Interacting Brownian dynamics in a nonequilibrium particle bath

Stefano Steffenoni,[1,2,*] Klaus Kroy,[2,†] and Gianmaria Falasco[2,‡]
[1]*Max Planck Institute for Mathematics in the Sciences, Inselstrasse 22, 04103 Leipzig, Germany*
[2]*Institut für Theoretische Physik, Universität Leipzig, Postfach 100 920, D-04009 Leipzig, Germany*
(Received 21 October 2016; published 28 December 2016)

We set up a mesoscopic theory for interacting Brownian particles embedded in a nonequilibrium environment, starting from the microscopic interacting many-body theory. Using nonequilibrium linear-response theory, we characterize the effective dynamical interactions on the mesoscopic scale and the statistics of the nonequilibrium environmental noise, arising upon integrating out the fast degrees of freedom. As hallmarks of nonequilibrium, the breakdown of the fluctuation-dissipation and action-reaction relations for Brownian degrees of freedom is exemplified with two prototypical models for the environment, namely active Brownian particles and stirred colloids.

DOI: 10.1103/PhysRevE.94.062139

I. INTRODUCTION

The notion of Brownian motion refers to the thermal fluctuations of some mesoscopic particles in contact with a bath of smaller particles. Colloidal beads dissolved in a simple fluid are the historical paradigm. But the concept generalizes to any slow mesoscopic degrees of freedom in contact with a bath of fast microscopic degrees of freedom. In fact, there are wide-ranging applications of the basic theme outside the realm of physics [1]. The essential feature is a scale separation between the Brownian and bath degrees of freedom that allows for some systematic coarse-graining of an otherwise intractable many-body system. A convenient approach to formalize this seminal insight is via (generalized) Langevin equations, which can be formulated for a wide variety of phenomena and have helped to rationalize a range of interesting phenomena from long-time tails [2] to critical fluctuations [3]. They have therefore become a prevalent tool in the quantitative description of soft matter and, more generally, noisy systems.

Even though there exist systematic derivations of such mesoscopic equations of motion from an underlying microscopic many-body Hamiltonian through the elimination of the fast degrees of freedom [4–6], one eventually typically appeals to equilibrium statistical mechanics in order to make the formal expressions practically useful. Namely, to bypass the explicit solution of the microscopic dynamic equations, the "noise" fluctuations that agitate the mesoscopic degrees of freedom are assigned a weight in accordance with Boltzmann's principle [7]. By construction, their correlations then satisfy detailed balance in the form of a fluctuation-dissipation theorem (FDT) [8]. This implies, in particular, that they induce mesoscopic correlations in accordance with equipartition. Moreover, the average mesoscopic dynamics is found to be a gradient flow in a convex free-energy landscape. Being derived by such a (thermodynamic) potential, the mean effective interactions of the Brownian degrees of freedom themselves obey the action-reaction principle. In other words, in equilibrium stochastic thermodynamics, the symmetries holding on the microscopic level can essentially be lifted up to the mesoscopic scale. The theory remains valid even when some of these mesoscopic degrees of freedom are externally driven out of equilibrium, as long as local detailed balance persists [9,10], i.e., under the assumption that the source of nonequilibrium does not appreciably affect the (many) bath degrees of freedom. For this reason, the concept of a Brownian scale separation, as embodied in the Langevin equation, has played a central role in the development of a framework of stochastic thermodynamics that reaches out to conditions far from equilibrium [11,12] and in the study of nonequilibrium fluctuation and work relations [13].

In contrast, none of the above symmetry properties generally survives on the Brownian scale if the bath itself is driven out of equilibrium. Not only is the detailed balance of the Brownian degrees of freedom then lost, but also equipartition gives way to a more complex energy partition rule [14], stochastic forces are no longer of gradient-type [15], and the action-reaction principle is violated [16]. In soft matter physics, one finds many examples for interacting probes in nonequilibrium baths. One may naturally think of a suspension of colloids immersed in a nonequilibrium solvent, such as a sheared fluid [17]; a granular [18], glassy [19], or active-particle suspension [20]; or even the cytoplasm of a living cell [21]. It would certainly be of great interest to establish a self-contained coarse-grained description for the colloids in such situations. Yet, the usual equilibrium arguments invoked in the construction of a coarse-grained Langevin description are not any more applicable. So the reduced stochastic description (assuming it still exists) must be found by other means, in the worst case by *explicitly* integrating out the dynamics of the nonequilibrium environment. It should go without saying that, for scientifically or technologically interesting systems, this is almost always an impossible task.

There is thus great interest in defining suitable conditions and finding general approximate methods [22,23] that allow for reliable and useful predictions on the Brownian scale, even if the microscopic degrees of freedom of the environment are driven far from equilibrium. Among other things, such methods should enable us to infer the key properties of the nonequilibrium environment from the observed mesoscopic dynamics. Ideally, they should moreover help to unravel the

*stefano.steffenoni@mis.mpg.de
†klaus.kroy@itp.uni-leipzig.de
‡falasco@itp.uni-leipzig.de

formal structure of the coarse graining, such that we can identify the mechanisms underlying the emergent violations of the detailed-balance and action-reaction principles, which are not always straightforwardly discernible on the mesoscopic scale. Finally, this would allow the logical chain of arguments used to infer environmental conditions from Brownian dynamics to be reversed, namely to tailor some desired mesoscopic properties by a fine-tuning of the nonequilibrium driving of the microstatistics of the environment, with some obvious technological implications.

A good candidate for such an approximate method for bypassing the integration of the microscopic dynamics is suggested by the theory of Brownian motion itself. If the "fast" bath degrees of freedom of some Brownian system themselves admit a coarse-grained description by a mesoscopically driven (generalized) Langevin theory routed in its own equilibrium bath, the resulting theory fulfills all of the above requirements. An example for a successful implementation of such a scheme is provided by the theory of nonequilibrium fluctuating hydrodynamics [24], on which theories of Brownian dynamics in nonequilibrium baths can be based [25,26].

In the following, we pursue this idea in a slightly more abstract and fully particle-based framework, i.e., without appealing to a hydrodynamic limit for the environment degrees of freedom. We do assume, instead, that the environment is made up of some sort of particles that evolve according to some driven Markovian stochastic dynamics enjoying local detail balance. In contrast to standard Brownian dynamics, we thus do not require a direct buffering of the probe degrees of freedom by some equilibrium thermal reservoir but only an indirect one, mediated by the nonequilibrium environment (cf. Fig. 1). Technically, we employ nonequilibrium linear-response theory [27–35] to derive a Langevin equation for the interacting probe particles that we assume to be weakly coupled to the interacting many-body system acting as the environment. Following [36,37], we then go beyond a merely static description that would only account for systematic probe interactions induced by the nonequilibrium environment, such as nonequilibrium depletion forces [38,39]. We explicitly look for the fluctuations of such induced forces around their average values. In Sec. II, we obtain formal expressions for these fluctuating forces, the friction, and the noise statistics. When the driving is off, we retrieve the expected detailed balance condition connecting the noise correlation to the friction memory kernel. But we can also analyze how this relation changes when the environment is driven (far) out of equilibrium and quantify the violations of detailed balance and the reciprocal relations in terms of both excess dynamical activity [40–43] and probability currents. The latter result in the lack of an action-reaction principle for the induced probe interactions [16,17,44,45]. Section III exemplifies the theoretical scheme with the help of two paradigmatic examples that can explicitly be worked out. First, we treat analytically a single probe linearly coupled to a fluid of self-propelled particles. This toy model clearly displays the breakdown of detailed balance and allows us to touch on the scope of the notion of effective temperature. Secondly, we employ Brownian dynamics simulations to analyze the effective friction forces induced between two probes suspended in a driven fluid. The numerical evaluation of our general analytic expressions for the time-dependent friction matrix nicely reveals the expected violations of the action-reaction principle, as well as the possible appearance of negative mobility. Finally, in Sec. IV, we conclude with a summary and an outlook.

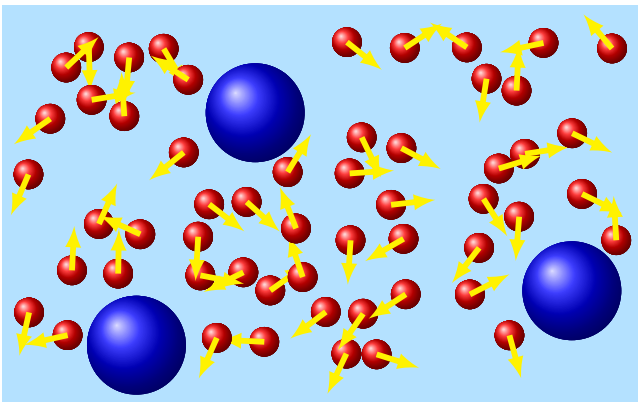

FIG. 1. Schematic representation of the three-level scheme employed by our theory: the probes (blue) representing the system are embedded in a nonequilibrium environment, e.g., a fluid of smaller driven particles (red), which are in contact with a stochastic equilibrium thermal bath (light blue).

II. GENERAL THEORY

We consider a d-dimensional system made up of M probe particles, with mass m_i and positions Q_i, which interact with an environment composed of $N \gg 1$ degrees of freedom denoted x_k. The environment is in contact with an equilibrium bath at inverse temperature β. The probes obey Newton's equation of motion,

$$m_i \ddot{Q}_i = K_i(Q_i) + g_i(\{x_k\}, Q_i), \tag{1}$$

where $g_i \equiv -\lambda \partial_{Q_i} U_i(\{x_k\}, Q_i)$ is the interaction force between the probe i and the environment, with λ a small dimensionless parameter. All the other forces are incorporated in K, which are (optional) direct interactions between the probes and additional external ones. Their specific form is irrelevant in the following. They are only required to be sufficiently confining so as to allow for a unique stationary state. Throughout the text, we use the shorthand $\{\ldots_k\}$ to denote the entire set of degrees of freedom labeled by k. We assume that the environment evolves according to a Markovian stochastic dynamics, enjoying local detail balance. Hence, with respect to standard approaches, we lift such a condition from the dynamics of the system to that of the environment. For concreteness, we can think of the overdamped Langevin equations

$$\dot{x}_k = \mu F - \lambda \mu \sum_{i=1}^{M} \partial_{x_k} U_i + \sqrt{2\mu/\beta}\xi_k. \tag{2}$$

Here $F(\{x_k\})$ consists of interparticle potential forces $-\partial_{x_k} V(\{x_k\})$, and external ones that may contain a nonpotential driving $f(\{x_k\})$ setting the environment out of equilibrium. The ξ's are centered Gaussian noises, white and uncorrelated.

Let $\{Y_i\}$ be the set of average positions around which the probes fluctuate as a consequence of the interactions with the

environment. Here we are concerned with the fluctuations induced by the presence of the environment, for which we seek a reduced description. Namely, we aim at integrating out of (1) the environment coordinates by averaging the probe-environment coupling with the appropriate distribution for x_k. We expect noise and friction to emerge in this process, together with indirect forces between the probes, mediated by the environment. Toward that end, we rewrite Eq. (1) as

$$m_i \ddot{Q}_i = K_i + \langle g_i \rangle + \eta_i, \tag{3}$$

where we split the environment-probe coupling into a systematic part $\langle g_i \rangle$ and a random contribution $\eta_i \equiv g_i - \langle g_i \rangle$. The former is defined as the mean force exerted by the environment on probe i, and it reads

$$\langle g_i(\{x_k\}, Q_i) \rangle \equiv \int d\{x_k\} g_i(\{x_k\}, Q_i) \rho(\{x_k\}|\{Q_i\}), \tag{4}$$

with $\rho(\{x_k\}|\{Q_i\})$ the probability density of the environment conditioned by the probes being in positions $\{Q_i\}$. We work under the usual assumptions made in the derivation of Langevin equations, i.e., a small variation of the probe momentum after a single particle-probe interaction (large mass difference), and a weak coupling between probes and environment. Under these conditions, the fluctuations of probe i around the preferred state Y_i are small, and its force on the whole environment can be expanded to linear order in the displacement from Y_i:

$$\lambda \sum_{k=1}^{N} \partial_{x_k} U_i = \lambda \sum_{k=1}^{N} \partial_{x_k} U_i \Big|_{Q_i = Y_i} - [Q_i(t) - Y_i] \sum_{k=1}^{N} \partial_{x_k} g_i \Big|_{Q_i = Y_i}. \tag{5}$$

Here it is useful to regard g_i as an external potential perturbing the environment, modulated in time via the protocol $Q_i(t) - Y_i$. In view of (5), it is then natural to express the conditional average (4) in terms of unperturbed averages $\langle \cdots \rangle^0$, corresponding to all probes sitting in the mean positions $\{Y_i\}$. To do so, we make use of the response theory for perturbations about nonequilibrium states. The linear-response formula in general reads [46]

$$\langle \mathcal{A}(t) \rangle = \langle \mathcal{A}(t) \rangle^0 + \frac{\beta}{2} \sum_j \int_{t_0}^{t} ds\, h_j(s) \times \left(\frac{d}{ds} \langle \mathcal{B}_j(s); \mathcal{A}(t) \rangle^0 - \langle L\mathcal{B}_j(s); \mathcal{A}(t) \rangle^0 \right), \tag{6}$$

where $\mathcal{A}$ is the observable of interest, and $\mathcal{B}_j$ are the perturbation potentials switched on at time t_0 and modulated in time through the protocol $h_j(s)$. The operator L and the average $\langle \ldots ; \ldots \rangle^0$ stand for, respectively, the backward generator of the unperturbed dynamics and the connected average with respect to it. In (6) the first integrand is the usual correlation of the observable with the entropy production, as appears in the Kubo formula. The second one is a frenetic contribution that contains the excess dynamical activity, $L\mathcal{B}_j$, caused by the perturbation. In equilibrium, they make equal contributions [34]:

$$\frac{d}{ds} \langle \mathcal{B}_j(s); \mathcal{A}_i(t) \rangle^{\mathrm{eq}} = -\langle L\mathcal{B}_j(s); \mathcal{A}_i(t) \rangle^{\mathrm{eq}}. \tag{7}$$

Here we are interested in the response of $g_i(\{x_k\}, Q_i)$ to the perturbations in (5). Hence, with the identifications $\mathcal{A} = g_i$, $\mathcal{B}_j = g_j$, and $h_j = Q_j - Y_j$, (6) becomes

$$\langle g_i(t) \rangle = \langle g_i \rangle^0 + \sum_{j=1}^{M} \frac{\beta}{2} \int_{t_0}^{t} ds [Q_j(s) - Y_j] \times \left[\frac{d}{ds} \langle g_j(s); g_i(t) \rangle^0 - \langle L g_j(s); g_i(t) \rangle^0 \right], \tag{8}$$

where L, the backward generator of the unperturbed dynamics of the environment, reads for (2)

$$L = \mu \sum_{k=1}^{N} \left[F_k \partial_{x_k} - \lambda \sum_{i=1}^{M} \partial_{x_k} U_i \Big|_{Q_i = Y_i} \partial_{x_k} + \frac{1}{\beta} \partial_{x_k}^2 \right]. \tag{9}$$

The summands in (8) are the forces due to the linearized fluctuations of the probes around their preferred states. Assuming that the environment was put in contact with the probes at time $t_0 = -\infty$, so that no correlation with the initial conditions is retained, an integration by parts yields

$$\langle g_i(t) \rangle = \langle g_i \rangle^0 + \sum_{j=1}^{M} \left[G_{ij}(t) - \int_{-\infty}^{t} ds\, \zeta_{ij}(t-s) \dot{Q}_i(s) \right]. \tag{10}$$

Here we defined the memory kernel

$$\zeta_{ij}(t-s) \equiv \frac{\beta}{2} \left(\langle g_j(s); g_i(t) \rangle^0 - \int_{-\infty}^{s} du \langle L g_j(u); g_i(t) \rangle^0 \right), \tag{11}$$

which enters both the friction and the statistical forces mediated by the environment,

$$G_{ij}(t) \equiv [Q_j(t) - Y_j] \zeta_{ij}(0), \tag{12}$$

including the "self-interaction" ($i = j$) and the forces between different probes ($i \neq j$).

Equation (12) establishes the connection between the friction kernel and the fluctuating statistical force, namely

$$\partial_{Q_j} G_{ij} = \zeta_{ij}(0). \tag{13}$$

For $i \neq j$, Eq. (13) relates environment-mediated interactions to cross-friction between probes. It was proposed by De Bacco *et al.* [47] for equilibrium systems arguing on the basis of Onsager's regression principle. Here we gave a formal proof of this relation that extends its validity to nonequilibrium states.

In equilibrium, where averages are denoted $\langle \cdots \rangle^{\mathrm{eq}}$, the frenetic contribution can be eliminated in favor of the entropic term according to (7),

$$\langle g_j(s); g_i(t) \rangle^{\mathrm{eq}} = -\int_{-\infty}^{s} du \langle L g_j(u); g_i(t) \rangle^{\mathrm{eq}}. \tag{14}$$

We thus retrieve that the friction kernel is a symmetric matrix,

$$\zeta_{ij}(t-s) = \beta \langle g_j(s); g_i(t) \rangle^{\mathrm{eq}} = \zeta_{ji}(t-s), \tag{15}$$

since correlations are functions of $|t-s|$ only, thanks to time-reversal invariance. The symmetry (15) translates into the condition $\partial G_{ij}/\partial Q_j = \partial G_{ji}/\partial Q_i$, which suffices to make g_i derive from an effective (thermodynamic) potential $\mathcal{F}(\{Q_i\})$. That such a potential is the Helmholtz free energy of the environment,

$$\mathcal{F} \equiv -\frac{1}{\beta}\ln\int d\{x_k\} e^{-\beta(\lambda\sum_{i=1}^{N}U_i+V)}, \tag{16}$$

is easily seen by introducing the Boltzmann factor in (4):

$$\begin{aligned}\langle g_i\rangle^{\mathrm{eq}} &= -\int d\{x_k\}\lambda\partial_{Q_i}U_i\, e^{-\beta(\sum_{i=1}^{N}U_i+V-\mathcal{F})}\\ &= \frac{1}{\beta}e^{\beta\mathcal{F}}\partial_{Q_i}\int d\{x_k\}e^{-\beta(\lambda\sum_{i=1}^{N}U_i+V)}\\ &= \frac{1}{\beta}e^{\beta\mathcal{F}}\partial_{Q_i}e^{-\beta\mathcal{F}} = -\partial_{Q_i}\mathcal{F}.\end{aligned} \tag{17}$$

This ensues the action-reaction principle for the fluctuating forces among probes. Contrarily, when the environment is driven away from equilibrium, (14) is not applicable in general, as frenetic and entropic terms remain distinct. Hence the reciprocal relations are not satisfied, $\zeta_{ij} \neq \zeta_{ji}$, which implies that the action-reaction symmetry is broken.

Now we turn to the random part of the interaction,

$$\eta_i \equiv g_i(\{x_k\},Q_i) - \langle g_i(\{x_k\},Q_i)\rangle. \tag{18}$$

It has zero mean by definition, and its two-times correlation is obtained again by application of the response formula (6), with $\mathcal{A} = g_i g_j$,

$$\begin{aligned}\langle\eta_i(t)\eta_j(s)\rangle &= \langle g_i(\{x_k(t)\},Q_i(t));\, g_j(\{x_k(s)\},Q_j(s))\rangle\\ &\simeq \langle g_i(t);g_j(s)\rangle^0.\end{aligned} \tag{19}$$

The weak-coupling approximation allowed us to drop higher orders in λ, so that (19) simplifies to

$$\langle\eta_i(t)\eta_j(s)\rangle = \frac{2}{\beta}\zeta_{ij}(t-s) + \int_{-\infty}^{s} du\big\langle Lg_j(u);g_i(t)\big\rangle^0. \tag{20}$$

In general, the noise correlation depends explicitly on the excess dynamical activity of the environment, Lg_i. Yet, in equilibrium, exploiting again the equality of the frenetic and entropic term, (20) reduces to the FDT,

$$\langle\eta_i(t)\eta_j(s)\rangle^{\mathrm{eq}} = \frac{1}{\beta}\zeta_{ij}(t-s). \tag{21}$$

Out of equilibrium (20) cannot be simplified further in general, and the FDT (21) is evidently broken, resulting in asymmetric noise cross-correlations. Such violation of the FDT appears more transparent if (20) is written in terms of the state velocity of the environment, i.e., the vector

$$v(\{x_k\},\{Q_i\}) \equiv \frac{J(\{x_k\},\{Q_i\})}{\rho^0(\{x_k\}|\{Q_i\})}, \tag{22}$$

with J the probability current of the environment, which vanishes identically in equilibrium. Even though it could be experimentally estimated [48–52], it has been analytically solved only in a few simple situations in which the stationary distribution is known [28,32,53]. From the identity $L = L^* + 2v\cdot\nabla$ [28,46,54], where ∇ is the vector of partial derivatives ∂_{x_k}, and L^* is the adjoint of L—the forward generator of the dynamics of the environment—one can easily prove that

$$\begin{aligned}\langle Lg_i(u);g_j(t)\rangle^0 = &-\frac{d}{du}\langle g_j(u);g_i(t)\rangle^0\\ &+ 2\langle v\cdot\nabla g_j(u);g_i(t)\rangle^0.\end{aligned} \tag{23}$$

Using Eqs. (11), (20), and (23), the broken FDT reads

$$\langle\eta_i(t)\eta_j(s)\rangle = \frac{1}{\beta}\zeta_{ij}(t-s) + \int_{-\infty}^{s} du\langle v\cdot\nabla g_j(u);g_i(t)\rangle^0, \tag{24}$$

where the deviation from the equilibrium Kubo formula appears explicitly.

In general, the noise (18) will not be Gaussian, and thus the two-times correlation is not enough to fully characterize its statistics. Higher moments can be calculated with the same procedure, though, by successive application of the response formula (6) together with the weak-coupling assumption.

Finally, we note that the restriction of time-independent mean states $\{Y_i\}$ can be easily lifted. If, instead, mean time-dependent trajectories $\{Y_i(t)\}$ are taken, our approach still holds with the caveat that the perturbation potentials, $g_i(\{x_k\},Y_i(s))$, now carry an explicit time dependence via $Y_i(t)$ [cf. Eq. (5)]. An extension of the response formula (6) needs to be applied [55], which features $\{Y_i(t)\}$ as a quasistatic protocol, but the remaining procedure is very analogous. Therefore, the theory naturally extends to probes that are, e.g., acted upon by external time-dependent forces, or in direct contact with the equilibrium bath, as well as with the environment.

III. EXAMPLES

In this section, we present two explicative examples. First, we consider a single probe coupled linearly to a fluid of non-interacting self-propelled particles. Equations (11) and (20) are calculated analytically and used to show the breakdown of (21). Second, we show how to extract from Brownian simulations the friction memory kernel of two confined probes immersed in a stirred fluid. We prove numerically the breakdown of the reciprocal relations, that is, the violation of the action-reaction principle for the fluid-mediated forces between the probes.

A. One probe in an active fluid

We consider a two-dimensional system ($d = 2$) in which a single probe under harmonic confinement,

$$K(t) = -\kappa_Q[Q(t)-Y], \tag{25}$$

interacts via a harmonic potential U (strength constant κ) with an environment of active Brownian particles [56]. The latter are not mutually interacting but (internally) driven so that they display a drift velocity of constant magnitude v_0 pointing along the random particle orientation $n_k(t)$, i.e.,

$$\mu F(\{x_k\}) = v_0 n_k(t). \tag{26}$$

Due to rotational Brownian motion, the unit vector $n_k(t)$ diffuses with a persistence time D_r [57]:

$$\langle n_k(t)n_{k'}(s)\rangle = \delta_{kk'}\, e^{-D_r|t-s|}. \tag{27}$$

Therefore, (2) takes the simple form of an Ornstein-Uhlenbeck process with an additional stochastic drift [58]. Thanks to the linearity of the system, the systematic part of the interaction,

$$\langle g\rangle = -\lambda\kappa\sum_{k=1}^{N}(Q-\langle x_k\rangle), \tag{28}$$

as well as the stochastic part,

$$\eta = -\lambda\kappa\sum_{k=1}^{N}(\langle x_k\rangle - x_k), \tag{29}$$

can be expressed analytically in terms of Q and Y only. Indeed, the terms in the large square brackets in (8), corresponding to the response function to a constant force,

$$\frac{N\beta\lambda^2\kappa^2}{2}\Big[\frac{d}{ds}\langle x_k(s);x_k(t)\rangle^0 - v_0\langle n_k(s);x_k(t)\rangle^0 + \lambda\kappa\mu\langle x_k(s);x_k(t)\rangle^0\Big], \tag{30}$$

contain simple correlation functions of the unperturbed Ornstein-Uhlenbeck steady state. From (11) we thus obtain the friction kernel

$$\zeta(t-s) = N\lambda\kappa e^{-\lambda\kappa\mu(t-s)}, \tag{31}$$

showing that dissipation happens on the characteristic time scale that it takes the active particles to relax in the coupling potential U. In contrast, the energy input due to the noise (29) is found to occur on multiple time scales,

$$\langle\eta(s)\eta(t)\rangle = \frac{1}{\beta}\zeta(t-s) + \frac{1}{2}\frac{N\lambda\kappa v_0^2}{(\lambda\kappa\mu)^2 - D_r^2} \times\left[\lambda\kappa e^{-D_r(t-s)} - \frac{D_r}{\mu}e^{-\lambda\kappa\mu(t-s)}\right]. \tag{32}$$

The disparity of the time scales for noise and friction entails the breakdown of the FDT, as predicted by (20). One may try to mend it by introducing an effective temperature [59] via

$$\beta T_{\rm eff}(\tau) = 1 + \frac{\beta v_0^2}{2\mu D_r}\left(1 - \frac{\lambda\kappa\mu}{D_r}e^{-D_r\tau}\right) + O(\lambda^2). \tag{33}$$

Thereby, the FDT (21) is formally restored, albeit with the time-dependent function $T_{\rm eff}(\tau)$ replacing the constant bath temperature $1/\beta$.

The deviation from equilibrium is seen to be governed by the two dimensionless numbers $\beta v_0^2/2\mu D_r$ and $\lambda\kappa\mu/D_r$. For $\lambda\kappa\mu/D_r \to 0$, the temperature renormalization becomes time-independent and independent of the weak-coupling parameter λ—it thus acquires the status of thermodynamic temperature. One can then be justified in saying that the probe acts as an ideal measurement device for the constant effective temperature

$$T_{\rm eff} \sim \beta^{-1} + \frac{v_0^2}{2\mu D_r} \tag{34}$$

of the active fluid itself, which coincides with the known value for a suspension of free active particles [60,61]. The strength of the temperature renormalization is controlled by the Peclét number $v_0(\mu D_r/\beta)^{-1/2}$ that weighs the relative importance of ballistic versus (translational and rotational) diffusive motion [62].

To first order in λ, Eq. (33) exhibits a crossover from a short-time temperature to a long-time temperature. Moreover, $T_{\rm eff}$ can no longer be interpreted as a property of the particle bath alone, but it characterizes its interaction with the embedded probe. In fact, the ratio $\lambda\kappa\mu/D_r$ can be interpreted as a measure for the interference of the coupling potential with the persistence of the active particle motion. We expect this particular feature to carry over to more general (strongly interacting) systems, where it would not be accessible within the weak-coupling formalism, however. The physical picture is that the apparent thermalization at the constant effective temperature (34) takes some finite time to happen. In our toy model, this "equilibration time" is given by the rotational diffusion time of the active particles, i.e., the active motion of the bath particles can only be subsumed into an enhanced fluid temperature once it has lost its orientational persistence. This very plausible condition has been pointed out before (e.g., in [63]), albeit not for the time domain. If (32) is extrapolated to values of the dimensionless coupling strength on the order of 1, the temporal growth of the corresponding effective temperature takes the form

$$\beta T_{\rm eff}(\tau) \overset{\lambda\kappa\mu\approx D_r}{\approx} 1 + \frac{\beta v_0^2}{4\mu D_r}(1+\tau D_r). \tag{35}$$

It may tentatively be interpreted as an indication of the onset of strong interactions and collective effects, such as a clustering of the bath particles around the probe, which would entail a progressive heating of the probe. While quantitatively inaccessible to the weak-coupling formalism, corresponding observations have indeed been made in numerical simulations [60,64].

Summing up, we arrived at the generalized Langevin equation for the probe,

$$M\ddot{Q}(t) = -\kappa[Q(t)-Y] - \int_{-\infty}^{t} ds\,\zeta(t-s)\dot{Q}(s) + \eta(t), \tag{36}$$

where the friction memory kernel and the noise covariance are given by (31) and (32), respectively. Note that, since n_k is not Gaussian in general, (29) is not Gaussian either. Nevertheless, in view of the central limit theorem, the probability distribution of η converges to a normal one for $N \gg 1$, $\{x_k\}$ being independent identically distributed random variables.

B. Two probes in a stirred fluid

We consider a one-dimensional system ($d = 1$) consisting of $M = 2$ probes under harmonic confinement and $N = 100$ fluid particles moving freely in a periodic domain $x_k \in [0, L]$, as sketched in Fig. 2. The fluid is driven out of equilibrium by an external constant force f that induces a net particle current J thanks to the periodic boundary conditions. The fluid particles interact (mutually and with the probes) through the same soft repulsive potential $V \equiv U$, such that they experience the total force

$$F_k = f - \sum_{k'=1}^{N}\frac{\partial V_{k'}(x_k, x_{k'})}{\partial x_k}. \tag{37}$$

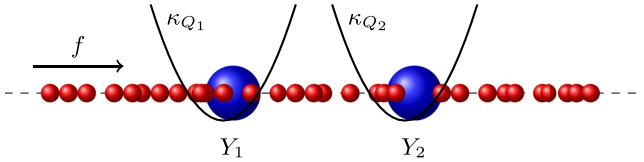

FIG. 2. Schematic illustration of the simulated system, composed of $N+2$ soft spheres in one spatial dimension with periodic boundary conditions. The probes (blue) have average positions $Y_i \sim Y_i^* + \jmath \int_0^\tau d\tau\, \zeta_{ii}(\tau)/\kappa_{Q_i}$ resulting from the balance of the drag force due to the steady current $\jmath$ of fluid particles (red) and to the harmonic confinement with minimum in Y_i^* and stiffness κ_{Q_i} ($i = 1,2$).

In the following, we present results obtained by using the Gaussian potential

$$V_{k'}(x_k, x_k') = e^{-\frac{1}{2\sigma^2}(x_k - x_{k'})^2}, \tag{38}$$

but we have checked numerically that anharmonic potentials lead to qualitatively similar results. In particular, we have calculated the time-dependent entries of the friction kernel ζ_{ij} from formula (11) for various values of the external driving f. This was done by letting the fluid relax from an initial uniform density, fixing the probes in their preferred positions Y_i, and then performing the steady-state averages in (11) over 2×10^4 independent simulation runs of duration $\mathcal{T} = 10^3$.

For $f \to 0$, equilibrium conditions are recovered. The diagonal elements ζ_{ii} of the friction kernel are positive and exhibit a monotonic time dependence. The two off-diagonal elements ζ_{12} and ζ_{21}, which quantify the mutual frictional forces between the probes, coincide. As expected, they are negative and decay to zero at late times. Their negative sign can be understood on the basis of global momentum conservation. For example, consider the drag force that probe 1 exerts on probe 2,

$$F_{1\to 2}^{\text{drag}} = -\int_{-\infty}^{t} ds\, \zeta_{21}(t-s)\dot{Q}_1(s).$$

It is easy to convince oneself that, given the configuration sketched in Fig. 2, a positive velocity $\dot{Q}_1$ will on average cause a positive displacement of the fluid particles surrounding probe 1. Such perturbation spreads along the coordinate axis reaching probe 2, ultimately resulting in a positive momentum transfer $F_{1\to 2}^{\text{drag}} > 0$. This suggests that $\zeta_{21} \leqslant 0$ for all times.

In contrast, with increasing nonequilibrium force $f > 0$, we observe a qualitative modification of the diagonal and nondiagonal elements of ζ_{ij}, as exemplified in Figs. 3 and 4, respectively. The diagonal elements ζ_{ii} develop a nonmonotonic time dependence and eventually turn negative. Physically, this corresponds to a viscoelastic recoiling of the individual probe particles. A more dramatic, genuinely nonequilibrium effect is found for the off-diagonal elements $\zeta_{i\neq j}$. As revealed by Fig. 4, the presence of a nonequilibrium flux in the bath breaks the symmetry of the friction matrix so that $\zeta_{ij} \neq \zeta_{ji}$, with $|\zeta_{21}|$ ($|\zeta_{12}|$) larger (smaller) with respect to equilibrium. Such an effect arises whenever a spatial asymmetry is imposed on top of broken detailed balance. Our periodic system is always spatially asymmetric unless $Y_1 - Y_2 = L/2$. Specifically, in the simulations, the probe

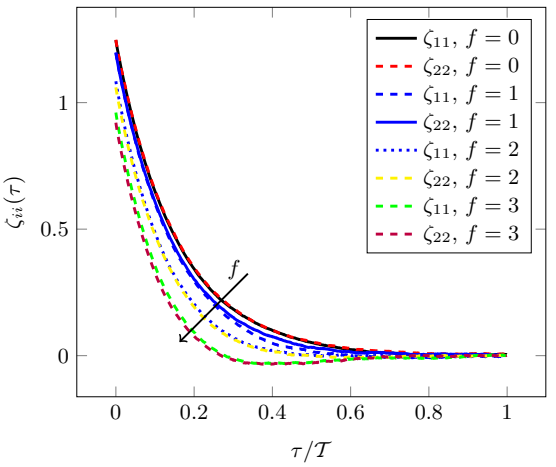

FIG. 3. Diagonal elements of the friction kernel $\zeta_{ij}(\tau)$ as a function of time $\tau = t - s$, obtained by numerical evaluation of (11) in Brownian dynamics simulations, for various values of the nonequilibrium driving force f and $\beta = 1$, $\mu = 1$, and $\sigma = 1$.

reference positions are set to $Y_1 \simeq L/3 < Y_2 \simeq L/2$, and, for convenience, the trap stiffnesses κ_{Q_i} are chosen large enough to make the position Y_i almost coincide with the trap minimum Y_i^*. By increasing L, we checked that interactions with the periodic image particles are negligible. We conclude that global momentum conservation does not hold anymore when the fluid dynamics becomes dissipative. This can be attributed to the asymmetric propagation (due to the current $\jmath$) of fluid perturbations. Namely, downstream propagation is progressively enhanced by increasing f, while upstream propagation is suppressed. As a result, the influence of probe 1 (2) on probe 2 (1) gets stronger (weaker) as we increase the driving. As for the diagonal elements, the sign of $\zeta_{12}(\tau)$ is transiently reversed. More remarkably, for sufficiently large values of f, the response coefficient of probe 2 to a uniform motion of probe 1, namely $-\int_0^\infty d\tau \zeta_{12}(\tau)$, turns negative. In contrast to the mentioned transient elastic recoil embodied in

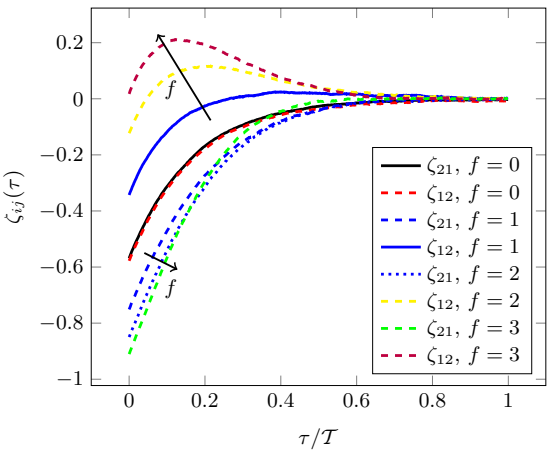

FIG. 4. Off-diagonal elements of the friction kernel $\zeta_{ij}(\tau)$ as a function of time $\tau = t - s$, obtained by numerical evaluation of (11) in Brownian dynamics simulations for various values of the nonequilibrium driving force f and $\beta = 1$, $\mu = 1$, and $\sigma = 1$.

the diagonal terms ζ_{ii}, this kind of "absolute negative mobility" [65–68] is strictly forbidden in equilibrium, where dissipative transport coefficients depend only on the (positive) entropy production but not on the dynamical activity [69].

IV. CONCLUSION

Employing nonequilibrium linear-response theory, we have derived generalized Langevin equations for probe particles interacting with a driven environment. The latter was described by an explicit interacting many-body theory for overdamped colloidal particles representing, e.g., active or sheared colloids. More generally, they can be understood as a set of mesoscopic degrees of freedom. Also, the theoretical framework developed above can be easily adapted to cope with different sources of nonequilibrium (other than the nonconservative force f), such as a nonuniform bath temperature field $\beta(x_k)$.

When only conservative forces are present, our theory correctly reproduces the expected equilibrium properties, i.e., it fulfils the FDT and conforms to Onsager's regression principle relating the fluctuations of statistical forces to the memory kernel. In general, it extends the Langevin approach into the nonequilibrium realm, predicting the violation of the FDT and the action-reaction law for the fluctuating effective forces. The breaking of these dynamical symmetries is traced back to the mismatch between the excess entropy and dynamical activity induced by probe fluctuations around their preferred states, or, equivalently, to the existence of dissipative currents in the environment. We have shown that these phenomena appear already in simple systems, unless special symmetries are present. Namely, noise and friction felt by a single probe in an active medium do not obey the FDT, except if the relaxation time scales of the system and the fluid are properly tuned, in which case a constant effective temperature can be defined. Also, the cross-frictions between two confined probes in a stirred periodic fluid are dissimilar, and they even change sign with respect to equilibrium whenever the probe reference positions break the spatial symmetry.

The theory allows us to obtain quantitative information about the parameters of the environment from measuring average properties of the probes. For example, from (31) and (32)—which are accessible by measuring, e.g., the spectral density of the probe fluctuations in the trap and its response to a small external kick—the values of the relaxation times $\mu\kappa$ and D_r can be inferred. Conversely, one may even speculate that some mesoscopic parameters [e.g., $\zeta_{ij}(0)$] might be fixed at will by properly designing the nonconservative driving. This is feasible in principle since formal procedures are available [70] that determine an appropriate environment dynamics conditioned on prescribed mean values [e.g., those entering (11)].

Finally, a remark on the status of the approximation of weak coupling to the nonequilibrium environment seems warranted. In a particle-based theory such as the one we employed, this approximation is explicitly enforced by introducing a small coupling constant λ. Physically, the appropriate values λ may depend on the average number of bath particles with which the probes interact. This should be clear from the example in Sec. III A, where the limit $N \to \infty$ produces an unphysical divergence of friction and noise strength if λ is not properly scaled. However, in practical applications, the weak coupling is often a dynamical, *emergent*, property resulting from the scale separation between the probe-particle system and the environment. For example, colloidal particles suspended in simple fluids are well described by a linear hydrodynamic theory, although the microdynamics of the fluid molecules is highly nonlinear. This feature is expected to be robust and to survive even far from equilibrium, as long as the driving energy input does not exceed the bath thermal energy [26]. Indeed, the peculiar feature of a time-dependent noise temperature, discovered within the weak-coupling approach mentioned above, was already explicitly demonstrated (and its time dependence analytically computed) in this setting [25].

Recently, new theoretical investigations [71,72] have been spurred by a surge of experimental interest in systems with strongly coupled components, such as in active nonlinear microrheology [73], single-molecule (force spectroscopy) experiments [74], and work extraction from active fluids [75]. Hence, it would be desirable to extend the above analysis to different dynamical descriptions of the environment, i.e., in terms of (hydrodynamic) fields or discrete-state variables. This may provide more versatile formal tools to account more reliably for the weak coupling and to address the strong-coupling problem in a larger variety of stochastic systems.

ACKNOWLEDGMENTS

G.F. and K.K. acknowledge funding by the Deutsche Forschungsgemeinschaft (DFG). S.S. acknowledges funding by International Max Planck Research Schools (IMPRS).

[1] E. Frey and K. Kroy, Ann. Phys. **14**, 20 (2005).
[2] J. A. McLennan, *Introduction to Nonequilibrium Statistical Mechanics* (Prentice Hall, Englewood Cliffs, NJ, 1989), pp. 53–54.
[3] P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).
[4] R. Zwanzig, J. Stat. Phys. **9**, 215 (1973).
[5] K. Kawasaki, J. Phys. A **6**, 1289 (1973).
[6] N. Van Kampen and I. Oppenheim, Physica A **138**, 231 (1986).
[7] L. Onsager and S. Machlup, Phys. Rev. **91**, 1505 (1953).
[8] R. Kubo, Rep. Prog. Phys. **29**, 255 (1966).
[9] S. Katz, J. L. Lebowitz, and H. Spohn, Phys. Rev. B **28**, 1655 (1983).
[10] T. Harada and S.-i. Sasa, Phys. Rev. E **73**, 026131 (2006).
[11] K. Sekimoto, *Stochastic Energetics*, Vol. 799 (Springer-Verlag, Berlin Heidelberg, 2010).
[12] U. Seifert, Phys. Rev. Lett. **95**, 040602 (2005).
[13] U. Seifert, Rep. Prog. Phys. **75**, 126001 (2012).
[14] G. Falasco, F. Baldovin, K. Kroy, and M. Baiesi, New J. Phys. **18**, 093043 (2016).

[15] U. Basu, C. Maes, and K. Netočný, Phys. Rev. Lett. **114**, 250601 (2015).
[16] A. V. Ivlev, J. Bartnick, M. Heinen, C.-R. Du, V. Nosenko, and H. Löwen, Phys. Rev. X **5**, 011035 (2015).
[17] J. Dzubiella, H. Löwen, and C. N. Likos, Phys. Rev. Lett. **91**, 248301 (2003).
[18] R. Candelier and O. Dauchot, Phys. Rev. Lett. **103**, 128001 (2009).
[19] B. Abou and F. Gallet, Phys. Rev. Lett. **93**, 160603 (2004).
[20] D. T. N. Chen, A. W. C. Lau, L. A. Hough, M. F. Islam, M. Goulian, T. C. Lubensky, and A. G. Yodh, Phys. Rev. Lett. **99**, 148302 (2007).
[21] A. W. C. Lau, B. D. Hoffman, A. Davies, J. C. Crocker, and T. C. Lubensky, Phys. Rev. Lett. **91**, 198101 (2003).
[22] H. Spohn, *Large Scale Dynamics of Interacting Particles* (Springer-Verlag, Berlin Heidelberg, 2012).
[23] N. G. Van Kampen, Phys. Rep. **124**, 69 (1985).
[24] J. M. O. De Zarate and J. V. Sengers, *Hydrodynamic Fluctuations in Fluids and Fluid Mixtures*, 1st ed. (Elsevier, Amsterdam, 2006).
[25] G. Falasco, M. V. Gnann, D. Rings, and K. Kroy, Phys. Rev. E **90**, 032131 (2014).
[26] G. Falasco and K. Kroy, Phys. Rev. E **93**, 032150 (2016).
[27] T. Nakamura and S.-i. Sasa, Phys. Rev. E **77**, 021108 (2008).
[28] R. Chetrite, G. Falkovich, and K. Gawędzki, J. Stat. Mech. (2008) P08005.
[29] T. Speck and U. Seifert, Phys. Rev. E **79**, 040102 (2009).
[30] U. Seifert and T. Speck, Europhys. Lett. **89**, 10007 (2010).
[31] J. Prost, J.-F. Joanny, and J. M. R. Parrondo, Phys. Rev. Lett. **103**, 090601 (2009).
[32] G. Verley, K. Mallick, and D. Lacoste, Europhys. Lett. **93**, 10002 (2011).
[33] E. Lippiello, F. Corberi, and M. Zannetti, Phys. Rev. E **71**, 036104 (2005).
[34] M. Baiesi, C. Maes, and B. Wynants, Phys. Rev. Lett. **103**, 010602 (2009).
[35] M. Baiesi, C. Maes, and B. Wynants, J. Stat. Phys. **137**, 1094 (2009).
[36] C. Maes, J. Stat. Phys. **154**, 705 (2014).
[37] C. Maes and S. Steffenoni, Phys. Rev. E **91**, 022128 (2015).
[38] I. Sriram and E. M. Furst, Soft Matter **8**, 3335 (2012).
[39] J. Harder, S. A. Mallory, C. Tung, C. Valeriani, and A. Cacciuto, J. Chem. Phys. **141**, 194901-1/7 (2014).
[40] V. Lecomte, C. Appert-Rolland, and F. van Wijland, Phys. Rev. Lett. **95**, 010601 (2005).
[41] M. Merolle, J. P. Garrahan, and D. Chandler, Proc. Natl. Acad. Sci. (USA) **102**, 10837 (2005).
[42] J. P. Garrahan, R. L. Jack, V. Lecomte, E. Pitard, K. van Duijvendijk, and F. van Wijland, J. Phys. A **42**, 075007 (2009).
[43] C. J. Fullerton and R. L. Jack, J. Chem. Phys. **138**, 224506 (2013).
[44] K. Hayashi and S.-i. Sasa, J. Phys.: Condens. Matter **18**, 2825 (2006).
[45] P. R. Buenzli and R. Soto, Phys. Rev. E **78**, 020102 (2008).
[46] M. Baiesi and C. Maes, New J. Phys. **15**, 013004 (2013).
[47] C. De Bacco, F. Baldovin, E. Orlandini, and K. Sekimoto, Phys. Rev. Lett. **112**, 180605 (2014).
[48] V. Blickle, T. Speck, U. Seifert, and C. Bechinger, Phys. Rev. E **75**, 060101(R) (2007).
[49] C. Battle, C. P. Broedersz, N. Fakhri, V. F. Geyer, J. Howard, C. F. Schmidt, and F. C. MacKintosh, Science **352**, 604 (2016).
[50] J. Mehl, B. Lander, C. Bechinger, V. Blickle, and U. Seifert, Phys. Rev. Lett. **108**, 220601 (2012).
[51] B. Lander, J. Mehl, V. Blickle, C. Bechinger, and U. Seifert, Phys. Rev. E **86**, 030401 (2012).
[52] J. R. Gomez-Solano, A. Petrosyan, S. Ciliberto, R. Chetrite, and K. Gawędzki, Phys. Rev. Lett. **103**, 040601 (2009).
[53] M. Baiesi, S. Ciliberto, G. Falasco, and C. Yolcu, Phys. Rev. E **94**, 022144 (2016).
[54] T. Speck and U. Seifert, Europhys. Lett. **74**, 391 (2006).
[55] C. Maes and K. Netočný, J. Stat. Phys. **154**, 188 (2014).
[56] A. P. Solon, Y. Fily, A. Baskaran, M. E. Cates, Y. Kafri, M. Kardar, and J. Tailleur, Nat. Phys. **11**, 3377 (2015).
[57] B. ten Hagen, S. van Teeffelen, and H. Löwen, Condens. Matter Phys. **12**, 725 (2009).
[58] C. W. Gardiner, *Handbook of Stochastic Methods for Physics, Chemistry and the Natural Sciences*, 3rd ed., Vol. 13 (Springer-Verlag, Berlin, 2004).
[59] L. F. Cugliandolo, J. Phys. A **44**, 483001 (2011).
[60] X. Yang, M. L. Manning, and M. C. Marchetti, Soft Matter **10**, 6477 (2014).
[61] S. C. Takatori and J. F. Brady, Soft Matter **11**, 7920 (2015).
[62] C. Bechinger, R. Di Leonardo, H. Löwen, C. Reichhardt, G. Volpe, and G. Volpe, Rev. Mod. Phys. **88**, 045006 (2016).
[63] A. Solon, M. Cates, and J. Tailleur, Eur. Phys. J. Spec. Top. **224**, 1231 (2015).
[64] E. Fodor, C. Nardini, M. E. Cates, J. Tailleur, P. Visco, and F. van Wijland, Phys. Rev. Lett. **117**, 038103 (2016).
[65] R. Eichhorn, P. Reimann, and P. Hänggi, Phys. Rev. Lett. **88**, 190601 (2002).
[66] A. Ros, R. Eichhorn, J. Regtmeier, T. T. Duong, P. Reimann, and D. Anselmetti, Nature (London) **436**, 928 (2005).
[67] L. Machura, M. Kostur, P. Talkner, J. Łuczka, and P. Hänggi, Phys. Rev. Lett. **98**, 040601 (2007).
[68] P. K. Ghosh, P. Hänggi, F. Marchesoni, and F. Nori, Phys. Rev. E **89**, 062115 (2014).
[69] P. Baerts, U. Basu, C. Maes, and S. Safaverdi, Phys. Rev. E **88**, 052109 (2013).
[70] R. Chetrite and H. Touchette, in *Annales Henri Poincaré* (Springer-Verlag, Berlin Heidelberg, 2015), Vol. 16, pp. 2005–2057.
[71] U. Seifert, Phys. Rev. Lett. **116**, 020601 (2016).
[72] M. Krüger and C. Maes, arXiv:1607.08790.
[73] J. R. Gomez-Solano and C. Bechinger, New J. Phys. **17**, 103032 (2015).
[74] O. Otto, S. Sturm, N. Laohakunakorn, U. F. Keyser, and K. Kroy, Nat. Commun. **4**, 1780 (2013).
[75] R. Di Leonardo, L. Angelani, D. Dell'Arciprete, G. Ruocco, V. Iebba, S. Schippa, M. Conte, F. Mecarini, F. De Angelis, and E. Di Fabrizio, Proc. Natl. Acad. Sci. (USA) **107**, 9541 (2010).