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Enhanced diffusion over a periodic trap by hydrodynamic coupling to an elastic mode

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In many physical systems, degrees of freedom are coupled *via* hydrodynamic forces, even in the absence of Hamiltonian interactions. A particularly important and widespread example concerns the transport of microscopic particles in fluids near deformable boundaries. In such a situation, the influence of elasto-hydrodynamic couplings on Brownian motion remains to be understood. Unfortunately, the temporal and spatial scales associated with the thermal fluctuations of usual surfaces are often so small that their deformations are difficult to monitor experimentally, together with the much slower and larger particle motion at stake. Here, we propose a minimal model describing the hydrodynamic coupling of a colloidal particle to a fluctuating elastic mode, in presence of an external periodic potential. We demonstrate that the late-time diffusion coefficient of the particle increases with the compliance of the elastic mode. Remarkably, our results reveal that, and quantify how: i) spontaneous microscopic transport in complex environments can be affected by soft boundaries – a situation with numerous practical implications in nanoscale and biological physics; ii) the effects of fast and tiny surface deformations are imprinted over the long-term and large-distance colloidal mobility – and are hence measurable in practice.

Brownian motion refers to the random movement of a colloid at equilibrium, induced by its collisions with the molecules of the surrounding fluid. It has played a foundational role in modern statistical physics since its discovery [1] and theoretical description [2, 3] over a century ago. The celebrated experiments of Perrin on colloids in a gravitational field [4] validated further the theoretical description and confirmed the atomic nature of matter. Extending the classical framework of Brownian motion in bulk Newtonian fluids to more realistic systems, such as particles diffusing in complex environments, opens up new challenges for nanoscale and biological physics. One common route is to study Brownian motion in viscoelastic fluids or intracellular media, as typically performed through microrheology [5], which revealed the emergence of anomalous diffusion [6]. Another source of complexity lies in the introduction of topological constraints and boundaries. For instance, a simple-yet-canonical situation where interfaces matter is the diffusion near rigid walls, that has been extensively studied both theoretically and experimentally [7–13]. Besides the practical interest of such a configuration towards the sensing of surface forces [14], one of the key aspects lies perhaps in the apparition of anisotropic and space-dependent mobilities [15] due to the frictional boundary condition at the walls. As a consequence, multiplicative noises emerge, leading to non-Gaussian diffusion and the probability enhancement of rare events [16–20].

However, in most practical situations where molecules or microorganisms diffuse near surfaces, the latter are typically soft. Therefore, the dynamical properties of Brownian particles near deformable boundaries must be

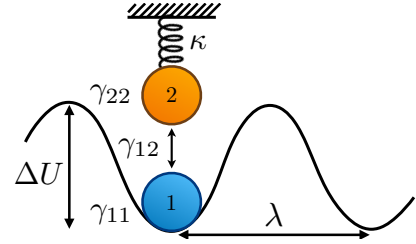


FIG. 1. Schematic of the system. A test Brownian particle (1) is trapped in a periodic energy potential of amplitude ΔU and spatial period λ . It is coupled via a hydrodynamic-like interaction of constant damping coefficient γ_{12} to a thermally-fluctuating elastic mode, represented by another Brownian particle (2) trapped in a harmonic trap of compliance (*i.e.* inverse stiffness) κ . Besides, both particles feel individual hydrodynamic drag forces represented by their constant self-drag coefficients γ_{11} and γ_{22} , and are described within the overdamped Langevin regime.

understood. Along this line of thought, a few studies, mostly theoretical, have addressed the case of tracers diffusing near soft surfaces. Examples include liquid interfaces [21, 22], and fluctuating ones [23]. The case of elastic interfaces was also addressed in the contexts of contactless microrheology [24] and biological membranes [25]. Nevertheless, these works focused on the low-coupling regime, where the particles are distant or point-like, and thus unable to generate rectified flows that lead to surface deformation. Interestingly though, within a purely deterministic context, near-contact elasto-hydrodynamic couplings have been studied in the past decades. Examples include contactless colloidal-probe rheology of soft substrates in normal mode [26, 27], as well as the emergence of lift forces at low Reynolds numbers [28, 29], that were first revealed theoretically [30–34], and then confirmed experimentally [35–39]. The leading-order corrections induced by elasticity to the full mobility tensor

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near a rigid wall [7] were obtained for various geometries and rheologies [40–42], and revealed a zoology of counter-intuitive inertial-like solutions.

In view of the above state of the art, the arising challenge is now to understand the influence of soft boundaries on the spontaneous motion of nearby finite-sized Brownian particles. Molecular-dynamics simulations were performed in such a context [43]. Besides, a recent experimental study indicated that soft colloids diffusing near rigid walls feel transient, non-conservative forces that could be related to an intricate combination of thermal fluctuations, soft boundaries and viscous flows [44]. Finally, at a practical level, a natural question to ask is whether or not the effects of fast and tiny surface fluctuations can be observed over the long times and large distances associated with colloidal diffusion. In this Letter, we thus introduce a minimal, but generic, theoretical model in order to investigate Brownian elastohydrodynamic effects. Specifically, we combine the hydrodynamic coupling of a colloidal particle with an elastic mode [45, 46] to an external periodic trapping [47–50]. We solve the problem both analytically and numerically, and demonstrate that the late-time diffusion coefficient of the particle increases notably with the compliance of the elastic mode.

The model. As depicted in Fig. 1, we consider an overdamped system made of two coupled degrees of freedom: i) a principal degree of freedom $q_1(t)$, corresponding to the spatial position of a test colloid over time t , which is free to explore space while being subject to a periodic trapping potential $\phi(q_1)$; and ii) an auxiliary degree of freedom $q_2(t)$, corresponding to an elastic mode, which is harmonically confined with a potential energy $q_2^2/(2\kappa)$, where κ is the compliance. Thus, the total energy of the system is given by the Hamiltonian

$$H(q_1, q_2) = \phi(q_1) + \frac{q_2^2}{2\kappa}. \quad (1)$$

At thermal equilibrium, the probability density function (PDF) of q_1 and q_2 is given by the Gibbs-Boltzmann distribution

$$P_{\text{eq}}(q_1, q_2) = \frac{\exp[-\beta H(q_1, q_2)]}{Z}, \quad (2)$$

where Z is the normalising partition function, and where $\beta = 1/(k_B T)$ with T the temperature and k_B Boltzmann's constant. The equilibrium statistics of q_1 and q_2 are independent due to the additivity of the Hamiltonian, which implies in particular that q_2 does not affect the equilibrium distribution of q_1 .

The system dynamics is described by the coupled overdamped Langevin equations

$$\gamma_{11} \frac{dq_1}{dt} + \gamma_{12} \frac{dq_2}{dt} = -\phi'(q_1) + \eta_1(t), \quad (3)$$

$$\gamma_{12} \frac{dq_1}{dt} + \gamma_{22} \frac{dq_2}{dt} = -\frac{q_2}{\kappa} + \eta_2(t), \quad (4)$$

where the constants γ_{ij} , with $i, j \in \{1, 2\}$, are the components of the friction tensor γ , which is related to the mobility tensor μ through $\mu = \gamma^{-1}$; and where the functions $\eta_i(t)$ are zero-mean Gaussian white noises, with a correlation function $\langle \eta_i(t) \eta_j(t') \rangle = 2k_B T \delta(t - t') \gamma_{ij}$, where $\langle \cdot \rangle$ is the ensemble average. This choice of noise correlation is self-consistent with the equilibrium distribution of q_1 and q_2 being given by Eq. (2).

Theoretical analysis. In the weakly-compliant limit ($\kappa \ll 1$), one can compute the late-time diffusion coefficient $D^*(\kappa)$ of the colloid using a perturbation analysis at $\mathcal{O}(\kappa)$, together with an exact representation of the effective diffusion coefficient obtained via the Stokes-Einstein relation, and a multiscale analysis [51, 52]. Doing so (see details in SM [53]), we obtain

$$D^*(\kappa) \simeq D^*(0) (1 + \alpha), \quad (5)$$

with

$$\alpha = \beta \kappa \frac{\gamma_{12}^2 \int_0^\lambda dq_1 \phi'(q_1)^2 \exp[\beta \phi(q_1)]}{\gamma_{11}^2 \int_0^\lambda dq_1 \exp[\beta \phi(q_1)]}, \quad (6)$$

and

$$D^*(0) = \frac{k_B T \lambda^2}{\gamma_{11} Z_+ Z_-}, \quad (7)$$

where

$$Z_\pm = \int_0^\lambda dq_1 \exp[\pm \beta \phi(q_1)]. \quad (8)$$

Since $\alpha \geq 0$, the hydrodynamic coupling of the colloid to the elastic mode always increases the late-time diffusion coefficient of the colloid. Moreover, the effect is larger at larger compliance κ . We also see that a non-flat periodic potential $\phi(q_1)$ is required for the effect to exist, within the assumptions of the model.

In the particular case of a sinusoidal potential, where $\phi(q_1) = \Delta U \sin(2\pi q_1/\lambda)$, with ΔU the amplitude of the potential, and λ its spatial period, we have

$$\alpha = 4\pi^2 \epsilon \frac{\gamma_{12}^2 v I_1(v)}{\gamma_{11}^2 I_0(v)}, \quad (9)$$

with $v = \beta \Delta U$, where I_n denotes the order- n modified Bessel function of the first kind, and where we introduced the dimensionless compliance

$$\epsilon = \frac{\kappa k_B T}{\lambda^2}. \quad (10)$$

As such, the relative increase of the late-time diffusion coefficient is larger for smaller λ . This is attributed to the fact that for a smaller λ the coupling to the elastic mode increases the jump rate of the colloid between neighbouring minima, instead of being averaged out as would be the case at large λ . We also see that the relative increase of the late-time diffusion coefficient depends linearly on the

function $vI_1(v)/I_0(v)$, which is a monotonically increasing function of v . This means that the relative magnitude of the effect increases with the depth of the trap – the obvious counterpart being that larger times are then needed to reach the late-time diffusive regime. Finally, the relative magnitude of the effect increases with the relative magnitude γ_{12}/γ_{11} of the hydrodynamic coupling to the elastic mode, as expected.

The multiscale analysis can also be employed in the limit where $\epsilon \rightarrow \infty$, which corresponds to a very compliant elastic mode. In this case, we also find (see details in SM [53]) that the late-time diffusion coefficient is increased relatively to the rigid-case value, and saturates to a maximum value at large ϵ , as

$$D^* \simeq \frac{k_B T \mu_{11} (\mu_{11} \mu_{22} - \mu_{12}^2)}{\lambda^{-2} Z_+ Z_- (\mu_{11} \mu_{22} - \mu_{12}^2) + \mu_{12}^2} + O\left(\frac{1}{\sqrt{\epsilon}}\right). \quad (11)$$

Such a plateau value can be notably higher than the rigid-case value, indicating the potential strength of the effect in practical systems relevant to soft matter and biophysics. Besides, once again, we note that when the periodic potential $\phi(q_1)$ is flat, the late-time diffusion coefficient is not modified by the hydrodynamic coupling to the elastic mode.

Heuristic argument. In order to understand the softness-induced late-time diffusivity enhancement from a physical point of view, we now present a much simpler heuristic argument giving the same result as the rigorous analysis in the small- ϵ limit (see details in SM [53]). The intrinsic time scale $\tau_2 = \kappa \gamma_{22}$ of the stochastic process $q_2(t)$ is assumed to be small with respect to all the other time scales. In this limit, the effective zero-temperature overdamped Langevin equation for $q_1(t)$ is

$$\frac{dq_1}{dt} = -\mu_e(q_1) \phi'(q_1), \quad (12)$$

where we have introduced the dressed mobility

$$\mu_e(q_1) = \frac{1}{\gamma_{11} \left[1 + \frac{\kappa \gamma_{12}^2 \phi''(q_1)}{\gamma_{11}^2} \right]}. \quad (13)$$

The equilibrium marginal PDF in position q_1 is $P_{\text{meq}}(q_1) = (Z_-)^{-1} \exp[-\beta \phi(q_1)]$. Therefore, assuming that the time-dependent marginal PDF $P_1(q_1, t)$ satisfies an effective Fokker-Planck equation, the latter must have the form

$$\frac{\partial P_1(q_1, t)}{\partial t} = \frac{\partial}{\partial q_1} \left\{ \mu_e(q_1) \left[k_B T \frac{\partial P_1(q_1, t)}{\partial q_1} + \phi'(q_1) P_1(q_1, t) \right] \right\}, \quad (14)$$

where the T -independent term on the right-hand side is determined from the zero-temperature dynamics, while the T -dependent term on the right-hand side is constructed to ensure convergence of P_1 to P_{meq} at infinite time. Note that we have implicitly assumed here that the dressed mobility $\mu_e(q_1)$ does not exhibit any temperature dependence.

The dressed diffusion coefficient is given by the Stokes-Einstein relation, $D_e(q_1) = k_B T \mu_e(q_1)$, which differs

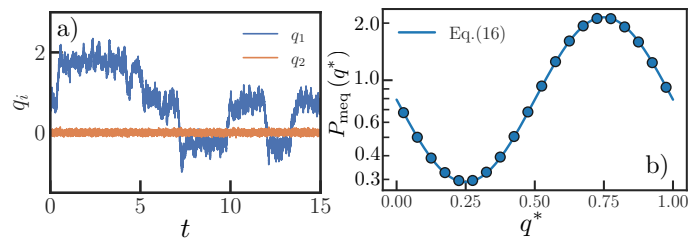


FIG. 2. a) Typical trajectories $q_i(t)$ of the test particle ($i = 1$, blue line) and the elastic mode ($i = 2$, orange line), simulated by numerically solving Eqs. (3) and (4), for $\mu_{11} = 1.0$, $\mu_{12} = 0.5$, $\mu_{22} = 0.5$, $\Delta U = 1.0$, $\lambda = 1.0$, $\kappa = 1.0 \times 10^{-3}$, $\Delta t = 1.0 \times 10^{-4}$, and $k_B T = 1.0$. b) Late-time marginal PDF P_{meq} for the periodised position q^* , given by $q_1 = q^* \bmod(\lambda)$, of the test particle, obtained by binning 10^2 trajectories of 10^7 points. The symbols are the results from the numerical simulations. The solid blue line corresponds to the Boltzmann distribution given in Eq. (16).

from the bare (or “microscopic”) diffusion coefficient $D_m = k_B T / \gamma_{11}$ of the colloid. Interestingly, the spatial dependence of the dressed diffusion coefficient implies that the effective Langevin equation must have a multiplicative noise, leading to a supplementary source of non-Gaussian displacements [14, 16–20, 54] in addition to the trivial one induced by the trapping potential.

We now invoke the Lifson-Jackson formula [55], providing the late-time diffusion coefficient in a periodic potential, which reads

$$D^*(\kappa) = \frac{\lambda^2}{\int_0^\lambda dq_1 \frac{\exp[\beta \phi(q_1)]}{D_e(q_1)} \int_0^\lambda dq_1 \exp[-\beta \phi(q_1)]}. \quad (15)$$

From this expression, we can recover Eqs. (5) and (6). We emphasize that the heuristic argument we presented here is not rigorous. Nevertheless, it can also be applied to evaluate the effective diffusion coefficient of an underdamped Brownian particle near the overdamped limit (see details in SM [53]), for which we once again recover the results obtained via rigorous multiscale analysis [51, 52]. The same method also yields the first-order correction to the effective diffusion coefficient for a particle carrying a dipole moment which interacts with a random or periodic electric field when the dipole dynamics is assumed to be much faster than the particle diffusion [56].

Numerical simulations. In order to verify the above predictions, Eqs. (3) and (4) are numerically solved for a sinusoidal trapping potential $\phi(q_1)$, using the Euler-Maruyama scheme [57], through an optimised *Cython* [58] code (see details in SM [53]). The initial positions of the test particle and the elastic mode are sampled from Eq. (2). Typical raw trajectories are shown in Fig. 2a). We observe that the elastic mode $q_2(t)$ fluctuates around its equilibrium value ($q_2 = 0$), within the harmonic potential it is subject to, whereas the position $q_1(t)$ of the test colloid fluctuates at short times in a minimum of the sinusoidal potential $\phi(q_1)$ before randomly jumping from one minimum to the other by passing the

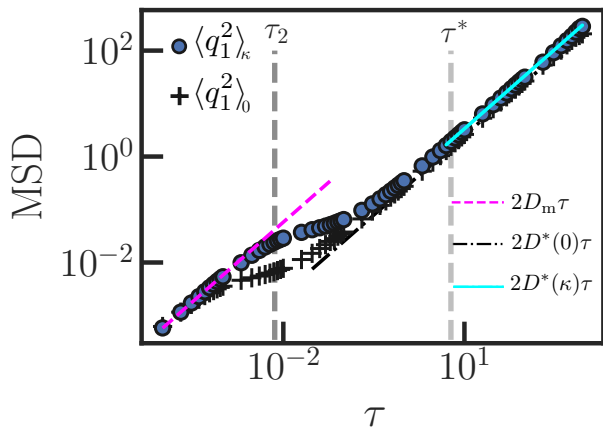


FIG. 3. Mean-squared displacements $\langle q_1^2 \rangle_\kappa$ and $\langle q_1^2 \rangle_0$ of the test particle as functions of the time increment τ , for $\kappa = 1.0 \times 10^{-3}$ (blue disks) and $\kappa = 0.0$ (black crosses) respectively, as computed from 10^2 trajectories of 10^7 points each, for $\gamma_{11} = 5.0$, $\gamma_{12} = 22.0$, $\gamma_{22} = 104.0$, $\Delta U = 1.0$, $\lambda = 1.0$, $\Delta t = 1.0 \times 10^{-4}$, and $k_B T = 1.0$. The symbols are the results from the numerical simulations. The dash-dotted black line indicates $2D^*(0)\tau$, where $D^*(0)$ is the classical Lifson-Jackson late-time diffusion coefficient [55]. The solid cyan line indicates $2D^*(\kappa)\tau$, where $D^*(\kappa)$ is the late-time diffusion coefficient given by Eq. (5). The dashed pink line indicates $2D_m\tau$, where $D_m = k_B T / \gamma_{11}$ is the bare diffusion coefficient. The characteristic times $\tau_2 = \kappa\gamma_{22}$ and $\tau^* = \lambda^2 / D^*(\kappa)$ are indicated with the dark-grey and light-grey vertical dashed lines, respectively.

potential barrier. From such trajectories, we can first compute the late-time PDF in positions. As an example, Fig. 2b) shows the equilibrium marginal PDF of the periodised variable q^* , defined by $q_1 = q^* \bmod(\lambda)$, which satisfies

$$P_{\text{meq}}(q^*) = \frac{\exp[-\beta\phi(q^*)]}{\int_0^\lambda dq \exp[-\beta\phi(q)]}. \quad (16)$$

In order to probe the dynamical effects of the hydrodynamic coupling of the trapped test particle to the elastic mode, we now turn to the analysis of the mean-squared displacement (MSD), defined as

$$\langle q_i^2 \rangle(\tau) = \langle [q_i(t+\tau) - q_i(t)]^2 \rangle, \quad (17)$$

where τ is a time increment. As shown in Fig. 3, the MSD of the test colloidal particle exhibits two distinct diffusive regimes separated by a crossover region. The short-time diffusive regime is characterized by the bare diffusion coefficient D_m . The late-time diffusive regime is characterized by the late-time diffusion coefficient D^* , and is reached when the particle has diffused over many periods of the trapping potential, i.e. when its MSD is large compared to λ^2 . This sets a time scale $\tau^* = \lambda^2 / D^*$, which must be exceeded to see the late-time diffusive regime.

Finally, we focus on the late-time diffusive regime of the simulated trajectories, and compute from the MSD

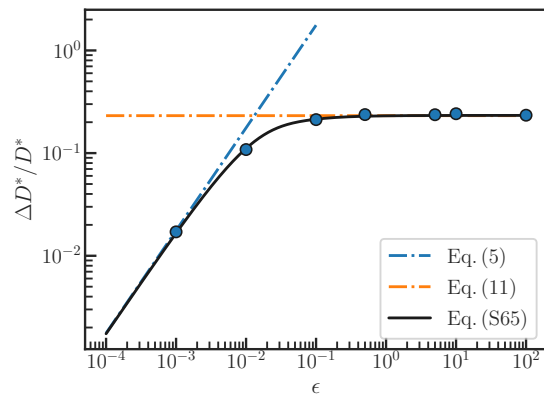


FIG. 4. Relative increase $\Delta D^*/D^* = [D^*(\epsilon) - D^*(0)]/D^*(0)$ of the late-time diffusion coefficient D^* of the test particle as a function of the dimensionless compliance ϵ , for $\mu_{11} = 1.0$, $\mu_{12} = 0.5$, $\mu_{22} = 0.5$, $\Delta U = 1.0$, $\lambda = 1.0$, and $k_B T = 1.0$. The symbols are the results from the numerical simulations. The dash-dotted blue line corresponds to the small- ϵ prediction of Eq. (5). The dash-dotted orange line corresponds to the large- ϵ prediction of Eq. (11). The solid black line corresponds to the Kubo formula of Eq. (S65) (see details in SM [53]).

the relative excess in late-time diffusion coefficient, i.e. $\Delta D^*/D^* = [D^*(\epsilon) - D^*(0)]/D^*(0)$, as a function of ϵ . The results are shown in Fig. 4 for a given set of parameters. We observe that the numerical-simulation data agree well with the small- ϵ and large- ϵ asymptotic predictions of Eqs. (5) and (11), respectively. Furthermore, the Kubo formula can be used to evaluate D^* for any value of ϵ (see details in SM [53], and in particular Eq. (S65)). We see that the numerical-simulation data agree at all ϵ values with the Kubo formula, and that the latter interpolates well the two asymptotic predictions.

In conclusion, we theoretically and numerically studied the Brownian motion of colloid-like particles near elastic interfaces using minimal ingredients. We showed that a Brownian particle trapped in a periodic potential sees its late-time diffusivity dressed and significantly enhanced by hydrodynamic coupling to a hidden elastic mode. Despite being minimalist, our description reveals generic, original and large effects, relevant for the transport of colloidal particles near complex boundaries. In particular, our results suggest the intriguing possibility of understanding, and even tuning, the diffusivity of colloids near deformable and patchy interfaces – with key implications for biophysics and nanofluidics.

Data availability. All codes used in this study to produce data are available through the online repository https://github.com/EMetBrown-Lab/Lacherez2024_Codes.

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