma), \tag{25}$$

where K and η only contain the propagator $\mathcal{L}_0$, while the variable p_z is propagated by $\mathcal{L}_0 + \mathcal{L}_1$. We have thus succeeded in constructing an exact generalized Langevin equation for a particle subjected to an external force, which contains the same memory kernel and fluctuating force as the generalized Langevin equation for the particle without the external force, Eq. (2). However, compared to Eq. (4) there is one additional term: the nonequilibrium response force $F_{\rm NER}(t,0;\Gamma)$. This force encodes the nonequilibrium response of the solvent to the driven particle. In general, it will depend on time and on the history of the process, and, as we will show below, it cannot be absorbed in the fluctuating force.

If a (quasi)stationary distribution is reached under sufficiently weak driving, one might use known approximations for the steady distribution to determine statistical properties of $F_{\rm NER}(t,0;\Gamma)$ [15]. However, in all generality this is not possible and thus we determine $F_{\rm NER}(t,0;\Gamma)$ numerically. To this end we make use of the fact that the functions $K(t)$ and $\eta(t,0;\Gamma)$ in the stationary GLE, Eq. (2), and in Eq. (25) are identical. We first run equilibrium molecular dynamics simulations of a microscopic N-particle system, "measure" p_z of one labeled particle, and compute the memory kernel of its effective equation of motion by means of the method described in Ref. [16]. The fluctuating forces for each trajectory then follow from Eq. (2). Then we run simulations with an external force applied to the particle for the exact same starting configurations and calculate $F_{\rm NER}(t,0;\Gamma)$ via Eq. (25).

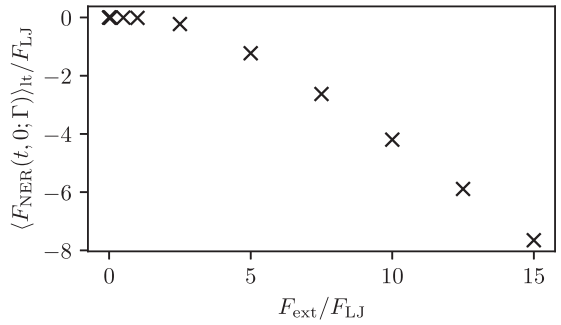

FIG. 1. The long-time ensemble average of the NER force, $\langle F_{\rm NER}(t,0;\Gamma)\rangle_{\rm lt}$, as a function of the external force.

The simulations contain 1024 particles interacting via a Lennard-Jones potential. We use Lennard-Jones units and define a Lennard-Jones force $F_{\rm LJ} := m_{\rm LJ}\sigma_{\rm LJ}/\tau_{\rm LJ}^2$, where $m_{\rm LJ}$ is the mass of the bath particles. In this Letter, we restrict the discussion to the case of a constant force applied to one particle for times $t > 0$ acting in the z direction $F_{\rm ext}(t,z(t)) = F_{\rm ext}\theta(t)$. In all our simulations, we found $F_{\rm NER}(t,0;\Gamma)$ to be nonzero.

If there is a force missing in the Langevin equation, under which nonequilibrium conditions do BD simulations and MD simulations with a Langevin thermostat work? After all, these methods are widely used for systems under external forces (see, e.g., BD simulations of colloidal particles in optical traps [17], Langevin MD simulations of proteins pulled through pores [18], or targeted MD simulations in the context of biomolecular modeling [19]). One conjecture could be that the stochastic interpretation of the fluctuating force absorbs the effect, i.e., when the deterministic fluctuating force η is replaced by a noise, perhaps the resulting dynamics is the same as if $\zeta(t,0;\Gamma) := \eta(t,0;\Gamma) + F_{\rm NER}(t,0;\Gamma)$ were replaced by noise of the same statistical properties. For this to hold, the sum $\zeta(t,0;\Gamma)$ must exhibit the same statistical properties as $\eta(t,0;\Gamma)$ itself. We now check if this conjecture holds, i.e., if $\langle\zeta(t,0;\Gamma)\rangle = 0$ and if $\zeta(t,0;\Gamma)$ fulfills the FKT. Since the ensemble average is linear and $\langle\eta(t,0;\Gamma)\rangle = 0$, it suffices to check if $\langle F_{\rm NER}(t,0;\Gamma)\rangle$ vanishes. For all external forces considered here, we see that the ensemble average of the NER force converges to a constant. Figure 1 shows this limit, calculated by taking the time average of $\langle F_{\rm NER}(t,0;\Gamma)\rangle$ for large times ($3 \leqslant t/\tau_{\rm LJ} \leqslant 25$). There is a crossover point when the external force has the same magnitude as the fluctuating forces, beyond which $\langle F_{\rm NER}(t,0;\Gamma)\rangle$ decreases linearly. Since the external force was applied in positive z direction, this plot also shows that, on average, the NER force counteracts the external force, making it relevant for the calculation of friction. Next, we test if the FKT holds true for $\zeta(t,0;\Gamma)$, cf. Fig. 2. The correlation functions of $\zeta(t,0;\Gamma)$ are calculated in the long-time limit where the ensemble average of $\zeta(t,0;\Gamma)$ has reached its plateau. For small external forces the FKT

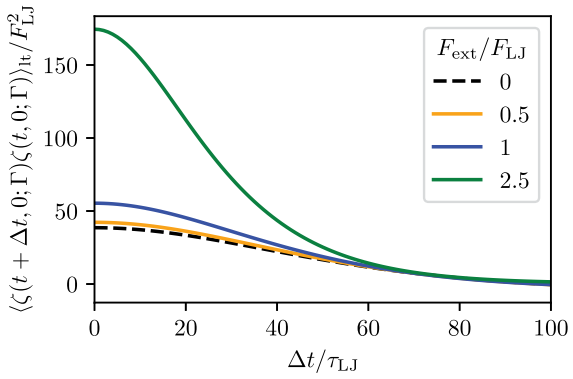

FIG. 2. The autocorrelation function of $\zeta(t,0;\Gamma)$ vs the time difference Δt for different constant external forces.

seems to be fulfilled. However, already at $F_{\rm ext} = F_{\rm LJ}$ a clear deviation can be seen.

We conclude that, in general, the nonequilibrium response cannot be absorbed in the noise by a simple rescaling of the temperature, because the effect of the collisions with the bath particles does not have a zero average.

If $F_{\rm NER}$ cannot be neglected in general, are there limits in which $F_{\rm NER}$ becomes irrelevant? By definition [cf. Eq. (22)] $F_{\rm NER}(t,0;\Gamma)$ vanishes for $t \to 0$ or $F_{\rm ext}(t,z(t)) \to 0\ \forall\, t$. Hence, Eq. (25) allows to analyze an expansion of the GLE around the well-understood stationary case without driving. The fact that $\langle F_{\rm NER}(t,0;\Gamma)\rangle$ vanishes faster than linearly with $F_{\rm ext}$ (as seen in the numerical analysis above) can be derived rigorously:

$$\langle F_{\rm NER}(t,0;\Gamma)\rangle = \int d\Gamma\, \rho_0(\Gamma) \int_0^t d\tau\, \mathcal{U}(\tau,0) F_{\rm ext}(\tau) \cdot \partial_p \eta(t-\tau,0;\Gamma) \tag{26}$$

$$= \int_0^t d\tau F_{\rm ext}(\tau) \int d\Gamma\, \rho_0(\Gamma) \frac{\beta p}{m} \eta(t-\tau,0;\Gamma) + \mathcal{O}\big(F_{\rm ext}^2\big) \tag{27}$$

$$= \mathcal{O}\big(F_{\rm ext}^2\big). \tag{28}$$

From the first to the second line we use that $\mathcal{U}(\tau,0) = \exp(\tau\mathcal{L}_0) + \mathcal{O}(F_{\rm ext})$, bring the time-evolution operator over to the phase-space density, and note that $\exp(-\tau\mathcal{L}_0)\rho_0(\Gamma) = \rho_0(\Gamma)$ because we start out from a stationary distribution. Further, we do an integration by parts to also bring the partial derivative with respect to p over to $\rho_0(\Gamma)$. Here, we exploit that we start out from a canonical distribution to calculate the derivative of $\rho_0(\Gamma)$. In the last step we use that the fluctuating force is orthogonal to the observable p by construction. For the system studied here, we can see from symmetry considerations that $\langle F_{\rm NER}(t,0;\Gamma)\rangle$ has to be an odd function of $F_{\rm ext}$ and thus, $\langle F_{\rm NER}(t,0;\Gamma)\rangle = \mathcal{O}(F_{\rm ext}^3)$.

If we are interested in the onset behavior of $\langle F_{\rm NER}(t,0;\Gamma)\rangle$ as a function of time, we can do a similar analysis as above where we use the series expansions of both $\mathcal{U}(\tau,0)$ and $\eta((t-\tau,0;\Gamma))$. By doing so we find from simple algebraic manipulations and symmetry considerations that

$$\langle F_{\rm NER}(t,0;\Gamma)\rangle = \mathcal{O}(t^6). \tag{29}$$

We see that $\langle F_{\rm NER}(t,0;\Gamma)\rangle$ is strongly suppressed at short times but reaches a nonnegligible plateau within a few Lennard-Jones times (cf. Fig. 1).

Thus, BD and MD with Langevin thermostats can statistically reproduce the first moment of the true dynamics for small perturbations out of equilibrium because the statistical properties of the bath resemble those of an equilibrium fluctuating force.

Note that in case the external force varies with time, $F_{\rm NER}$ does not necessarily reach a stationary limit and the dependence of $F_{\rm NER}$ on the mass ratio and on the magnitude of the force will be more complex. Recent work by Español on the GENERIC framework under external forcing implies that even in the Markovian limit the dynamics is nontrivial [20].

We conclude that the application of an external driving force produces an additional force on the level of the Langevin equation. This additional force, which encodes the nonequilibrium response of the solvent, is in general nonlocal in time. Its statistics is different from the statistics of the fluctuating force. It is probably possible to measure this force in single molecule pulling experiments or in experiments on tracer particles driven through complex liquids following the same protocol that we applied in our simulations. Considering simulations, we stress that the application of a Langevin thermostat to MD simulations of particles which are not subject to external forces is, of course, perfectly fine, as $F_{\rm NER}$ does not appear there. However, the application to more complex systems is less trivial. Regarding simulations of interacting particles immersed in a solvent, the impact of the potential of mean force on the structure of the Langevin equation has recently been discussed elsewhere [21–23]. The statement made in this Letter refers to systems under external driving such as, e.g., particles in optical traps or biomolecules in constrained MD simulations. Here, if one wishes to obtain quantitative results, care is required. In general, it is misleading to interpret the Langevin equation as a Newtonian equation of motion.

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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