Dynamic Density Functional Theory
with hydrodynamic interactions and fluctuations

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We derive a closed equation for the empirical concentration of colloidal particles in the presence of both hydrodynamic and direct interactions. The ensemble average of our functional Langevin equation reproduces known deterministic Dynamic Density Functional Theory (DDFT) [*Rex and Löwen, Phys. Rev. Lett., 101(14):148302, 2008*], and, at the same time, it also describes the microscopic fluctuations around the mean behavior. We suggest separating the ideal (non-interacting) contribution from additional corrections due to pairwise interactions. We find that, for an incompressible fluid and in the absence of direct interactions, the mean concentration follows Fick's law just as for uncorrelated walkers. At the same time, the nature of the stochastic terms in fluctuating DDFT is shown to be distinctly different for hydrodynamically-correlated and uncorrelated walkers. This leads to striking differences in the behavior of the fluctuations around Fick's law, even in the absence of pairwise interactions. We connect our own prior work [*A. Donev, T. G. Fai, E. Vanden-Eijnden, J. Stat. Mech., P04004, 2014*] on fluctuating hydrodynamics of diffusion in liquids to the DDFT literature, and demonstrate that the fluid cannot easily be eliminated from consideration if one wants to describe the collective diffusion in colloidal suspensions.

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I. Introduction

Over the past decade and a half there has been considerable interest in extending traditional (static) Density Functional Theory (DFT) for liquids to account for dynamics, leading to Dynamic DFT (DDFT) [1–5]. Recently, attempts have been made to additionally account for hydrodynamic interactions (HI) among the particles due to the presence of a viscous solvent [6–10], as necessary when modeling colloidal suspensions. A key feature of these proposed HI+DDFT theories is that even for the simple case of non-interacting Brownian particles suspended in a fluid the resulting equations appear to be different from Fick's law, in contrast to the case of independent (uncorrelated) Brownian walkers. Here we show that for the case of non-interacting but hydrodynamically-correlated Brownian particles one can write down a closed equation for the average density that is exactly Fick's law, without uncontrolled approximations such as closures of the BBGKY hierarchy. Furthermore, our equation includes fluctuations around Fick's law, and sheds light on the controversy over the difference between deterministic and fluctuating DDFT [1–4]. The derivation presented here follows on our previous work [11] in which we obtain the same result by using a fluctuating hydrodynamic formalism. Here we follow an approach originally proposed by Dean [3] for the case of uncorrelated Brownian walkers, and obtain the same equation as derived in [11] by rather different means. Our work demonstrates that hydrodynamics is not something that is to be added to Fick's law as non-local correction; rather, fluctuating hydrodynamics underlies diffusion and gives rise to Fick's law. This simple yet seemingly frequently missed point is silently evidenced by the well-known Stokes-Einstein relation, which relates the diffusion coefficient $\chi \sim k_B T / (\eta \sigma)$ to the temperature T, the size of the particles σ, *and* the viscosity of the fluid η.

For consistency, in this paper we use the notation of our prior closely-related work [11] instead of the notation more common in the DDFT literature. We start from the overdamped Langevin equations of Brownian Dynamics (BD), which are often used to model dynamics of colloidal particles or polymer chains in flow. The Ito equations of motion for the (correlated) positions of the N particles $\boldsymbol{Q}(t) = \{\boldsymbol{q}_1(t), \ldots, \boldsymbol{q}_N(t)\}$ are

$$d\boldsymbol{Q} = -\boldsymbol{M}\left(\partial_{\boldsymbol{Q}} U\right) dt + \left(2 k_B T\, \boldsymbol{M}\right)^{\frac{1}{2}} d\boldsymbol{\mathcal{B}} + k_B T \left(\partial_{\boldsymbol{Q}} \cdot \boldsymbol{M}\right) dt, \tag{1}$$

where $\boldsymbol{\mathcal{B}}(t)$ is a collection of independent Brownian motions, $U(\boldsymbol{Q})$ is a conservative interaction potential, $\boldsymbol{M}(\boldsymbol{Q}) \succeq \boldsymbol{0}$ is a symmetric positive semidefinite *mobility* block matrix for the collection of particles. The Fokker-Planck equation (FPE) for the probability density $P(\boldsymbol{Q}, t)$ corresponding to (1) is

$$\frac{\partial P}{\partial t} = \frac{\partial}{\partial \boldsymbol{Q}} \cdot \left\{ \boldsymbol{M} \left[\frac{\partial U}{\partial \boldsymbol{Q}} P + (k_B T) \frac{\partial P}{\partial \boldsymbol{Q}} \right] \right\}, \tag{2}$$

and is in detailed-balance (i.e., is time reversible) with respect to the Gibbs-Boltzmann distribution $\sim \exp\left(-U(\boldsymbol{Q})/k_B T\right)$. A commonly-used model of the mobility matrix, suitable for dilute suspensions, is the Rotne-Prager pairwise approximation [12].

We will assume here that the mobility is *pairwise* additive, and that the block of the mobility corresponding to the pair of particles i and j is a smooth function of *only* the positions of those particles,

$$\forall (i,j): \quad \boldsymbol{M}_{ij}\left(\boldsymbol{q}_i, \boldsymbol{q}_j\right) = \frac{\boldsymbol{\mathcal{R}}\left(\boldsymbol{q}_i, \boldsymbol{q}_j\right)}{k_B T}, \tag{3}$$

where $\boldsymbol{\mathcal{R}}\left(\boldsymbol{r}, \boldsymbol{r}'\right)$ is a symmetric positive-semidefinite (SPD) tensor kernel (linear operator mapping vector fields to vector fields) [1]. The assumption of pairwise additivity is appropriate for low-density colloidal suspensions, when the typical distance between particles is significantly larger than the typical size of a particle; at higher densities complex many-body effects appear which are beyond the scope of this work. Because we assume that (3) holds even if $i = j$, the self-diffusion tensor of a particle with position $\boldsymbol{r}$ is

$$\boldsymbol{\chi}\left(\boldsymbol{r}\right) = \boldsymbol{\mathcal{R}}\left(\boldsymbol{r}, \boldsymbol{r}\right).$$

For confined systems, $\boldsymbol{\mathcal{R}}\left(\boldsymbol{r}, \boldsymbol{r}'\right)$ depends on the positions of the two particles relative to the boundaries and $\boldsymbol{\chi}\left(\boldsymbol{r}\right)$ may be anisotropic and may depend on the position; for a translationally-invariant and isotropic

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[1] Here we adopt the notation of our previous work [11], except that we have included an additional factor of 2 in $\boldsymbol{\mathcal{R}}$ to simplify some of the expressions.

$_{43}$ system $\boldsymbol{\mathcal{R}}\left(\boldsymbol{r}, \boldsymbol{r}'\right) \equiv \boldsymbol{\mathcal{R}}\left(\boldsymbol{r} - \boldsymbol{r}'\right)$ and $\boldsymbol{\chi}\left(\boldsymbol{r}\right) = \chi\boldsymbol{I}$, where χ is the self-diffusion coefficient of the particles. Rex
$_{44}$ and Löwen [6, 7] assume translational invariance but take a form for the mobility in which the diagonal
$_{45}$ elements of the mobility are treated differently from the off-diagonal ones,

$$\boldsymbol{M}_{ij}\left(\boldsymbol{Q}\right) \equiv \boldsymbol{M}_{ij}\left(\boldsymbol{q}_i, \boldsymbol{q}_j\right) = \frac{\chi}{k_B T}\left[\delta_{ij}\boldsymbol{I} + \left(1 - \delta_{ij}\right)\boldsymbol{\omega}_{12}\left(\boldsymbol{q}_i - \boldsymbol{q}_j\right)\right],$$

$$= \frac{\chi}{k_B T}\left[\delta_{ij}\left(\boldsymbol{I} - \boldsymbol{\omega}_{12}\left(0\right)\right) + \boldsymbol{\omega}_{12}\left(\boldsymbol{q}_i - \boldsymbol{q}_j\right)\right] \tag{4}$$

$_{46}$ where we have neglected higher-order hydrodynamic corrections and taken $\omega_{11} = 0$, which is appropriate
$_{47}$ for dilute suspensions. The Rotne-Prager [12] form for $\boldsymbol{\omega}_{12}\left(\boldsymbol{r}\right)$, which is what Rex and Löwen used in
$_{48}$ their numerical simuations, can be written in the form

$$\boldsymbol{\omega}_{12}\left(\boldsymbol{r}\right) = \begin{cases} \left(\dfrac{3\sigma}{4r} + \dfrac{\sigma^3}{2r^3}\right)\boldsymbol{I} + \left(\dfrac{3\sigma}{4r} - \dfrac{3\sigma^3}{2r^3}\right)\dfrac{\boldsymbol{r} \otimes \boldsymbol{r}}{r^2}, & r > 2\sigma \\[2ex] \left(1 - \dfrac{9r}{32\sigma}\right)\boldsymbol{I} + \left(\dfrac{3r}{32\sigma}\right)\dfrac{\boldsymbol{r} \otimes \boldsymbol{r}}{r^2}, & r \le 2\sigma \end{cases} \tag{5}$$

$_{49}$ where σ is the radius of the colloidal particles, and satisfies the key condition $\boldsymbol{\omega}_{12}(0) = \boldsymbol{I}$. Therefore, the
$_{50}$ term involving δ_{ij} in (4) can be deleted and (4) becomes of the form (3) with

$$\boldsymbol{\mathcal{R}}\left(\boldsymbol{q}_i, \boldsymbol{q}_j\right) \equiv \chi\boldsymbol{\omega}_{12}\left(\boldsymbol{q}_i - \boldsymbol{q}_j\right).$$

$_{51}$ Note that in colloidal suspensions there is typically a hard-core repulsion that ensures that particles
$_{52}$ essentially never overlap, which implies that the behavior of $\boldsymbol{\omega}_{12}\left(\boldsymbol{r}\right)$ for $r \le 2\sigma$ is not expected to be
$_{53}$ important. Since the effect of hydrodynamic interactions is distinct from that of direct interactions, it
$_{54}$ is important to also consider the case of an ideal gas in which the only interparticle interactions are of
$_{55}$ hydrodynamic origin. Furthermore, particles can overlap relative to their far-field hydrodynamic radius
$_{56}$ for suspensions of soft particles such as star polymer chains [13].
$_{57}$ In a clever but formal derivation [4], Dean started from the overdamped Langevin equations for a
$_{58}$ collection of N interacting Brownian walkers driven by *independent* noise, i.e., a diagonal mobility matrix
$_{59}$ $\boldsymbol{M} = \left(k_B T\right)^{-1}\chi\boldsymbol{I}$, to obtain a closed-form equation for the *empirical* or *fluctuating* density (concentration)
$_{60}$ of particles

$$c\left(\boldsymbol{r}, t\right) = \sum_{i=1}^{N}\delta\left(\boldsymbol{q}_i\left(t\right) - \boldsymbol{r}\right). \tag{6}$$

$_{61}$ For non-interacting particles, this equation can formally be written as an Ito stochastic partial differential
$_{62}$ equation (SPDE) [4],

$$\partial_t c = \chi\boldsymbol{\nabla}^2 c + \boldsymbol{\nabla} \cdot \left(\sqrt{2\chi c}\,\boldsymbol{\mathcal{W}}_c\right), \tag{7}$$

$_{63}$ where $\boldsymbol{\mathcal{W}}_c\left(\boldsymbol{r}, t\right)$ denotes a spatio-temporal white-noise vector field. As pointed out in Ref. [4] and further
$_{64}$ elaborated in [2], equation (7) is simply a formal rewriting of (1). The only difference is that the identity of
$_{65}$ the different particles has been removed by going from a Lagrangian to an Eulerian description. Importantly,
$_{66}$ the solution of (7) should forever remain a sum of delta functions (whose positions diffuse independently).
$_{67}$ In fact, the multiplicative noise SPDE (7) as written has no clear mathematical interpretation, and neither
$_{68}$ does the square root of a sum of delta functions in the noise amplitude.
$_{69}$ Of primary interest in practice are expectation values of the instantaneous concentration $c\left(\boldsymbol{r}, t\right)$, such
$_{70}$ as the *average* concentration, which is also the single-particle distribution function $c^{(1)}\left(\boldsymbol{r}, t\right) = \langle c\left(\boldsymbol{r}, t\right)\rangle$.
$_{71}$ Taking an ensemble average of (7) is trivial because of the linearity of the deterministic term and the
$_{72}$ fact that the noise term averages to zero due to its Ito interpretation, and for the case of non-interacting
$_{73}$ particles one simply obtains Fick's law,

$$\partial_t c^{(1)} = \chi\boldsymbol{\nabla}^2 c^{(1)}. \tag{8}$$

$_{74}$ If direct interactions among the particles are included, one cannot write a closed form equation and an infinite
$_{75}$ hierarchy of BBGKY equations arises; a closure approximation for the higher-order correlation functions is

⁷⁶ required, as explained by Marconi and Tarazona [4]. It is important to note that (7) and (8) describe rather dif-
⁷⁷ ferent objects; the solution to (7) is a spiky sum of delta functions, and not a smooth average density or single-
⁷⁸ particle distribution function as Fick's *deterministic* law (8) or traditional (static) DFT describes [1, 2, 4].
⁷⁹ As summarized in Ref. [2], Fick's law (8) can also be obtained by starting from the FPE (2) and
⁸⁰ integrating over $N-1$ particles to get an equation for the single-particle distribution function $c^{(1)}(\boldsymbol{r}, t)$.
⁸¹ This route was followed by Rex and Löwen [6, 7] in order to include the effect of hydrodynamic interactions
⁸² in (8) and obtain an equation that, at first sight, appears distinctly different from Fick's law. It is important
⁸³ to note that in order to close the BBGKY hierarchy some uncontrolled approximations are made in Refs.
⁸⁴ [6, 7]; we will not require such approximations until Section II. For non-interacting particles, in our notation,
⁸⁵ eq. (5) in Ref. [7] reduces to

$$\partial_t c^{(1)}(\boldsymbol{r}, t) = \chi \boldsymbol{\nabla}^2 c^{(1)}(\boldsymbol{r}, t) + \chi \boldsymbol{\nabla} \cdot \left(\int \boldsymbol{\omega}_{12}(\boldsymbol{r} - \boldsymbol{r}') \, \boldsymbol{\nabla}' c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t) \, d\boldsymbol{r}' \right), \tag{9}$$

⁸⁶ where $c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t)$ is the two-particle distribution function, and we use $\boldsymbol{\nabla}$ to denote gradient with respect to $\boldsymbol{r}$
⁸⁷ and $\boldsymbol{\nabla}'$ with respect to $\boldsymbol{r}'$. In this work, we derive an equation for the empirical (fluctuating) concentration in
⁸⁸ the presence of hydrodynamic interactions similar to (7), whose expectation gives (9). In the absence of direct
⁸⁹ interactions this equation is given by (12) and was previously derived by us using a different approach in
⁹⁰ Ref. [11]. In addition to reproducing Fick's law for the average, (12) also describes the long-range correlated
⁹¹ fluctuations around the mean. Here we also include the effect of direct interactions among the particles.
⁹² The first term on the right hand side of (9) is the familiar *local* Fick's law; but the second term is a
⁹³ *non-local* diffusion term. It is important to note that the far-field behavior of the mobility (5) is given
⁹⁴ by the scaled Oseen tensor

$$\boldsymbol{\omega}_{12}(\boldsymbol{r}) = \frac{3}{4} \frac{\sigma}{r} \left(\boldsymbol{I} + \frac{\boldsymbol{r} \otimes \boldsymbol{r}}{r^2} \right) + O\left(\left(\frac{\sigma}{r} \right)^3 \right), \tag{10}$$

⁹⁵ which is long-ranged and decays as r^{-1}. While it may at first sight look like $\boldsymbol{\omega}_{12}(\boldsymbol{r})$ is small for $r \gg \sigma$,
⁹⁶ it should be recalled that the Stokes-Einstein formula $\chi = k_B T / (6\pi\eta\sigma)$ implies that the second term in (9)
⁹⁷ is independent ² of σ since $\chi \boldsymbol{\omega}_{12}(\boldsymbol{r}) \sim (k_B T) / (\eta r)$. The equation of Rex and Löwen (9) therefore implies
⁹⁸ that Fick's law needs to be amended with a long-ranged non-local term even for *dilute* suspensions with
⁹⁹ *no* direct interactions among the diffusing particles.
¹⁰⁰ Let us observe, however, that the Rotne-Prager mobility (5) satisfies an additional key property,
¹⁰¹ $\boldsymbol{\nabla} \cdot \boldsymbol{\omega}_{12}(\boldsymbol{r}) = 0$, or more generally,

$$\boldsymbol{\nabla} \cdot \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') = 0. \tag{11}$$

¹⁰² This is a direct consequence of the fact that hydrodynamic interactions (correlations) are mediated by an
¹⁰³ incompressible fluid [12]. In this case the second term on the right hand side of (9) in fact *vanishes* after a
¹⁰⁴ simple integration by parts. Therefore, Fick's law (8) for the average concentration remains valid even in the
¹⁰⁵ presence of long-ranged hydrodynamic correlations among the Brownian walkers. This important physical
¹⁰⁶ implication of (11) seems to be have been missed in [6, 7] and subsequent works because the focus in DFT,
¹⁰⁷ and therefore DDFT, is almost exclusively on interacting particles and nonlocal free-energy functionals, and
¹⁰⁸ comparatively little attention seems to have been given to the nonlocal difffusion aspect of (9). Following
¹⁰⁹ the completion of this work, we learned of an early derivation by Altenberger and Deutch that showed
¹¹⁰ that, indeed, (8) holds even in the presence of hydrodynamic interactions (correlations), see (3.10) in Ref.
¹¹¹ [14]. These authors also made use of and noted the importance of the divergence-free condition (11).
¹¹² It is important to also note another derivation aiming to include hydrodynamics in DDFT, developed by the
¹¹³ authors of Refs. [8–10]. These authors argue that inertia also needs to be included, and arrive at an equation
¹¹⁴ that has even more non-local terms than (9). We believe that these derivations, while careful (even rigorous),
¹¹⁵ start from an incorrect inertial formulation of the equations of motion of colloidal particles immersed in fluid.
¹¹⁶ As explained by Hinch [15] and later summarized eloquently and clearly by Roux [16], any equation of motion
¹¹⁷ that accounts for inertial effects *must* include the inertia of the fluid in addition to any excess inertia of the
¹¹⁸ particles over the fluid they expel. This is because the time it takes for momentum to diffuse through the liquid,

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² This is expected since the leading-order hydrodynamic correction comes from a monopole term (Stokeslet) that corresponds to a *point* force in a Stokesian fluid.

₁₁₉ with diffusion coefficient equal to the kinematic viscosity $\nu = \eta/\rho$ (note the appearance of the fluid inertia here
₁₂₀ via the density ρ), is in fact *longer* than inertial time scales. It is therefore inconsistent to use hydrodynamic
₁₂₁ friction or mobility functions such as the Rotne-Prager tensor, which assume steady Stokes flow, i.e., infinitely
₁₂₂ fast momentum diffusion, while including inertia of the particles explicitly. The only Markovian formulation
₁₂₃ of the hydrodynamics of colloidal suspensions that includes *both* hydrodynamics and thermal fluctuations
₁₂₄ (Brownian motion) consistently is that of *fluctuating hydrodynamics* [15, 17, 18]. Roux starts from the inertial
₁₂₅ formulation of Hinch [15] and derives the overdamped equation of motion (1) from those inertial equations
₁₂₆ [16]. We therefore consider the overdamped equation (1), rather than the inertial Langevin equations used
₁₂₇ by a number of authors [8, 19, 20], as the correct starting point for including hydrodynamics in DDFT.

In our own recent work [11], we started from a simplified version of the complete formulation of Hinch [15]
and Roux [16]. In this approximation [21–25], the no-slip condition resolved over the surface of the particles
is approximated by an average no-slip condition at the centroid of each of the particles, and the particles are
assumed to be neutrally-buoyant (but see Ref. [24] for an extension to account for excess inertia). Another
way to think of the approximation is as a low-order multipole approximation of the complete hydrodynamics,
suitable for dilute suspensions, and accurate to the *same* order as the Rotne-Prager far-field approximation
[25, 26]. By starting from the simplified fluctuating hydrodynamic formulation and eliminating the fluid
velocity as a fast variable, one can obtain the overdamped Lagrangian equation (1) [11, 21]. In Ref. [11] we
started from an inertial *Eulerian* description, i.e., a description involving not the positions of the individual
particles but rather the empirical concentration $c\,(\boldsymbol{r}, t)$, and obtained, by adiabatic elimination of the fast
fluid velocity, the overdamped Eulerian Ito SPDE

$$\partial_t c = \boldsymbol{\nabla} \cdot [\boldsymbol{\chi}\,(\boldsymbol{r})\,\boldsymbol{\nabla} c] - \boldsymbol{w} \cdot \boldsymbol{\nabla} c. \tag{12}$$

Here $\boldsymbol{w}\,(\boldsymbol{r}, t)$ is a random velocity field that is white in time and has a spatial covariance [11],

$$\langle \boldsymbol{w}\,(\boldsymbol{r}, t) \otimes \boldsymbol{w}\,(\boldsymbol{r}', t') \rangle = 2\boldsymbol{\mathcal{R}}\,(\boldsymbol{r}, \boldsymbol{r}')\,\delta\,(t - t')\,, \tag{13}$$

₁₂₈ and the incompressibility condition (11) is assumed to hold. The ensemble average of (12) is nothing other
₁₂₉ than Fick's law (8), and does not include any non-local diffusion terms because of the incompressibility
₁₃₀ of the fluid. It is important to point out that (12), just like (7), describes a spiky sum of delta functions
₁₃₁ which are advected by a rapidly-decorrelating random velocity field. However, (12) is distinctly different
₁₃₂ from (7): while both equations have multiplicative noise, (12) is *linear*, while (7) is *nonlinear*. As we discuss
₁₃₃ in more detail in the Conclusions, one can obtain (7) from (12) upon taking a suitable (nontrivial) limit
₁₃₄ in which $\boldsymbol{\mathcal{R}}\,(\boldsymbol{r}, \boldsymbol{r}')$ becomes highly localized around $\boldsymbol{r} = \boldsymbol{r}'$.
₁₃₅ Here, we connect our prior work to the DDFT literature, by obtaining the overdamped Eulerian (fluctuating
₁₃₆ DDFT) equation (12) starting from the overdamped Lagrangian equation (1), rather than from the inertial
₁₃₇ Eulerian formulation as we did in Ref. [11]. Our argument is essentially a generalization of that of Dean [3]
₁₃₈ and makes specific use of the hydrodynamic formulation that is hidden in Rotne-Prager-like approximations
₁₃₉ to the mobility matrix. As it must, for non-interacting particles the present calculation gives exactly the same
₁₄₀ result (12) for the empirical concentration and Fick's law (8) for the average concentration. Furthermore,
₁₄₁ here we extend our previous work to account for direct interactions (as opposed to hydrodynamic interactions)
₁₄₂ among the diffusing particles. Just as in the work of Dean [4], we obtain a *closed* but *nonlinear* and
₁₄₃ *nonlocal* equation for the empirical (fluctuating) concentration. As expected, in the presence of interactions
₁₄₄ it is not possible to write down a closed form for the ensemble-averaged concentration, and approximate
₁₄₅ closures are required for two-particle and three-particle correlation functions [4, 6, 7].
₁₄₆ This paper is organized as follows. In Section II we summarize and then derive our key result (15),
₁₄₇ a fluctuating diffusion equation for a collection of particles interacting both hydrodynamically and via
₁₄₈ conservative potentials. In Section III we discuss coarse-graining (averaging) and the relation of our work
₁₄₉ to density functional theory, Fick's macroscopic law, and fluctuating hydrodynamics, and point to several
₁₅₀ important open problems. Finally, we give some conclusions in Section IV.

₁₅₁ II.   Fluctuating DDFT with Hydrodynamic Interactions

₁₅₂ In this section we summarize our main results, and defer the detailed derivations to Appendix A. For
₁₅₃ completeness, we will include here a direct interaction among the particles in the form of a conservative

154 potential that includes an external potential $U_1(\boldsymbol{r})$ and a pairwise additive potential $U_2(\boldsymbol{r}, \boldsymbol{r}')$,

$$U(\boldsymbol{Q}) = \sum_{i=1}^{N} U_1(\boldsymbol{q}_i) + \frac{1}{2} \sum_{\substack{i,j=1 \\ i \neq j}}^{N} U_2(\boldsymbol{q}_i, \boldsymbol{q}_j) \tag{14}$$

155 where, without loss of generality, we can assume that $U_2(\boldsymbol{r}, \boldsymbol{r}') = U_2(\boldsymbol{r}', \boldsymbol{r})$ and $[\boldsymbol{\nabla} U_2(\boldsymbol{r}, \boldsymbol{r}')]_{\boldsymbol{r}'=\boldsymbol{r}} = 0$. Note
156 that such an interaction was not included in our prior work [11].
157 Here we use (1,3) to formally derive a closed-form SPDE for the empirical concentration (6). Our
158 calculation mimics the one performed by Dean for the case of uncorrelated walkers [3]. The result of the
159 calculations detailed in Appendix A is the fluctuating hydrodynamic equation (conservation law)

$$\begin{aligned}
\partial_t c(\boldsymbol{r}, t) = &-\boldsymbol{\nabla} \cdot (\boldsymbol{w}(\boldsymbol{r}, t) c(\boldsymbol{r}, t)) + \boldsymbol{\nabla} \cdot (\boldsymbol{\chi}(\boldsymbol{r}) \boldsymbol{\nabla} c(\boldsymbol{r}, t) + \boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r}) c(\boldsymbol{r}, t)) \\
&+ \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' c(\boldsymbol{r}', t) \, d\boldsymbol{r}' \right) \\
&+ (k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' U_1(\boldsymbol{r}') c(\boldsymbol{r}', t) \, d\boldsymbol{r}' \right) \\
&+ (k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' U_2(\boldsymbol{r}', \boldsymbol{r}'') c(\boldsymbol{r}', t) c(\boldsymbol{r}'', t) \, d\boldsymbol{r}' d\boldsymbol{r}'' \right),
\end{aligned} \tag{15}$$

160 where $\boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r}') = \boldsymbol{\nabla}' \cdot \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}')$ and $\boldsymbol{w}(\boldsymbol{r}, t)$ is a random velocity field with covariance (13), see (A14) for
161 a derivation of the stochastic term in the Ito convention and (A5) for the Stratonovich interpretation.
162 Compare (15) to the equation obtained by following the same procedure for the case of uncorrelated
163 particles, $\boldsymbol{M}_{ij} = \delta_{ij} (k_B T)^{-1} \boldsymbol{\chi}(\boldsymbol{q}_i)$,

$$\begin{aligned}
\partial_t c(\boldsymbol{r}, t) = &\boldsymbol{\nabla} \cdot \left((2\boldsymbol{\chi}(\boldsymbol{r}) c(\boldsymbol{r}, t))^{\frac{1}{2}} \boldsymbol{\mathcal{W}}_c \right) + \boldsymbol{\nabla} \cdot (\boldsymbol{\chi}(\boldsymbol{r}) \boldsymbol{\nabla} c(\boldsymbol{r}, t)) \\
&+ (k_B T)^{-1} \boldsymbol{\nabla} \cdot (\boldsymbol{\chi}(\boldsymbol{r}) c(\boldsymbol{r}, t) \boldsymbol{\nabla} U_1(\boldsymbol{r})) \\
&+ (k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(\boldsymbol{\chi}(\boldsymbol{r}) c(\boldsymbol{r}, t) \int \boldsymbol{\nabla}' U_2(\boldsymbol{r}, \boldsymbol{r}') c(\boldsymbol{r}', t) \, d\boldsymbol{r}' \right)
\end{aligned} \tag{16}$$

164 which is a slight generalization of Eq. (17) in [3] to account for the one-particle potential and the possible
165 anisotropy and spatial dependence of the diffusion tensor $\boldsymbol{\chi}(\boldsymbol{r})$.
166 Ensemble averaging (15) gives the first member of a BBGKY-like hierarchy of equations for the
167 single-particle distribution function,

$$\begin{aligned}
\partial_t c^{(1)}(\boldsymbol{r}, t) = &\boldsymbol{\nabla} \cdot \left(\boldsymbol{\chi}(\boldsymbol{r}) \boldsymbol{\nabla} c^{(1)}(\boldsymbol{r}, t) \right) + \boldsymbol{\nabla} \cdot \left(\int \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t) \, d\boldsymbol{r}' \right) \\
&+ (k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(\boldsymbol{\chi}(\boldsymbol{r}) \boldsymbol{\nabla} U_1(\boldsymbol{r}) c^{(1)}(\boldsymbol{r}, t) + \int \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' U_1(\boldsymbol{r}') c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t) \, d\boldsymbol{r}' \right) \\
&+ (k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(\int (\boldsymbol{\chi}(\boldsymbol{r}) \boldsymbol{\nabla} U_2(\boldsymbol{r}, \boldsymbol{r}') + \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' U_2(\boldsymbol{r}, \boldsymbol{r}')) c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t) \, d\boldsymbol{r}' \right) \\
&+ (k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(\int \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') c^{(3)}(\boldsymbol{r}, \boldsymbol{r}', \boldsymbol{r}'', t) \boldsymbol{\nabla}' U_2(\boldsymbol{r}', \boldsymbol{r}'') \, d\boldsymbol{r}'' d\boldsymbol{r}' \right),
\end{aligned} \tag{17}$$

168 which is a slight generalization of equation (5) in Ref. [6, 7] with $\omega_{11} = 0$. Here $c^{(3)}(\boldsymbol{r}, \boldsymbol{r}', \boldsymbol{r}'', t)$ denotes
169 the three-particle correlation function. We note that the term involving $c^{(3)}$ is missing in (4.4) in Ref. [14],
170 as well as (3.1) in Ref. [27], apparently because of an additional low-density approximation in the spirit
171 of kinetic theory.
172 When the incompressibility condition (11) is satisfied, Eqs. (15) and (17) simplify in a key way; as also
173 observed in Ref. [14], after an integration by parts the nonlocal diffusion term on the second line of (15) and

174 the second term on the right hand side in the first line of (17) disappear, see (A17) in the Ito convention
175 and (A6) for the Stratonovich interpretation [3]. Therefore, in the absence of interactions the fluctuating
176 DDFT equation (15) reduces to (12) and the mean follows the local Fickian diffusion equation (8), *even*
177 in the presence of hydrodynamic correlations among the particles. This important physical consequence of
178 incompressibility was not observed by Rex and Löwen [6, 7], and this omission may have lead some readers to
179 the wrong conclusion that hydrodynamic interactions lead to nonlocal corrections to Fick's law for the mean.
180 Although not apparent at first sight, (15) has the same structure of an overdamped Langevin equation
181 as does (1), namely, we can formally write it in the compact notation [28]

$$\partial_t c = -\boldsymbol{\mathcal{M}}\left[c(\cdot, t)\right] \frac{\delta H}{\delta c\left(\cdot, t\right)} + \left(2 k_B T \boldsymbol{\mathcal{M}}\left[c(\cdot, t)\right]\right)^{\frac{1}{2}} \boldsymbol{\mathcal{W}}_c(\cdot, t) + k_B T \left(\frac{\delta}{\delta c\left(\cdot, t\right)} \cdot \boldsymbol{\mathcal{M}}\left[c(\cdot, t)\right]\right), \qquad (18)$$

182 where the mobility $\boldsymbol{\mathcal{M}}\left[c(\cdot)\right]$ is a positive-semidefinite linear operator that is a functional of the function
183 of position c, denoted here by the notation $[c(\cdot)]$, and products imply a contraction over spatial position.
184 More precisely,

$$\partial_t c(\boldsymbol{r}, t) = -\int d\boldsymbol{r}' \mathcal{M}\left[c(\cdot, t); \boldsymbol{r}, \boldsymbol{r}'\right] \frac{\delta H}{\delta c\left(\boldsymbol{r}', t\right)} \qquad (19)$$

$$+ \left(2 k_B T\right)^{\frac{1}{2}} \int d\boldsymbol{r}' \boldsymbol{\mathcal{M}}^{\frac{1}{2}}\left[c(\cdot, t); \boldsymbol{r}, \boldsymbol{r}'\right] \boldsymbol{\mathcal{W}}_c(\boldsymbol{r}', t)$$

$$+ \left(k_B T\right) \int d\boldsymbol{r}' \left(\frac{\delta \mathcal{M}\left[c(\cdot, t); \boldsymbol{r}, \boldsymbol{r}'\right]}{\delta c\left(\boldsymbol{r}', t\right)}\right),$$

185 where the mobility $\mathcal{M}\left[c(\cdot)\right]\left(\boldsymbol{r}, \boldsymbol{r}'\right) \equiv \mathcal{M}\left[c(\cdot); \boldsymbol{r}, \boldsymbol{r}'\right]$ is defined by its action on a scalar field $f(\boldsymbol{r})$,

$$\int d\boldsymbol{r}' \mathcal{M}\left[c(\cdot); \boldsymbol{r}, \boldsymbol{r}'\right] f(\boldsymbol{r}') \equiv -\left(k_B T\right)^{-1} \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}) \int \boldsymbol{\mathcal{R}}\left(\boldsymbol{r}, \boldsymbol{r}'\right) c(\boldsymbol{r}') \boldsymbol{\nabla}' f(\boldsymbol{r}') d\boldsymbol{r}'\right).$$

186 Here $H\left[c(\boldsymbol{r})\right]$ is an energy functional consisting of an ideal and an excess (potential) contribution,

$$H\left[c\left(\cdot\right)\right] = H_{\mathrm{id}}\left[c\left(\cdot\right)\right] + H_{\mathrm{exc}}\left[c\left(\cdot\right)\right] = H_{\mathrm{id}}\left[c\left(\cdot\right)\right] + \int U_1(\boldsymbol{r}) c(\boldsymbol{r}) d\boldsymbol{r} + \frac{1}{2} \int U_2(\boldsymbol{r}, \boldsymbol{r}') c(\boldsymbol{r}) c(\boldsymbol{r}') \, d\boldsymbol{r} d\boldsymbol{r}',$$

187 where the ideal gas energy functional is

$$H_{\mathrm{id}}\left[c\left(\cdot\right)\right] = k_B T \int c\left(\boldsymbol{r}\right) \left(\ln\left(\Lambda^3 c\left(\boldsymbol{r}\right)\right) - 1\right) \, d\boldsymbol{r},$$

188 Λ is a constant (e.g., the thermal de Broglie wavelength), and H_{exc} is the excess free energy functional. It
189 is important to note that when incompressibility condition (11) holds, we can remove the ideal contribution
190 from H and define $H \equiv H_{\mathrm{exc}}$ without affecting (19), because

$$\int \boldsymbol{\mathcal{R}}\left(\boldsymbol{r}, \boldsymbol{r}'\right) c(\boldsymbol{r}') \boldsymbol{\nabla}' \left(\frac{\delta H_{\mathrm{id}}}{\delta c\left(\boldsymbol{r}'\right)}\right) \, d\boldsymbol{r}' = \int \boldsymbol{\mathcal{R}}\left(\boldsymbol{r}, \boldsymbol{r}'\right) \boldsymbol{\nabla}' c(\boldsymbol{r}') \, d\boldsymbol{r}' = 0.$$

191 Also note that in the case of independent (uncorrelated) particles, (16) can be written as a functional
192 Langevin equation (19) with the same free-energy functional but with a different mobility operator $\boldsymbol{\mathcal{M}}_{\mathrm{ind}}$,
193 defined by its action on a scalar field $f(\boldsymbol{r})$,

$$\int d\boldsymbol{r}' \mathcal{M}_{\mathrm{ind}}\left[c(\cdot); \boldsymbol{r}, \boldsymbol{r}'\right] f(\boldsymbol{r}') \equiv -\left(k_B T\right)^{-1} \boldsymbol{\nabla} \cdot \left(\boldsymbol{\chi}(\boldsymbol{r}) c(\boldsymbol{r}) \boldsymbol{\nabla} f(\boldsymbol{r})\right).$$

---

[3] Note that for incompressible $\boldsymbol{w}$ we have $\boldsymbol{\nabla} \cdot (\boldsymbol{w} c) = \boldsymbol{w} \cdot \boldsymbol{\nabla} c$.

The kinetic form [29] of the (formal) functional FPE associated with (19) implies that the equilibrium distribution associated with (15), assumed to be unique, is the formal Gibbs-Boltzmann distribution

$$P\left[c(\cdot)\right] = Z^{-1} \exp\left(-\frac{H\left[c\left(\cdot\right)\right]}{k_B T}\right), \tag{20}$$

which is the field representation of the equilibrium distribution $\exp\left(-U(\boldsymbol{Q})/k_B T\right)$ associated with the particle description (1). In the incompressible case, uniqueness of the Gibbs-Boltzmann distribution can be ensured by adding a small multiple of the identity (so-called bare diffusion [11]) to the mobility matrix $\boldsymbol{M}$, that is, by adding a small multiple of $\boldsymbol{\mathcal{M}}_{\text{ind}}$ to the mobility operator $\boldsymbol{\mathcal{M}}$.

III. Coarse-Graining

As noted by Marconi and Tarazona [4], (15) contains the same physical content as (1) because we have not performed any *coarse graining* or averaging, and have not lost any information except the particle numbering. Nevertheless, (15) is an informative nontrivial rewriting of (1) that can be used to perform additional *coarse-graining* and attempt to describe the behavior of collective diffusion in colloidal suspensions at a spectrum of length (and thus also time) scales, going from a microscopic scale ξ to macroscopic scales. Here we discuss three distinct types of coarse-graining one can perform on (15): an ensemble average over the realizations of the noise, an average over an ensemble of initial conditions, and spatial averaging over a large number of particles [30]. Spatial averaging is of great interest in practice since colloidal suspensions are typically observed at mesoscopic scales larger than the size of individual particles. For example, in typical experiments such as light scattering from colloidal suspensions, concentration fluctuations are averaged over a region containing many particles (e.g., the thickness of the sample).

One of the simplest, though by no means the only [31], ways to approach such spatial coarse graining is to define a smoothed empirical concentration that averages over particles in a physical region of typical size ξ (see Section 4 in Ref. [2] and Section IV in Ref. [11]),

$$c_\xi\left(\boldsymbol{r}, t\right) = \sum_{i=1}^{N} \delta_\xi\left(\boldsymbol{q}_i\left(t\right) - \boldsymbol{r}\right), \tag{21}$$

where δ_ξ is a smoothing kernel with support $\sim \xi$ that converges to a delta function as $\xi \to 0$ (e.g., a Gaussian with standard deviation ξ). For ξ smaller than the typical particle size or interparticle distance, we have little to no coarse-graining and detailed microstructural information (e.g., layering in a hard-core fluid) is encoded in c_ξ. For ξ much larger than some characteristic correlation length (e.g., decay length of the pair correlation function), microstructural information will no longer be encoded in c_ξ, although fluctuations in c_ξ may still be non-negligible. Ultimately, for very large ξ we expect c_ξ to become *macroscopic* with negligible fluctuations, although it is not *a priori* obvious how large ξ needs to be for this to become the case.

A. Ensemble Averaging

For simplicity, and in order to facilitate a direct comparison with prior work by others, in this section we will assume there is no external potential, $U_1 = 0$. We will also assume an isotropic homogeneous (translationally- and rotationally-invariant) system,

$$\boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') \equiv \boldsymbol{\mathcal{R}}(\boldsymbol{r} - \boldsymbol{r}') \quad \text{and} \quad \boldsymbol{\chi}(\boldsymbol{r}) \equiv \chi\boldsymbol{I}.$$

Furthermore, we will assume that the incompressibility condition (11) holds, which we again emphasize is true for the Rotne-Prager mobility.

Direct ensemble averaging of the functional Langevin equation (15) gives

$$\partial_t c^{(1)}(\boldsymbol{r}, t) = -\int d\boldsymbol{r}' \left\langle \mathcal{M}\left[c(\cdot, t); \boldsymbol{r}, \boldsymbol{r}'\right] \frac{\delta H_{\text{exc}}}{\delta c\left(\boldsymbol{r}', t\right)} \right\rangle \tag{22}$$
$$+ (k_B T) \int d\boldsymbol{r}' \left\langle \frac{\delta \mathcal{M}\left[c(\cdot, t); \boldsymbol{r}, \boldsymbol{r}'\right]}{\delta c\left(\boldsymbol{r}', t\right)} \right\rangle,$$

where we used the fact that for incompressible $\boldsymbol{\mathcal{R}}$ we can replace H by H_{exc}, and the fact that in the Ito interpretation the stochastic term vanishes in expectation. As derived more carefully in Appendix

A of our prior work [11], the thermal or stochastic drift term on the second line of (22) can be averaged explicitly due to linearity, and leads to the first term on the right hand side of (17). This demonstrates that Fickian diffusion is already included in the hydrodynamic correlation tensor $\boldsymbol{\mathcal{R}}$, as evidenced by the Stokes-Einstein-like relation $\chi \boldsymbol{I} = \boldsymbol{\mathcal{R}}(0)$. It also shows that all of the terms in the second, third and fourth lines of (17) come from the closure of the term $\langle -\boldsymbol{\mathcal{M}} \, \delta H_{\mathrm{exc}}/\delta c \rangle$. Recall that the second term on the first line of (17) disappears for incompressible $\boldsymbol{\mathcal{R}}$.

In order to make (17) useful in practice, some closure approximation for the two-particle correlation function is required, and it is here that equilibrium statistical mechanical quantities such as free energy functionals enter in the calculations, as first discussed by Marconi and Tarazona [4] in the absence of hydrodynamic correlations and then generalized by Rex and Löwen [6, 7] to account for hydrodynamics. Namely, by assuming that the higher-order correlation functions can be approximated by those of the equilibrium system kept at the same density profile by an external potential, system (17) can be *approximated* with (c.f. (14) in [7])

$$\partial_t c^{(1)}(\boldsymbol{r}, t) = (k_B T)^{-1} \chi \boldsymbol{\nabla} \cdot \left(c^{(1)}(\boldsymbol{r}, t) \, \boldsymbol{\nabla} \frac{\delta F}{\delta c^{(1)}(\boldsymbol{r}, t)} \right) \tag{23}$$
$$+ (k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(\int \boldsymbol{\mathcal{R}}(\boldsymbol{r} - \boldsymbol{r}') \, c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t) \, \boldsymbol{\nabla}' \frac{\delta F}{\delta c^{(1)}(\boldsymbol{r}', t)} \, d\boldsymbol{r}' \right),$$

where $F\left[c^{(1)}(\cdot) \right]$ is the *equilibrium* density functional familiar from static DFT, which is only explictly known for the ideal-gas, see the discussion around (24). This microscopic equilibrium density functional captures microstructural information about the colloidal system at scales comparable to the size of the colloidal particles. Español and Löwen [5] explain how to connect the equilibrium free-energy functional with a non-Markovian non-local equation for $c^{(1)}$ *without* making approximations; after making a Markovian (separation of time scales) approximation they obtain a non-local diffusion equation (c.f. (32) in Ref. [5]), and after a further approximation of the diffusion kernel they obtain the equation of Marconi and Tarazona. Note that in the presence of hydrodynamic correlations the second line of (23) involves $c^{(2)}$, which makes the equation unclosed and therefore not yet useful in practice without a further closure approximation for $c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t)$. Rex and Löwen [6, 7] suggest such an approximation in terms of the equilibrium pair correlation function.

It is important to note that, in general, the free-energy functional F (defined on a space of functions) that enters in the equation for the ensemble average is *different* from the energy functional H (formally defined on a space of distributions) that enters in the functional Langevin equation (19). In fact, a precise thermodynamic definition can be given to the classical DDFT functional $F\left[c^{(1)}(\cdot) \right]$ as an expectation value over a Gibbs-Boltzmann distribution related to (20). However, for noninteracting particles (an ideal gas) F and H have formally the same functional form,

$$F = F_{\mathrm{id}} = H_{\mathrm{id}}.$$

Equation (23) as written contains a long-ranged nonlocal diffusion term on the second line, which is there even when there are no direct interactions. For an ideal gas, the flux in the parenthesis on the second line of (23) becomes

$$\int \boldsymbol{\mathcal{R}}(\boldsymbol{r} - \boldsymbol{r}') \frac{c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t)}{c^{(1)}(\boldsymbol{r}', t)} \boldsymbol{\nabla}' c^{(1)}(\boldsymbol{r}', t) \, d\boldsymbol{r}',$$

which is still not closed. For an ideal gas, the closure for the two-particle correlation function that Rex and Löwen [6, 7] suggest becomes

$$c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t) \approx c^{(1)}(\boldsymbol{r}, t) \, c^{(1)}(\boldsymbol{r}', t).$$

After also making this approximation we can write the second line of (23) in the form

$$c^{(1)}(\boldsymbol{r}, t) \int \boldsymbol{\mathcal{R}}(\boldsymbol{r} - \boldsymbol{r}') \, \boldsymbol{\nabla}' c^{(1)}(\boldsymbol{r}', t) \, d\boldsymbol{r}',$$

which vanishes after an integration by parts due to the incompressibility condition (11).

The above considerations for an ideal gas suggest that (23) should be written in a form that separates the ideal from the non-ideal contributions,

$$\partial_t c^{(1)}\left(\boldsymbol{r}, t\right) = \chi \boldsymbol{\nabla}^2 c^{(1)}\left(\boldsymbol{r}, t\right) + \left(k_B T\right)^{-1} \chi \boldsymbol{\nabla} \cdot \left(c^{(1)}\left(\boldsymbol{r}, t\right) \boldsymbol{\nabla} \frac{\delta F_{\mathrm{exc}}}{\delta c^{(1)}\left(\boldsymbol{r}, t\right)}\right)$$

$$+ \left(k_B T\right)^{-1} \boldsymbol{\nabla} \cdot \left(\int \boldsymbol{\mathcal{R}}\left(\boldsymbol{r} - \boldsymbol{r}'\right) c^{(2)}\left(\boldsymbol{r}, \boldsymbol{r}', t\right) \boldsymbol{\nabla}' \frac{\delta F_{\mathrm{exc}}}{\delta c^{(1)}\left(\boldsymbol{r}', t\right)} d\boldsymbol{r}'\right). \tag{24}$$

where F_{exc} is the *excess* (over the ideal gas) free-energy functional. The first line is the equation obtained for uncorrelated walkers by Marconi and Tarazona [4]. In the last line of (24), $\boldsymbol{\mathcal{R}}$ is long-ranged but one expects that the remainder of the integrand is short-ranged far from phase transitions in some sense [27] and therefore the result will be nonlocal only over scales that represents that typical correlation length in the microstructure of the system. Making this more precise requires some further approximations and is beyond the scope of this work. It is interesting to note that the first line in (24) can be written in functional notation as

$$- \int d\boldsymbol{r}' \, \mathcal{M}_{\mathrm{ind}} \left[c^{(1)}(\cdot, t); \, \boldsymbol{r}, \boldsymbol{r}'\right] \frac{\delta F}{\delta c^{(1)}\left(\boldsymbol{r}', t\right)},$$

which, surprisingly, involves $\mathcal{M}_{\mathrm{ind}}$ even though $\mathcal{M}_{\mathrm{ind}}$ does not appear in the original dynamics. Further work is necessary to explore how well closures such as (24) describe collective diffusion in both confined and unconfined dilute and semi-dilute colloidal suspensions.

B. Averaging over initial conditions

As written, the fluctuating DDFT equation (15) is a nonlinear non-local SPDE that appears of little practical utility; solving it is no easier than solving (1) using Brownian Dynamics [25]. This is so even in the absence of direct interactions because of the nonlocal diffusive flux term $c(\boldsymbol{r}, t) \int \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' c(\boldsymbol{r}', t) \, d\boldsymbol{r}'$. However, an important observation, previously missed, is that the incompressibility of the fluid mediating the hydrodynamic correlations implies that the correlation tensor is divergence free. This implies that the nonlocal diffusive flux term vanishes, and therefore, in the absence of direct interactions the fluctuating DDFT equation is the *linear* and *local* stochastic advection-diffusion equation (12).

It is important to emphasize that (12) is mathematically well-behaved and *does* have utility beyond that of formal equations such as (16) because it can be averaged over initial conditions (rather than over realizations of the noise) [11]. Specifically, let us assume that the initial positions of the particles are uniformly sampled from an equilibrium ensemble *constrained* to have a specified mean $c_0\left(\boldsymbol{r}, t\right)$ via a suitable external or chemical potential [4, 7]. For noninteracting walkers, this simply amounts to choosing the initial particle positions independently from a probability distribution $\sim c_0\left(\boldsymbol{r}, t\right)$. Because of the linearity of (12) we can trivially average it over this ensemble of initial conditions; the equation remains the same but now the initial condition is the *smooth* $c\left(\boldsymbol{r}, 0\right) = c_0$ rather than a spiky sum of delta functions. This is useful if one wants to describe particular instances (realizations) of the dynamics starting from a random configuration of particles. For example, consider a fluorescence recovery after photobleaching (FRAP) experiment [32] in which a random subset of the particles uniformly distributed *below* a given plane are fluorescently labeled at $t = 0$ and then allowed to diffuse freely. This can be modeled by solving (1) for a finite collection of particles, but, equivalently, one can solve (using computational fluid dynamics techniques) the Eulerian equation (12) with $c\left(\boldsymbol{r}, 0\right) =$ const. above the given plane and $c\left(\boldsymbol{r}, 0\right) = 0$ below it, to obtain the probability $\sim c\left(\boldsymbol{r}, t\right)$ of finding a particle at position $\boldsymbol{r}$ for a *specific* instance of the noise $\boldsymbol{w}\left(\boldsymbol{r}, t\right)$. More general smooth initial conditions are also possible, e.g., a Gaussian profile corresponding to a nonuniform laser beam intensity in a FRAP experiment.

Because of its nonlinearity, averaging (15) over initial conditions is nontrivial and requires further approximations that are beyond the scope of this work. We believe such averaging could lead to descriptions that describe collective diffusion at *all* scales, from the microscopic to the macroscopic, in a manner more suitable for numerical approximations than (1).

C. Spatial Averaging

It is important to contrast the fluctuating diffusion (24) that describes the microscopic dynamics to the equation obtained by considering a *macroscopic* limit and coarse-graining the concentration over many particles, rather than over realizations of the noise. The literature on the subject is large [14, 27, 33–35] and we make no attempt to review it here, rather, we summarize some key results. Let us denote with $\bar{c}\left(\boldsymbol{r}, t\right) \approx$

$_{310}$ $c_\xi(\boldsymbol{r}, t)$ the macroscopic concentration, which, roughly speaking, can be thought of as $c(\boldsymbol{r}, t)$ averaged over
$_{311}$ a region of macroscopic size ξ (i.e., a region containing many particles and typical size much larger than the
$_{312}$ interaction range of the pairwise potential). A precise mathematical definition is possible by suitable rescaling
$_{313}$ of space and time, see Refs. [36–39]; equivalently, one can consider the Fourier transform of $c(\boldsymbol{r}, t)$ in the limit
$_{314}$ of small wavenumbers. It has been demonstrated rigorously [37] that for uncorrelated walkers interacting
$_{315}$ with short-ranged potentials the macroscopic concentration obeys a nonlinear but local Fick's law [33]

$$\partial_t \bar{c} = \chi \boldsymbol{\nabla}^2 \Pi(\bar{c}) = \chi \boldsymbol{\nabla} \cdot \left(\frac{d\Pi(\bar{c})}{d\bar{c}} \boldsymbol{\nabla} \bar{c} \right).$$

$_{316}$ Here $\Pi(\bar{c})$ is the osmotic pressure of the suspension at thermodynamic (local) equilibrium with uniform
$_{317}$ concentration $\bar{c}$ (for an ideal gas $\Pi(\bar{c}) = \bar{c}\, k_B T$), $\Pi(\bar{c}) = \bar{c}(df/d\bar{c}) - f$, where $f(\bar{c})$ is the thermodynamic
$_{318}$ *equilibrium* free-energy density of a *macroscopic* system with uniform density $\bar{c}$.
$_{319}$ We are, however, not aware of any mathematical techniques that can be used to rigorously justify Fick's
$_{320}$ law in the presence of long-ranged hydrodynamic correlations. Felderhof [27] argues that from a variant
$_{321}$ of (17) one can obtain Fick's law with a diffusion coefficient that depends on concentration and gives
$_{322}$ a low-density expansion of the collective diffusion coefficient (c.f. (4.24) in [27]) that matches the one
$_{323}$ obtained by Batchelor [35] using Einstein's formula. It is important to point out that at later stages of his
$_{324}$ argument Felderhof makes key use of the divergence-free nature of the hydrodynamic correlations [4], which
$_{325}$ he also emphasizes follows from the incompressibility of the fluid (c.f. (4.13) in [27]). While Felderhof and
$_{326}$ other authors in the physics literature write Fick's law as an equation for $c^{(1)}$ it is clear from the derivations
$_{327}$ that an assumption is being made that $c^{(1)}$ varies little and slowly in space. It is important to remember
$_{328}$ that $c^{(1)}(\boldsymbol{r}, t)$ and $\bar{c}(\boldsymbol{r}, t)$ are different objects, although one expects that in cases where $c^{(1)}$ varies slowly
$_{329}$ in space the two are closely related since ensemble and spatial averaging are expected to commute.
$_{330}$ Of particular interest is to understand collective diffusion over the broad-spectrum of *mesoscopic*
$_{331}$ length-scales, i.e., scales that are larger than σ, where σ is a typical microscopic length, but not so large
$_{332}$ that the hydrodynamic limit applies. For non-interacting uncorrelated walkers, the ensemble-averaged
$_{333}$ concentration follows the *same* diffusion equation (Fick's law) with the same diffusion coefficient at *all*
$_{334}$ scales, as seen from the linearity of (8). We demonstrated here that the same holds even in the presence
$_{335}$ of hydrodynamic correlations among the particles. Direct interactions appear to, however, complicate the
$_{336}$ picture and lead to non-local nonlinear terms like those in (24), and we do not know of any rigorous results
$_{337}$ in the mesoscopic regime. Non-equilibrium thermodynamics [28] and the theory of coarse-graining [30]
$_{338}$ provide guidance on the structure of the resulting equations but not their explicit form.
$_{339}$ In principle, an equation for the coarse-grained concentration (21) can be carried out by convolving
$_{340}$ (filtering) the right hand side of (15) with the kernel δ_ξ. In general this leads to an unclosed equation and
$_{341}$ further approximations are required. Once again the special case of an ideal gas is much simpler to tackle
$_{342}$ because (15) becomes the linear (12). In Ref. [11] we proposed how to carry out spatial coarse-graining
$_{343}$ by performing a *partial* ensemble average of (12) over fluctuations of the random velocity field $\boldsymbol{w}$ below
$_{344}$ the coarse-graining scale. Our argument, however, closely relied on the linearity of (12) and therefore only
$_{345}$ applies when there are incompressible hydrodynamic correlations but no direct interactions among the
$_{346}$ particles. The general conclusion of our work and other related works in the literature is that coarse-graining
$_{347}$ leads to effective dissipation (entropy production) with transport coefficients that must be *renormalized*
$_{348}$ in a way that takes into account the mesoscopic observation scale. The same undoubtly holds for any
$_{349}$ "free energy functional" that may appear in the mesoscopic equations. Carrying out such a renormalization
$_{350}$ of (15) remains a difficult but important challenge for the future.

$_{351}$ IV.   Conclusions

$_{352}$ Hydrodynamics plays an important role in colloidal suspensions and must be included in DDFT theories.
$_{353}$ Momentum transport in the fluid leads to hydrodynamic correlations among the diffusing particles and
$_{354}$ has important consequences for the collective diffusion not seen if one looks at the self-diffusion of a single
$_{355}$ particle in suspension. Starting from (3) as a model of these hydrodynamic correlations, we obtained a closed
$_{356}$ equation (15) for the instantaneous, fluctuating, or empirical concentration, the ensemble average of which
$_{357}$ (17) matches the DDFT equation previously obtained by Rex and Löwen [6, 7]. This generalizes the results

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[4] This part of the derivation of Felderhof inspired the rewriting (24).

358 of Dean [3] for the case of uncorrelated (independent) Brownian walkers to account for hydrodynamics,
359 and generalizes our prior results [11] to account for direct interactions among the particles.

360 A few comments about the physical reasoning behind (3) are in order. Note that the generic form (4)
361 does not fit (3) because the appearance of the Kronecker δ_{ij}. It can be shown that the requirement that the
362 mobility be positive semidefinite for any configuration of particles and any N implies that $\|\boldsymbol{\omega}_{12}(0)\|_2 \leq 1$;
363 if this holds as an equality then ^{5} $\boldsymbol{\omega}_{12}(0) = \boldsymbol{I}$ and therefore (3) holds. This has important physical
364 consequences that do not appear to have been widely appreciated. Notably, for two overlapping particles,
365 $\boldsymbol{q}_i = \boldsymbol{q}_j$, (3) predicts $\boldsymbol{M}_{ii} = \boldsymbol{M}_{jj} = \boldsymbol{M}_{ij} = \boldsymbol{M}_{ji}$, which implies that, in fact, the two particles continue
366 to move in synchrony forever, and $\boldsymbol{q}_i = \boldsymbol{q}_j$ for all times. By contrast, if $\|\boldsymbol{\omega}_{12}(0)\|_2 < 1$, as for the case of
367 independent Brownian walkers $\boldsymbol{\omega}_{12} = 0$, two particles released from the same position separate immediately.

368 We believe that it is physically more realistic to assume that the trajectories of nearby particles become
369 highly correlated rather than remain independent. Furthermore, two perfectly overlapping particles should
370 behave as if there is only a single particle at that location. The well-known Rotne-Prager mobility [12],
371 which was used by Rex and Löwen [6, 7] in their numerical calculations, *does* conform to (3). In our prior
372 work [11], we used a model based on fluctuating hydrodynamics [23, 40, 41], which, in the limit of infinite
373 Schmidt number (momentum diffusion much faster than particle diffusion) converges to (1) with (3) and
374 a covariance operator [21, 25]

$$\boldsymbol{\mathcal{R}}\left(\boldsymbol{r}_1, \boldsymbol{r}_2\right) = \frac{k_B T}{\eta} \int \boldsymbol{\sigma}\left(\boldsymbol{r}_1, \boldsymbol{r}'\right) \boldsymbol{G}\left(\boldsymbol{r}', \boldsymbol{r}''\right) \boldsymbol{\sigma}^T\left(\boldsymbol{r}_2, \boldsymbol{r}''\right) d\boldsymbol{r}' d\boldsymbol{r}'', \tag{25}$$

375 where $\boldsymbol{G}$ is the Green's function for the steady Stokes equation with unit viscosity and appropriate
376 boundary conditions. For unbounded three-dimensional systems $\boldsymbol{G}$ is the Oseen tensor $\boldsymbol{G}\left(\boldsymbol{r}', \boldsymbol{r}''\right) =$
377 $(8\pi r)^{-1}\left(\boldsymbol{I} + r^{-2}\boldsymbol{r} \otimes \boldsymbol{r}\right)$, where $\boldsymbol{r} = \boldsymbol{r}' - \boldsymbol{r}''$. Here $\boldsymbol{\sigma}$ is a smoothing kernel that filters out features at scales
378 below the size of the diffusing particles σ, e.g., $\boldsymbol{\sigma}$ could be a Gaussian with standard deviation σ. The
379 self-diffusion coefficient χ given by (25) obeys a Stokes-Einstein formula, in three dimensions, $\chi \sim k_B T / (\eta \sigma)$
380 [11]. When the particles are far apart, $\|\boldsymbol{q}_i - \boldsymbol{q}_j\| \gg \sigma$, the mobility is well-approximated by the Oseen
381 tensor, $\boldsymbol{M}_{ij}\left(\boldsymbol{q}_i, \boldsymbol{q}_j\right) \approx \eta^{-1}\boldsymbol{G}\left(\boldsymbol{q}_i, \boldsymbol{q}_j\right)$. At short distances the divergence of the Oseen tensor is mollified
382 by the filter, and (25) gives a pairwise mobility very similar to the Rotne-Prager mobility (5) widely-used
383 in BD simulations [25]. Note that (11) follows from the incompressibility of the Green's function $\boldsymbol{G}$.

384 Numerical methods to solve (12) and (1), along with an extensive visual and quantitative analysis of the
385 surprising characteristics of the solution can be found in Ref. [11]. A key observation is that, due to the Ito
386 nature of the hydrodynamic term $-\boldsymbol{w} \cdot \boldsymbol{\nabla} c$ in (12), the ensemble-averaged concentration continues to follow
387 the local Fick's law (8), despite the presence of hydrodynamic correlations among the diffusing particles. Note,
388 however, that the behavior of each *instance* (relization) of the stochastic process $c(\boldsymbol{r}, t)$ is rather distinct from
389 the behavior of the mean concentration, as discussed extensively in our prior work [11]. In particular, the
390 fluctuating equation (12) is non-dissipative (reversible), while Fick's law (8) is dissipative (irreversible). In the
391 presence of large concentration gradients the solutions of (12) exhibit characteristic long-ranged correlations
392 (giant fluctuations) that are quite distinct from the case of uncorrelated walkers [42–45]. This indicates that
393 the mathematical structure and the physical behavior of (16) is very different from that of (15) because
394 hydrodynamics affects the fluctuations of the concentration in crucial ways. This fact is well-known in nonequi-
395 librium statistical mechanics circles, and recent experiments [44] have demonstrated how *giant* concentration
396 fluctuations can arise for a simple polymer solution out of equilibrium in the absence of gravity. Nonequi-
397 librium concentration fluctuations have also been measured in gravity for a nanocolloidal suspension [45].

398 The striking difference between correlated and uncorrelated walkers is somewhat surprising. After all, one
399 would expect that, if the correlations are sufficiently weak in a certain sense (e.g., they decay rapidly with
400 distance ^{6}), (15) should converge to (16). It is important to emphasize, however, that (16) corresponds to the
401 physically unrealistic case of particles performing uncorrelated random motions even when they overlap. In
402 reality, it is the solvent molecules that have to kick the colloidal particles, and nearby particles must become
403 correlated because their diffusion is caused by the motion of correlated fluid molecules. Let us assume for a
404 moment that (1) holds with an isotropic smooth $\boldsymbol{\mathcal{R}}\left(\boldsymbol{r} - \boldsymbol{r}'\right)$ that is nonzero only if the two particles are within

---

^{5} Observe that $\boldsymbol{\omega}_{12}(0)$ must be rotationally invariant and therefore has to be the identity matrix.

^{6} It must be pointed out, however, that incompressible hydrodynamic correlations such as the Rotne-Prager tensor, must be long ranged because of the incompressibility constraint.

a distance σ' apart, and has a finite value at the origin, $\boldsymbol{\mathcal{R}}(0) = \chi \boldsymbol{I}$. Let us also account for the fact that the diffusing particles themselves are not point particles but have a physical size σ, and consider the coarse-grained concentration (21) for $\xi \sim \sigma$. The case considered by Dean corresponds to the double limit $\sigma \to 0$ and $\sigma' \to 0$, but the order of these limits is not *a priori* clear. Formal manipulations show that (15) converges in a certain sense to (16) if one takes the limit $\sigma \to 0$ first and then takes the limit $\sigma' \to 0$. It is an interesting open question what happens if the order of the limits is reversed, or if σ and σ' go to zero simultaneously. Such calculations will shed further light on the nature of diffusion in liquid suspensions and mixtures over a much broader spectrum of length and time scales than described by Fick's law with phenomenological diffusion constants.

Appendix

A. Equations for the Empirical Concentration

In this Appendix we present the detailed derivation of (15) and (17). In the beginning, we will consider the case of no direct interactions among the particles, $U = 0$, and subsequently add the direct forces. The assumption that the covariance operator $\boldsymbol{\mathcal{R}}$ is symmetric positive-semidefinite is equivalent to the requirement that the mobility matrix $\boldsymbol{M}(\boldsymbol{Q})$ be symmetric positive semi-definite for all $\boldsymbol{Q}$, and implies that there exists an infinite countable set of eigenfunctions $\boldsymbol{\phi}_k(\boldsymbol{r})$ that factorize (diagonalize) the covariance operator,

$$\sum_k \boldsymbol{\phi}_k(\boldsymbol{r}) \otimes \boldsymbol{\phi}_k(\boldsymbol{r}') = \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}').$$

Note that if (11) holds then the eigenfunctions of $\boldsymbol{\mathcal{R}}$ are also incompressible, $\boldsymbol{\nabla} \cdot \boldsymbol{\phi}_k(\boldsymbol{r}) = 0$.

1. Stratonovich form

It is not hard to show that in the absence of direct interactions (1,3) corresponds to the Stratonovich equation for the position of an individual tracer $i = 1, \ldots, N$,

$$d\boldsymbol{q}_i = \sum_{j=1}^N \boldsymbol{b}(\boldsymbol{q}_i, \boldsymbol{q}_j)dt + \sqrt{2}\sum_k \boldsymbol{\phi}_k(\boldsymbol{q}_i) \circ dB_k, \tag{A1}$$

where $\circ$ denotes a Stratonovich product, B_k are independent Brownian motions, and we defined

$$\boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r}') = \boldsymbol{\nabla}' \cdot \boldsymbol{\mathcal{R}}(\boldsymbol{r}, \boldsymbol{r}') = \sum_k \boldsymbol{\phi}_k(\boldsymbol{r}) \boldsymbol{\nabla} \cdot \boldsymbol{\phi}_k(\boldsymbol{r}'). \tag{A2}$$

Note that when the incompressibility condition (11) holds $\boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r}') = 0$.

For the Stratonovich interpretation we can use ordinary calculus to write

$$
\begin{aligned}
dc(\boldsymbol{r}, t) = &-\sum_{i,j=1}^N \boldsymbol{b}(\boldsymbol{q}_i(t), \boldsymbol{q}_j(t)) \cdot \boldsymbol{\nabla}\delta(\boldsymbol{r} - \boldsymbol{q}_i(t))dt \\
&- \sqrt{2}\sum_{i=1}^N \sum_k \boldsymbol{\phi}_k(\boldsymbol{q}_i(t)) \cdot \boldsymbol{\nabla}\delta(\boldsymbol{r} - \boldsymbol{q}_i(t)) \circ dB_k(t)
\end{aligned}
\tag{A3}
$$

Using integration by parts and properties of the delta function we can write this as a closed-form equation for c,

$$
\begin{aligned}
dc(\boldsymbol{r}, t) = &-\boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r}')c(\boldsymbol{r}', t)d\boldsymbol{r}' \right) dt \\
&- \sqrt{2}\sum_k \boldsymbol{\nabla} \cdot (\boldsymbol{\phi}_k(\boldsymbol{r})c(\boldsymbol{r}, t)) \circ dB_k(t)
\end{aligned}
\tag{A4}
$$

or, after recalling the definition of $\boldsymbol{b}$ in (A2) and performing an integration by parts to transfer the gradient to c,

$$dc(\boldsymbol{r}, t) = \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' c(\boldsymbol{r}', t) d\boldsymbol{r}' \right) dt \\ - \sqrt{2} \sum_k \boldsymbol{\nabla} \cdot (\boldsymbol{\phi}_k(\boldsymbol{r}) c(\boldsymbol{r}, t)) \circ dB_k(t). \tag{A5}$$

When the incompressibility condition (11) is satisfied, $\boldsymbol{b} = 0$ and (A4) implies that

$$dc(\boldsymbol{r}, t) = -\sqrt{2} \sum_k \boldsymbol{\phi}_k(\boldsymbol{r}) \cdot \boldsymbol{\nabla} c(\boldsymbol{r}, t) \circ dB_k(t), \tag{A6}$$

which is exactly identical to the Stratonovich form of the equation we obtained in Ref. [11] by rather different means. Here we can identify $\boldsymbol{w}(\boldsymbol{r}, t) = \sqrt{2} \sum_k \boldsymbol{\phi}_k(\boldsymbol{r}) dB_k(t)$ as a random velocity field with covariance given by (13). While the Stratonovich form of the equation is the simplest, the Ito form is the most convenient for performing an ensemble average to obtain an equation for the average concentration $c^{(1)}$.

2. Ito form

In the Ito interpretation, (A1) reads

$$d\boldsymbol{q}_i = \boldsymbol{a}(\boldsymbol{q}_i) dt + \sum_{j \neq i}^N \boldsymbol{b}(\boldsymbol{q}_i, \boldsymbol{q}_j) dt + \sqrt{2} \sum_k \boldsymbol{\phi}_k(\boldsymbol{q}_i) dB_k, \tag{A7}$$

where we defined

$$\boldsymbol{a}(\boldsymbol{r}) = \boldsymbol{\nabla} \cdot \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}) = \sum_k \boldsymbol{\phi}_k(\boldsymbol{r}) \boldsymbol{\nabla} \cdot \boldsymbol{\phi}_k(\boldsymbol{r}) + \sum_k \boldsymbol{\phi}_k(\boldsymbol{r}) \cdot \boldsymbol{\nabla} \boldsymbol{\phi}_k(\boldsymbol{r}) = \boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r}) + \boldsymbol{g}(\boldsymbol{r}), \tag{A8}$$

and

$$\boldsymbol{g}(\boldsymbol{r}) = \sum_k \boldsymbol{\phi}_k(\boldsymbol{r}) \cdot \boldsymbol{\nabla} \boldsymbol{\phi}_k(\boldsymbol{r}). \tag{A9}$$

The Ito equation (A7) can also be written as

$$d\boldsymbol{q}_i = \boldsymbol{g}(\boldsymbol{q}_i) dt + \sum_{j=1}^N \boldsymbol{b}(\boldsymbol{q}_i, \boldsymbol{q}_j) dt + \sqrt{2} \sum_k \boldsymbol{\phi}_k(\boldsymbol{q}_i) dB_k, \tag{A10}$$

which will be the most convenient for our calculation. Note that when the incompressibility condition (11) holds,

$$\boldsymbol{a}(\boldsymbol{r}) = \boldsymbol{g}(\boldsymbol{r}) = \boldsymbol{\nabla} \cdot \boldsymbol{\chi}(\boldsymbol{r}) \tag{A11}$$

is the divergence of the diffusion tensor, which vanishes for translationally-invariant systems.

Using Ito calculus, we can now write an equation for the empirical concentration,

$$dc(\boldsymbol{r}, t) = -\sum_{i=1}^N \left(\boldsymbol{g}(\boldsymbol{q}_i(t)) + \sum_{j=1}^N \boldsymbol{b}(\boldsymbol{q}_i(t), \boldsymbol{q}_j(t)) \right) \cdot \boldsymbol{\nabla} \delta(\boldsymbol{r} - \boldsymbol{q}_i(t)) dt \\ - \sqrt{2} \sum_{i=1}^N \sum_k \boldsymbol{\phi}_k(\boldsymbol{q}_i(t)) \cdot \boldsymbol{\nabla} \delta(\boldsymbol{r} - \boldsymbol{q}_i(t)) dB_k(t) \\ + \sum_{i=1}^N \sum_k \boldsymbol{\phi}_k(\boldsymbol{q}_i(t)) \boldsymbol{\phi}_k(\boldsymbol{q}_i(t)) : \boldsymbol{\nabla}\boldsymbol{\nabla} \delta(\boldsymbol{r} - \boldsymbol{q}_i(t)) dt \tag{A12}$$

Using integration by parts and properties of the delta function we can write this as a closed-form equation for c,

$$
dc(\boldsymbol{r}, t) = -\boldsymbol{\nabla} \cdot (\boldsymbol{g}(\boldsymbol{r})c(\boldsymbol{r}, t)) \, dt + \boldsymbol{\nabla}\boldsymbol{\nabla} : (\mathcal{R}(\boldsymbol{r}, \boldsymbol{r})c(\boldsymbol{r}, t)) \, dt
$$
$$
- \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r}')c(\boldsymbol{r}', t)d\boldsymbol{r}' \right) dt \tag{A13}
$$
$$
- \sqrt{2} \sum_k \boldsymbol{\nabla} \cdot (\boldsymbol{\phi}_k(\boldsymbol{r})c(\boldsymbol{r}, t)) \, dB_k(t),
$$

which can further be simplified to

$$
dc(\boldsymbol{r}, t) = \boldsymbol{\nabla} \cdot (\mathcal{R}(\boldsymbol{r}, \boldsymbol{r})\boldsymbol{\nabla} c(\boldsymbol{r}, t) + \boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r})c(\boldsymbol{r}, t)) \, dt
$$
$$
+ \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}')\boldsymbol{\nabla}'c(\boldsymbol{r}', t)d\boldsymbol{r}' \right) dt \tag{A14}
$$
$$
- \sqrt{2} \sum_k \boldsymbol{\nabla} \cdot (\boldsymbol{\phi}_k(\boldsymbol{r})c(\boldsymbol{r}, t)) \, dB_k(t).
$$

Here we can identify $\boldsymbol{w}(\boldsymbol{r}, t) = \sqrt{2}\sum_k \boldsymbol{\phi}_k(\boldsymbol{r})dB_k(t)$ as a random velocity field with covariance given by (13). Upon averaging over realizations of the noise the fluctuating term drops out in the Ito interpretation, giving a non-local diffusion equation for the mean concentration $c^{(1)}(\boldsymbol{r}, t) = \langle c(\boldsymbol{r}, t) \rangle$

$$
\partial_t c^{(1)}(\boldsymbol{r}, t) = \boldsymbol{\nabla} \cdot \left(\boldsymbol{\chi}(\boldsymbol{r})\boldsymbol{\nabla} c^{(1)}(\boldsymbol{r}, t) + \boldsymbol{b}(\boldsymbol{r}, \boldsymbol{r})c^{(1)}(\boldsymbol{r}, t) \right)
$$
$$
+ \boldsymbol{\nabla} \cdot \left(\int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}') \langle c(\boldsymbol{r}, t) \, \boldsymbol{\nabla}'c(\boldsymbol{r}', t) \rangle \, d\boldsymbol{r}' \right). \tag{A15}
$$

By noting that the two-particle correlation function is

$$
c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t) = \langle c(\boldsymbol{r}, t) \, c(\boldsymbol{r}', t) \rangle - \langle c(\boldsymbol{r}, t) \rangle \, \delta(\boldsymbol{r} - \boldsymbol{r}'), \tag{A16}
$$

we see after an integration by parts that (A15) is equivalent to

$$
\partial_t c^{(1)}(\boldsymbol{r}, t) = \boldsymbol{\nabla} \cdot \left(\boldsymbol{\chi}(\boldsymbol{r})\boldsymbol{\nabla} c^{(1)}(\boldsymbol{r}, t) \right) + \boldsymbol{\nabla} \cdot \left(\int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}')\boldsymbol{\nabla}'c^{(2)}(\boldsymbol{r}, \boldsymbol{r}', t) \, d\boldsymbol{r}' \right),
$$

which, for a translationally-invariant system, is exactly the equation (9) obtained by Rex and Löwen.
When the incompressibility condition (11) holds, Eq. (A13) reduces to the fluctuating Fick's law

$$
dc(\boldsymbol{r}, t) = \boldsymbol{\nabla} \cdot (\boldsymbol{\chi}(\boldsymbol{r})\boldsymbol{\nabla} c(\boldsymbol{r}, t)) \, dt - \sqrt{2} \sum_k \boldsymbol{\phi}_k(\boldsymbol{r}) \cdot \boldsymbol{\nabla} c(\boldsymbol{r}, t)dB_k(t), \tag{A17}
$$

which is exactly the stochastic advection-diffusion equation (12). In this case the mean follows Fick's law

$$
\partial_t c^{(1)}(\boldsymbol{r}, t) = \boldsymbol{\nabla} \cdot \left(\boldsymbol{\chi}(\boldsymbol{r})\boldsymbol{\nabla} c^{(1)}(\boldsymbol{r}, t) \right),
$$

and the non-local diffusion term involving $c^{(2)}$ disappears since $\boldsymbol{b} = 0$.

3. Direct interactions

If we include direct interactions among the particles of the form (14), the Stratonovich equation of motion (A1) becomes

$$
d\boldsymbol{q}_i = \sum_{j=1}^N \boldsymbol{b}(\boldsymbol{q}_i, \boldsymbol{q}_j)dt + \sqrt{2} \sum_k \boldsymbol{\phi}_k(\boldsymbol{q}_i) \circ dB_k
$$
$$
+ (k_B T)^{-1} \sum_{j=1}^N \mathcal{R}(\boldsymbol{q}_i, \boldsymbol{q}_j) \left(\boldsymbol{f}_1(\boldsymbol{q}_j) + \sum_{k \neq j} \boldsymbol{f}_2(\boldsymbol{q}_j, \boldsymbol{q}_k) \right) dt \tag{A18}
$$

where we have defined the external and pairwise forces

$$\boldsymbol{f}_1(\boldsymbol{r}) = -\boldsymbol{\nabla} U_1(\boldsymbol{r}), \qquad \boldsymbol{f}_2(\boldsymbol{r}, \boldsymbol{r}') = -\boldsymbol{\nabla} U_2(\boldsymbol{r}, \boldsymbol{r}'). \tag{A19}$$

For simplicity, and without loss of generality, we will assume that there is no self-force coming from the pairwise interactions, $\boldsymbol{f}_2(\boldsymbol{r}, \boldsymbol{r}) = 0$.

The new term in (A18) adds the following term to the drift in (A3):

$$
\begin{aligned}
&-(k_B T)^{-1} \sum_{i,j=1}^{N} \mathcal{R}(\boldsymbol{q}_i, \boldsymbol{q}_j) \left(\boldsymbol{f}_1(\boldsymbol{q}_j) + \sum_{k \neq j} \boldsymbol{f}_2(\boldsymbol{q}_j, \boldsymbol{q}_k) \right) \cdot \boldsymbol{\nabla} \delta(\boldsymbol{r} - \boldsymbol{q}_i) \\
&= -(k_B T)^{-1} \boldsymbol{\nabla} \cdot \left[\sum_{i,j=1}^{N} \mathcal{R}(\boldsymbol{q}_i, \boldsymbol{q}_j) \left(\boldsymbol{f}_1(\boldsymbol{q}_j) + \sum_{k=1}^{N} \boldsymbol{f}_2(\boldsymbol{q}_j, \boldsymbol{q}_k) \right) \delta(\boldsymbol{r} - \boldsymbol{q}_i) \right] \\
&= -(k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(\sum_{i,j=1}^{N} \int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{f}_1(\boldsymbol{r}') \delta(\boldsymbol{r} - \boldsymbol{q}_i) \delta(\boldsymbol{r}' - \boldsymbol{q}_j) \, d\boldsymbol{r}' \right) \\
&\quad - (k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(\sum_{i,j,k=1}^{N} \int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{f}_2(\boldsymbol{r}', \boldsymbol{r}'') \delta(\boldsymbol{r} - \boldsymbol{q}_i) \delta(\boldsymbol{r}' - \boldsymbol{q}_j) \delta(\boldsymbol{r}'' - \boldsymbol{q}_k) \, d\boldsymbol{r}' d\boldsymbol{r}'' \right)
\end{aligned}
\tag{A20}
$$

This can also be written in terms of the empirical concentration as

$$
\begin{aligned}
&-(k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{f}_1(\boldsymbol{r}') c(\boldsymbol{r}', t) \, d\boldsymbol{r}' \right) \\
&-(k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{f}_2(\boldsymbol{r}', \boldsymbol{r}'') c(\boldsymbol{r}', t) c(\boldsymbol{r}'', t) \, d\boldsymbol{r}' d\boldsymbol{r}'' \right),
\end{aligned}
\tag{A21}
$$

or, in terms of the potentials U_1 and U_2, as

$$
\begin{aligned}
&(k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' U_1(\boldsymbol{r}') c(\boldsymbol{r}', t) \, d\boldsymbol{r}' \right) \\
&+(k_B T)^{-1} \boldsymbol{\nabla} \cdot \left(c(\boldsymbol{r}, t) \int \mathcal{R}(\boldsymbol{r}, \boldsymbol{r}') \boldsymbol{\nabla}' U_2(\boldsymbol{r}', \boldsymbol{r}'') c(\boldsymbol{r}', t) c(\boldsymbol{r}'', t) \, d\boldsymbol{r}' d\boldsymbol{r}'' \right).
\end{aligned}
\tag{A22}
$$

Adding these terms to the right hand side of (A14) (or, in the Stratonovich interpretation, to (A5)) gives our final result (15). Remarkably, this is a closed equation for the fluctuating concentration just as in the case of uncorrelated particles [3].

Taking an ensemble average of the new terms (A22) leads to terms involving the two-particle (A16) and three-particle correlation function

$$
\begin{aligned}
c^{(3)}(\boldsymbol{r}, \boldsymbol{r}', \boldsymbol{r}'', t) = {}& \langle c(\boldsymbol{r}, t) c(\boldsymbol{r}', t) c(\boldsymbol{r}'', t) \rangle \\
&- \tfrac{1}{2} \langle c(\boldsymbol{r}, t) c(\boldsymbol{r}', t) \rangle \left(\delta(\boldsymbol{r} - \boldsymbol{r}'') + \delta(\boldsymbol{r}' - \boldsymbol{r}'') \right) \\
&- \tfrac{1}{2} \langle c(\boldsymbol{r}, t) c(\boldsymbol{r}'', t) \rangle \left(\delta(\boldsymbol{r} - \boldsymbol{r}') + \delta(\boldsymbol{r}' - \boldsymbol{r}'') \right) \\
&- \tfrac{1}{2} \langle c(\boldsymbol{r}', t) c(\boldsymbol{r}'', t) \rangle \left(\delta(\boldsymbol{r} - \boldsymbol{r}') + \delta(\boldsymbol{r} - \boldsymbol{r}'') \right) \\
&+ \langle c(\boldsymbol{r}, t) \rangle \delta(\boldsymbol{r} - \boldsymbol{r}') \delta(\boldsymbol{r}' - \boldsymbol{r}''),
\end{aligned}
\tag{A23}
$$

which, after some algebra, gives our final result (17) for the average concentration.

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