Colored-noise Fokker-Planck equation for the shear-induced self-diffusion process of non-Brownian particles

Laura J. Lukassen* and Martin Oberlack†

Graduate School of Excellence Computational Engineering, TU Darmstadt and Department of Mechanical Engineering, Chair of Fluid Dynamics, TU Darmstadt, Germany

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In the literature, it is pointed out that non-Brownian particles tend to show shear-induced diffusive behavior due to hydrodynamic interactions. Several authors indicate a long correlation time of the particle velocities in comparison to Brownian particle velocities modeled by a white noise. This work deals with the derivation of a Fokker-Planck equation both in position and velocity space which describes the process of shear-induced self-diffusion, whereas, so far, this problem has been described by Fokker-Planck equations restricted to position space. The long velocity correlation times actually would necessitate large time-step sizes in the mathematical description of the problem in order to capture the diffusive regime. In fact, time steps of specific lengths pose problems to the derivation of the corresponding Fokker-Planck equation because the whole particle configuration changes during long time-step sizes. On the other hand, small time-step sizes, i.e., in the range of the velocity correlation time, violate the Markov property of the position variable. In this work we regard the problem of shear-induced self-diffusion with respect to the Markov property and reformulate the problem with respect to small time-step sizes. In this derivation, we regard the nondimensionalized Langevin equation and develop a new compact form which allows us to analyze the Langevin equation for all time scales of interest for both Brownian and non-Brownian particles starting from a single equation. This shows that the Fokker-Planck equation in position space should be extended to a colored-noise Fokker-Planck equation in both position and colored-noise velocity space, which we will derive.

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I. INTRODUCTION

Non-Brownian particles in shear flow have become a considerably discussed topic throughout the literature. As the name implies, non-Brownian particles do not perform the well-known Brownian motion, which is an infinitely short correlated motion due to the molecule pushes of the surrounding solvent. Still, also non-Brownian particles are found to perform diffusive motion. This type of diffusion has been found to occur for particles with negligible inertia in Stokes flow in the absence of any kind of Brownian or turbulent diffusion [1]. Indeed, the so-called shear-induced self-diffusion results from hydrodynamic interactions between particles in the flow. This remarkable phenomenon has been subject to various theoretical, experimental, and numerical works, which are summarized below. The starting point of theoretical considerations is the two-particle interaction of purely hydrodynamically interacting particles with negligible inertia in Stokes flow as described by Batchelor and Green [2], [3] and in a review by Morris [4]. Though a viscous fluid is considered, mathematically this implies fore-aft symmetry of particle pair trajectories and reversibility [2–5].

From a theoretical point of view, this fore-aft symmetry and reversibility is refracted, the reasons being manifold, cf. Refs. [4,6–8], including surface roughness [9,10], weak Brownian motion and interparticle forces [5], and inertia [11].

The underlying process of shear-induced self-diffusion was evaluated experimentally, see, e.g., Refs. [1,12,13]. Numerical considerations can be found in, e.g., Refs. [14–17] using the Stokesian dynamics method [18] and in, e.g., Ref. [7] using so-called accelerated Stokesian dynamics [19]. Further, Pine *et al.* [20] compare experimental data with numerical results obtained by the Stokesian dynamics method. Even though Drazer *et al.* [15] use a repulsive force in their simulation, they argue that the diffusive behavior should also follow in the case of purely hydrodynamically interacting particles. In the present paper, which focuses on theoretical derivations, we also regard the case of purely hydrodynamically interacting particles and follow the assumptions in Refs. [7,8] that enough many-particle interactions suffice to create diffusive behavior.

The common models to describe particle flows either refer to an equation of motion (Langevin equation or Langevin-*like* equation) or the corresponding Fokker-Planck equation. One essential part of the present work is an asymptotics of the equation of motion in Sec. II, which includes the analysis for Brownian and non-Brownian particles for all apparent time scales, here the inertial relaxation time scale and the time scale of configurational changes, in a single starting equation. The asymptotics to be discussed here can be placed in a context of other asymptotics, e.g., Refs. [11,18,21–23], whereby we derive a new compact form which includes all other cases as special cases.

The corresponding Fokker-Planck equation depends on the time scale of interest and differs from the Brownian to the non-Brownian case. The basic version of the Fokker-Planck equation derived for Brownian particles provides an equation in position-velocity space (also indicated as phase space distribution), cf. Refs. [21,24,25]. Under the assumption that the velocity relaxes to equilibrium on a smaller time scale than the position, this form can be reduced to position space (Smoluchowski equation) [24,25]. Further, the reduction of the

*lukassen@gsc.tu-darmstadt.de
†oberlack@fdy.tu-darmstadt.de

phase space form to position form is analyzed by regarding multiple time scales in a single Fokker-Planck equation by Wycoff and Balazs [26] and extended to Brownian particles in shear flow by Subramanian and Brady [27].

Besides our interest in non-Brownian suspensions, we also pay attention to the analysis of Brownian particles because there are still cases when a reduction of the observed time scale reinforces the use of a coupled position-velocity space variable, namely the Kramers equation, see, e.g., Ref. [23]. In the non-Brownian case, we identify a similar problem and therefore propose to also switch to a coupled variable.

Instead of the N-particle Fokker-Planck equation, a one-particle representation decreases the complexity of the problem enormously which can be done, e.g., by deriving the coefficients for a one-particle Fokker-Planck equation from a one-particle equation of motion, as described technically in Ref. [23].

One very important aspect that was found in experiments, e.g., in Ref. [28], and numerical simulations, e.g., in Ref. [7] or Ref. [15], is that the shear-induced diffusion needs a certain time to develop, i.e., it is a long-time diffusion. Section II C gives a summary on diffusion. The time has to be long enough so enough many-particle interactions can have taken place [7]. In experiments, the authors in Ref. [28] indicate a different kind of diffusion for the small and intermediate time scales which is considerably smaller than the shear induced diffusion. Hence, we stick to Sierou and Brady‘s simulation results [7] which show a nondiffusive behavior of particle positions for short times due to long correlation times in the non-Brownian case. This poses limits to the derivation of the non-Brownian Fokker-Planck equation.

Existing Fokker-Planck equations for non-Brownian particles rather selectively model shear-induced diffusion in position space only using modified diffusion tensors. Santamaría-Holek *et al.* [29,30] study coupled Fokker-Planck equations in velocity and position space. They assume that the coupled Fokker-Planck equation can be transformed into a Fokker-Planck equation in position space only with a modified diffusion tensor incorporating thermal and nonthermal effects considering the second law of thermodynamcis. In this context, they report a breaking of the fluctuation-dissipation theorem due to an introduced shear flow which also has been investigated, e.g., by Mauri and Leporini [31]. Sierou and Brady [7] use a time-dependent diffusion coefficient for the small and intermediate time scales which becomes constant for long times and derive a Fokker-Planck equation in position space. Breedveld *et al.* [32] use the Fokker-Planck equation only in position space for the case of long-enough time intervals and measure the shear-induced self-diffusivity by use of a new correlation technique. Further, in Ref. [1], some of the authors from Ref. [32] and coworkers work with a Langevin equation with a colored-noise force with zero mean which contains the hydrodynamic influence of the particles onto each other. Based on that, they also aim at deriving the full position diffusion tensor in a modified form, with a colored-noise force in the equation of motion and, hence, overcome the common white-noise assumption. Nevertheless, they do not derive a Fokker-Planck equation in a coupled variable based on their Langevin equation.

Our work aims at deriving an alternative Fokker-Planck equation. Based on the assumption from Breedveld *et al.* [1] that the hydrodynamic influence of the particles is colored noise correlated, we transform the N-particle system of equations of motion to a one-particle equation for the particle velocity with a colored-noise-correlated hydrodynamic velocity component. The equations of motion for all N particles become independent. From that we aim at deriving the corresponding one-particle Fokker-Planck equation. Most importantly, we follow a new approach based on the Markov process assumption. The Markov process assumption, see, e.g., Ref. [23], allows us to combine the usage of a small time step for the underlying equations of motion and, at the same time, to describe the long-time behavior by the Fokker-Planck equation. The present paper primarily focuses on the investigation of the Markov process validity in the context of shear-induced diffusion. We will prove, by the appropriate choice of the time-step size, which is an important issue of the present work, that large time-step sizes are not allowed and small time-step sizes result in violation of the Markov property of the position variable. In this context we use the approach of a so-called colored-noise Fokker-Planck equation which has not been introduced in that context so far and model the hydrodynamic interaction via a hydrodynamic velocity component as an Ornstein-Uhlenbeck process. The general mathematical framework of the underlying colored-noise Fokker-Planck equation and Ornstein-Uhlenbeck processes can be found, e.g., in Ref. [23]. Ornstein-Uhlenbeck processes with exponential autocorrelation functions are a common scheme to describe colored-noise variables [23]. Further, the principle of velocity autocorrelation functions decaying exponentially in time has been observed in several particle-type situations as, e.g., sedimenting non-Brownian particles [33]. Our alternative equation is a colored-noise Fokker-Planck equation meant to carefully fulfill the Markov assumption and will be presented in Sec. III. Additionally, the solution based on a Gaussian distribution is shown and analyzed in Sec. IV. The resulting probability distribution is well founded.

II. FUNDAMENTAL MODEL DERIVATION

We study a three-dimensional (3D) homogeneous shear flow with purely hydrodynamically interacting non-Brownian particles. We assume a particle volume fraction such that enough particle-particle interactions can take place and shear-induced diffusion can arise [7]. For appropriate values of such particle volume fractions we refer to Ref. [7]. The works of Pine *et al.* [20], Santamaría-Holek *et al.* [30], and Sierou and Brady [7] show that the intensity of the position diffusion coefficients and the exact time after which diffusive behavior starts is influenced by the particle volume fraction. For our purposes it is most important that shear-induced behavior sets in generally so the influences of varying particle volume fractions are not investigated in the present work. Further, we consider periodic boundary conditions such that any wall interaction is avoided.

For the purpose of clearness we briefly define the difference among time, time step, and time scale, because of extensive use below. Time t is the independent variable on which the process may vary on the rather generic range $t \in [t_0,\infty)$. A time scale τ

defines the length of a process on which a significant change is observed, e.g., change in particle configuration, the period of a cyclic process, or the relaxation time scale of an exponentially decaying event. Finally, a time step $\triangle t$ is the fraction of the contemplated time scale (unless otherwise specified), e.g., $\triangle t = \tau/q$ with $q \in \mathbb{N}$, and therefore inherently coupled to τ. For the time scales to appear in the present work, it has to be chosen accordingly such that even in the limiting process of $\triangle t \to 0$, it is a given fraction of this time scale. That means that a time step $\triangle t \to 0$ with respect to a certain time scale will still be larger than the next smaller time scale (cf. Ref. [34]). Further, we would like to point out that a $\triangle t \ll$ or $\gg \tau$ in the present work refers to a $\triangle t$ on the next smaller or larger time scale, whereby a $\triangle t <$ or $> \tau$ indicates a time step on the very same time scale τ.

A. Particle motion described by Langevin equations

The starting point of the present considerations is an equation of motion for N particles in the system, which is built up according to the following [18]:

$$\boldsymbol{m}\frac{d\boldsymbol{U}}{dt} = \boldsymbol{F}^H + \boldsymbol{F}^P + \boldsymbol{F}^B. \tag{1}$$

The system consists of $6N$ equations, i.e. three degrees of freedom, respectively, for both translational and rotational motion for all N particles in the system. $\boldsymbol{m}$ is the generalized mass- and moment-of-inertia matrix and $\boldsymbol{U}$ is the combined vector of particle translational and rotational velocities. In the following, the hydrodynamic force-torque $\boldsymbol{F}^H$, the interparticle or external force-torque $\boldsymbol{F}^P$, and the Brownian force-torque $\boldsymbol{F}^B$ will be introduced. The particle Reynolds number, $\mathrm{Re}_p = \rho_f a^2\dot{\gamma}/\eta$, gives the inertia of the fluid with ρ_f as the fluid density, a as particle radius, $\dot{\gamma}$ as shear rate, and η as the suspending medium viscosity.

For $\mathrm{Re}_p \ll 1$, which is one of the key assumptions regarded in the present work, the hydrodynamic force-torque can be built up as shown in Refs. [18,35–37],

$$\boldsymbol{F}^H = -\boldsymbol{R}_{\mathrm{FU}}(\boldsymbol{U} - \boldsymbol{U}^\infty) + \boldsymbol{R}_{\mathrm{FE}} : \boldsymbol{E}^\infty. \tag{2}$$

The operator ":" is a contraction which reduces the multiplication of the rank 3 tensor $\boldsymbol{R}_{\mathrm{FE}}$ with the rank 2 tensor $\boldsymbol{E}^\infty$ to a vector (cf. Ref. [34]). $\boldsymbol{U}^\infty$ is the bulk shear flow. The translational components of the velocity vector of the bulk shear flow for one particle α are $(\dot{\gamma} y_\alpha,0,0)$, with y_α as the y position of this particle α. $\boldsymbol{E}^\infty$ is the symmetric part of the velocity gradient tensor, or the rate of strain tensor, i.e.,

$$\boldsymbol{E}^\infty = \frac{1}{2}\begin{bmatrix} 0 & \dot{\gamma} & 0 \\ \dot{\gamma} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}. \tag{3}$$

$\boldsymbol{R}_{\mathrm{FU}}$ and $\boldsymbol{R}_{\mathrm{FE}}$ are the configuration-dependent resistance matrices describing the connection between the force-torque ($\boldsymbol{F}$) and the relative velocity ($\boldsymbol{U} - \boldsymbol{U}^\infty$), as well as ($\boldsymbol{F}$) and the rate of strain ($\boldsymbol{E}^\infty$), respectively. $\boldsymbol{R}_{\mathrm{FU}}$ is used to describe the hydrodynamic interaction of the particles on each other while $\boldsymbol{R}_{\mathrm{FE}}$ gives the shear-induced disturbances in the flow field [34]. $\boldsymbol{R}_{\mathrm{FU}}$ and $\boldsymbol{R}_{\mathrm{FE}}$ can be built up such that they contain hydrodynamic far-field many-body interactions and near-field lubrication interactions, see Refs. [18,38] for the full derivation.

In a physical experiment, particles rather naturally do not coincide and, hence, it is also mandatory for numerical simulations to introduce an interparticle force $\boldsymbol{F}^P$ [35] to model physical effects as, for instance, particle roughness or residual Brownian motion [15]. In our theoretical considerations, we consider purely hydrodynamically interacting particles, and, thus, particles without interparticle forces, so subsequently $\boldsymbol{F}^P$ is set to zero.

Generally, for the present analysis, we regard a physical regime where particles are considered to have a larger size and mass than the particles or molecules of the surrounding fluid so the time scale of molecular collisions onto the particle is smaller than any other time scale of interest here [18,23]. Thus, the molecule impacts of the surrounding fluid onto a particle result in a randomly fluctuating force, i.e. the Brownian force $\boldsymbol{F}^B$ (cf. [21]). The Brownian force $\boldsymbol{F}^B$ is modeled such that it has zero average, i.e. $\langle \boldsymbol{F}^B \rangle = 0$, and an infinitely short autocorrelation time [18],

$$\langle \boldsymbol{F}^B(0)\boldsymbol{F}^B(t)\rangle = 2kT\,\boldsymbol{R}_{\mathrm{FU}}\delta(t). \tag{4}$$

In this context, the $\langle\rangle$ brackets are the ensemble average, k is the Boltzmann constant, and T is the absolute temperature, thus kT is the thermal energy. $\delta(t)$ is a Dirac correlated white noise which models the infinitely short autocorrelated molecule pushes onto the particle [39]. The procedure of adding white-noise sources, i.e., $\boldsymbol{F}^B$ in Eq. (1), will subsequently be called the Langevin approach (see Ref. [23]) while the resulting equation (1) is referred to as the Langevin equation [18]. As an alternative to the Langevin equation, stochastic processes can be described equivalently by a differential equation for the probability density $\mathcal{P}$, i.e., the Fokker-Planck equation, cf. Ref. [23], which will be widely employed below. The Fokker-Planck equation can be set up for Markov processes, which means that the probability density for a process at time t_n does not depend on earlier values than at time t_{n-1}, with $t_n - t_{n-1} = \triangle t$ and $n \in \mathbb{N}$ with $1 \leqslant n \leqslant q$ [23]. The Fokker-Planck equation enables us to calculate any averaged values of the regarded variable by integration [39,40].

For the following derivations, we limit ourselves to the translational components of Eq. (1) and, hence, the rotational degrees of freedom are not considered. Still, the influence of shear-induced rotation onto the translational motion is included due to $(\boldsymbol{R}_{\mathrm{FE}} : \boldsymbol{E}^\infty)_\alpha$ (the component of $\boldsymbol{R}_{\mathrm{FE}} : \boldsymbol{E}^\infty$ acting on the particle α) (see Ref. [34]).

B. Asymptotic expansion in the colored-noise regime

In the present subsection, we employ a time-scale asymptotics applied to the dimensionless form of Eq. (1).

Below, we define the three elementary time scales on which the essential physical phenomena will take place, cf. Refs. [7,18],

$$\tau_p = \frac{m}{6\pi\eta a}, \quad \text{the inertial relaxation time scale,} \tag{5}$$

$$\tau_f = \frac{1}{\dot{\gamma}}, \quad \text{the flow time scale, with the shear rate } \dot{\gamma}, \tag{6}$$

and

$$\tau_D = \frac{a^2}{D_0}, \quad \text{the diffusive time scale, with } D_0 = \frac{kT}{6\pi\eta a}, \tag{7}$$

TABLE I. Summary of Secs. II B and II C for Brownian particles with $\tau_p \ll \tau_D \ll \tau_f$.

	On τ_p		On τ_D	
t, $\triangle t$	$<\tau_p$	$>\tau_p$	$<\tau_D$	$>\tau_D$
Brownian velocity	Correlated	Uncorrelated		
Configuration			Constant	Changing
Diffusion	Velocity diffusive		Position short-time	Position long-time
$\langle yy\rangle$, $\langle zz\rangle$	$\sim t^2$		$\sim t$	$\sim t$
$\langle \triangle y \triangle y\rangle$, $\langle \triangle z \triangle z\rangle$	$\sim \triangle t^2$		$\sim \triangle t$	$\sim \triangle t$
$\langle U_i U_j\rangle$ with $i, j = x, y, z$	$\sim t$	Constant		

where m is the mass of one particle. D_0 is also known as the Stokes-Einstein diffusivity and captures the diffusivity of an isolated particle. Of the above-mentioned time scales it is possible to define dimensionless numbers in order to describe the significance of different phenomena. The ratio of τ_D to τ_f defines the Peclet number as follows:

$$\mathrm{Pe} = \frac{\tau_D}{\tau_f} = \frac{6\pi \eta a^3 \dot{\gamma}}{kT}. \tag{8}$$

As the Peclet number may also be interpreted as the ratio of shear forces to Brownian forces, the suspension is Brownian for a vanishing Peclet number while it refers to a non-Brownian suspension for a Peclet number tending to infinity [18].

The influence of particle inertia is determined by the Stokes number,

$$\mathrm{St} = \frac{\tau_p}{\tau_f}, \tag{9}$$

i.e. the ratio of the inertial relaxation time to the shear time scale. In the literature various authors investigated the influence of inertia in terms of finite Stokes numbers, e.g., Drossinos and Reeks [41] and Subramanian and Brady [11]. Presently, however, we limit ourselves to a τ_p, which is considerably smaller than τ_f, and, hence, particle inertia shall be neglected.

For the successional analysis, we may furthermore need to introduce three placeholder time scales as follows:

$$\tau_a, \quad \text{a placeholder for the regarded time scale,} \tag{10}$$

$$\tau_c, \quad \text{the time scale on which the particle configuration of the system changes, and} \tag{11}$$

$$\tau_{ac}, \quad \text{the time scale for the velocity autocorrelation.} \tag{12}$$

The system can be analyzed with respect to any of the given time scales, here τ_p, τ_f, and τ_D from Eqs. (5)–(7), by setting τ_a to the time scale of interest. Then the time t and the time-step size $\triangle t$ are taken in relation to τ_a. τ_c is defined such that during a time step $\triangle t$ on τ_c, the characteristic distance a particle has moved is a fraction of its own radius a. The determination of τ_c and τ_{ac} differs significantly from Brownian to non-Brownian particles.

For Brownian particles (see Refs. [21,23,24,27]) there is a so-called separation of time scales. This means, in the Brownian case, that the position of the particle changes on a time scale that differs from that of the velocity, so $\tau_c = \tau_D$ and $\tau_{ac} = \tau_p$ with $\tau_D \gg \tau_p$. Therefore it is constructive to define a time step $\triangle t$ in the range $\tau_p \ll \triangle t < \tau_D$ for the integration of the Langevin equation (1) such that the configuration (and thus $\boldsymbol{R}_{\mathrm{FU}}$, $\boldsymbol{R}_{\mathrm{FE}}$, and $\boldsymbol{U}^{\infty}$) and forces on the particle, e.g., $\boldsymbol{F}^H$ and $\boldsymbol{F}^P$ effectively stay unchanged over the time step while the random part of the velocity resulting from the Brownian motion is completely uncorrelated with the random part from the previous time step [21,24].

For non-Brownian particles (see Ref. [7]), the diffusive behavior results from the hydrodynamic part of the velocity. In contrast to the Brownian velocity with short autocorrelation times, the hydrodynamic velocity component directly depends on the configuration. Therefore, the time scale of the hydrodynamic velocity correlation is the same as the time scale of position change. In the non-Brownian case, $\tau_{ac} = \tau_f$ and $\tau_c = \tau_f$. This means, for $\triangle t \to 0$ on $\tau_a = \tau_f$, i.e., $\triangle t < \tau_f$, the present velocity is still correlated with velocities from previous time steps while the configuration is considered as unchanged over the time step. In contrast, for large $\triangle t > \tau_f$, the forces and configuration are not constant over the time step, and, in return, the velocities are uncorrelated. A summary of all the time scales and phenomena that occur on these time

TABLE II. Summary of Secs. II B, II C, and III for non-Brownian particles with $\tau_p \ll \tau_f \ll \tau_D$.

	On τ_p		On τ_f	
t, $\triangle t$	$<\tau_p$	$>\tau_p$	$<\tau_f$	$>\tau_f$
Velocity			Correlated	Uncorrelated
Configuration			Constant	Changing
Diffusion			Colored-noise velocity diffusive	Position long-time
$\langle yy\rangle$, $\langle zz\rangle$			$\sim t^2$	$\sim t$
$\langle \triangle y \triangle y\rangle$, $\langle \triangle z \triangle z\rangle$			$\sim \triangle t^2$	$\sim \triangle t$
$\langle V_i V_j\rangle$ with $i, j = x, y, z$			$\sim t$	Constant

scales can be found in Tables I and II. Note that $\langle xx\rangle$ and $\langle\triangle x\triangle x\rangle$ are not listed as this behavior differs from the y and z directions in the case of shear flow in the x direction. Note that these tables also contain information that will be referred to in later sections.

In addition to the different time scales, the system imbeds different length scales. On $\tau_a=\tau_c$, the appropriate length to nondimensionalize all lengths is the particle radius a, cf. Ref. [18].

In the case where the considered time scale is $\tau_a=\tau_p$, the characteristic length scale is the correlation length of a Brownian particle or the distance the particle moves during τ_p with the velocity $\sqrt{\frac{kT}{m}}$, see [40],

$$l\approx\sqrt{\frac{kT}{m}}\tau_p, \tag{13}$$

in the absence of any other forces which could introduce additional length scales.

From $\tau_p\ll\tau_D$ with τ_p from Eq. (5) and τ_D from Eq. (7) it follows $l\ll a$.

In the following, the nondimensionalized form of the Langevin equation (1) is analyzed. Depending on the physical situation, special forms of nondimensionalzing (1) were developed, cf. Refs. [11,18,21,22]. In this context, multiple time scale analysis has been used to receive reduced forms of the Fokker-Planck equation for Brownian particles [26,27]. However, we derive a new form of multiple time and length scale analysis which includes all time and length scales of interest for both Brownian and non-Brownian particles. This will be our starting equation, which has not been derived so far. This new compact form that we will expose for Brownian and non-Brownian particles shall consolidate the relation between the equations of motions and their corresponding Fokker-Planck equations as we will outline which variable is a Markov variable in the corresponding Fokker-Planck equation. The above-mentioned different time scales for Brownian and non-Brownian particles will become apparent below. Further, the analysis of the Brownian particle case which incorporates the white noise due to Brownian motion is done in order to show the special case of the Kramers equation (see, e.g., Ref. [23]) which poses a fundamental mean for our argumentation for the non-Brownian case.

For the nondimensionalization of Eq. (1), we employ the assumptions from [18] or [22], which imply that the hydrodynamic resistance matrices $\boldsymbol{R}_{\mathrm{FU}}$ and $\boldsymbol{R}_{\mathrm{FE}}$ are respectively nondimensionalized with the friction coefficient $f_r=6\pi\eta a$ and the product $f_r a$ whereby in our work we modify the nondimensionalization of $\boldsymbol{R}_{\mathrm{FE}}$ to $f_r h$ with h as the placeholder for the length scales a and l, depending on the respective time scale that is regarded. The asymptotics will show that the component referring to l is negligible; however, it is introduced to present the full analysis on both the time scales. The components of $\boldsymbol{E}^\infty$ are nondimensionalized by $\tau_f=1/\dot\gamma$. The mass m of one particle, is subsequently used for any nondimensionalization of quantities with mass dimension.

In the following, all components of the Langevin equation (1) with respect to all time scales are considered in the dimensionless equation. For that, first, all components are decomposed into their different parts in order to understand which processes are active on the respective time scales. Note that we only work with the translational components of Eq. (1). We will not use any identification mark or index to point that out. The $\boldsymbol{m}$ reduces to a simple m, since in the translational components $\boldsymbol{m}=m\cdot\boldsymbol{I}$, with $\boldsymbol{I}$ as a unity matrix of size $3N\times 3N$. In the analysis to follow, we need to introduce two expansion parameters,

$$\epsilon_1=\frac{\tau_p}{\tau_D}\quad\text{and}\quad\epsilon_2=\frac{\tau_p}{\tau_f}, \tag{14}$$

i.e., ϵ_1 as a small parameter for the asymptotic analysis of the Brownian case and ϵ_2 as a small parameter for the asymptotic analysis of the non-Brownian case.

For a better understanding of the time-scale expansion to follow, we consider the key time scales τ_p and τ_c for each term separately in an additive form, whereby Eq. (20) is received by inserting τ_p and τ_D into Eq. (4),

$$\tilde t_1=t/\tau_p,\ \ \tilde t_2=t/\tau_c, \tag{15}$$

$$\boldsymbol{x}=l\,\tilde{\boldsymbol{x}}_1(\tilde t_1)+a\,\tilde{\boldsymbol{x}}_2(\tilde t_2), \tag{16}$$

$$\boldsymbol{U}=\frac{d\boldsymbol{x}}{dt}=\frac{l}{\tau_p}\tilde{\boldsymbol{U}}_1(\tilde t_1)+\frac{a}{\tau_c}\tilde{\boldsymbol{U}}_2(\tilde t_2), \tag{17}$$

$$\boldsymbol{U}_\alpha^\infty=(\dot\gamma y_\alpha,0,0)=\left(\frac{1}{\tau_f}(l\ \tilde y_{1\alpha}(\tilde t_1)+a\ \tilde y_{2\alpha}(\tilde t_2)),0,0\right), \tag{18}$$

$$\boldsymbol{U}^\infty=\frac{1}{\tau_f}\big(l\ \tilde{\boldsymbol{U}}_1^\infty(\tilde{\boldsymbol{x}}_1)+a\ \tilde{\boldsymbol{U}}_2^\infty(\tilde{\boldsymbol{x}}_2)\big), \tag{19}$$

$$\boldsymbol{F}^B=\frac{m}{\tau_p}\frac{l}{\tau_p}\tilde{\boldsymbol{F}}_1^B(\tilde t_1)+\frac{m}{\tau_p}\frac{a}{\tau_D}\tilde{\boldsymbol{F}}_2^B(\tilde t_2). \tag{20}$$

The tilde represents the dimensionless components. The index α represents the α-th particle. In contrast, the indices 1 and 2 refer to the two components of the variables on the respective time scales. The components with index 1 create equations on the time scale $\tau_a=\tau_p$ and the components with index 2 create equations on $\tau_a=\tau_c$. Implementing (15)–(20) into Eq. (1) where $\boldsymbol{F}^H$ has been replaced by (2) and $\boldsymbol{F}^P$ is set to zero, we obtain:

$$m\left(\frac{l}{\tau_p\tau_p}\frac{d\tilde{\boldsymbol{U}}_1}{d\tilde t_1}+\frac{a}{\tau_c\tau_c}\frac{d\tilde{\boldsymbol{U}}_2}{d\tilde t_2}\right)=-f_r\tilde{\boldsymbol{R}}_{\mathrm{FU}}\left(\frac{l}{\tau_p}\tilde{\boldsymbol{U}}_1+\frac{a}{\tau_c}\tilde{\boldsymbol{U}}_2\right)+f_r\tilde{\boldsymbol{R}}_{\mathrm{FU}}\left(\frac{l}{\tau_f}\tilde{\boldsymbol{U}}_1^\infty+\frac{a}{\tau_f}\tilde{\boldsymbol{U}}_2^\infty\right)+f_r\frac{l}{\tau_f}(\tilde{\boldsymbol{R}}_{\mathrm{FE}}:\tilde{\boldsymbol{E}}^\infty)_1$$
$$+f_r\frac{a}{\tau_f}(\tilde{\boldsymbol{R}}_{\mathrm{FE}}:\tilde{\boldsymbol{E}}^\infty)_2+\frac{ml}{\tau_p\tau_p}\tilde{\boldsymbol{F}}_1^B+\frac{ma}{\tau_p\tau_D}\tilde{\boldsymbol{F}}_2^B. \tag{21}$$

1. Brownian particles

First, Eq. (21) is considered for Brownian particles. That implies that $\tau_c = \tau_D$ is inserted into Eq. (21). Furthermore, we multiply Eq. (21) by $\frac{\tau_p \tau_p}{ma}$. After applying Eq. (14), to get $\frac{\tau_p}{\tau_f} = \mathrm{Pe}\,\epsilon_1$ and $\frac{l}{a} = \sqrt{\frac{\tau_p}{\tau_D}} = \epsilon_1^{1/2}$, we receive

$$\left(\epsilon_1^{1/2}\frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} + \epsilon_1^2\frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2}\right) = -\epsilon_1^{1/2}\tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_1 - \epsilon_1\tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_2 + \mathrm{Pe}\,\epsilon_1^{3/2}\tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_1^\infty + \mathrm{Pe}\,\epsilon_1\tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_2^\infty + \mathrm{Pe}\,\epsilon_1^{3/2}(\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^\infty)_1 + \mathrm{Pe}\,\epsilon_1(\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^\infty)_2 + \epsilon_1^{1/2}\tilde{\boldsymbol{F}}_1^B + \epsilon_1\tilde{\boldsymbol{F}}_2^B. \tag{22}$$

Ordering (22) according to the small parameter $\epsilon_1 = \frac{\tau_p}{\tau_D}$ yields, to leading order, the asymptotics for the Brownian case in the separate equations (23)–(25) and (28)–(29),

$$\epsilon_1^{1/2}\ \text{terms:}\quad \frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} = -\tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_1 + \tilde{\boldsymbol{F}}_1^B, \tag{23}$$

where (23) is an equation of motion on $\tau_a = \tau_p$, so Δt is a fraction of τ_p here. As already indicated above, this is the time scale of velocity correlation for Brownian particles, i.e., $\tau_{ac} = \tau_p$. An analog equation in 1D can be found in Ref. [23]. There is derived the Fokker-Planck equation in correspondence to Eq. (23), namely Rayleigh‘s equation [23], which is a Fokker-Planck equation in velocity space, as the velocity is a Markov variable here, while the position is not. Of course, the appendant redimensionalized $\boldsymbol{R}_{\mathrm{FU}}$ has to be known (note that on the time scale τ_p, $\boldsymbol{R}_{\mathrm{FU}}$ is constant).

Further,

$$\epsilon_1\ \text{terms:}\quad 0 = -\tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_2 + \mathrm{Pe}\tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_2^\infty + \mathrm{Pe}(\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^\infty)_2 + \tilde{\boldsymbol{F}}_2^B, \tag{24}$$

$$\text{thus, for Pe} \to 0:\quad 0 = -\tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_2 + \tilde{\boldsymbol{F}}_2^B. \tag{25}$$

Equation (24) is an equation of motion on $\tau_a = \tau_D$, thus Δt is a fraction of τ_D. Isolating $\tilde{\boldsymbol{U}}_2$ in (24) by multiplying the inverse of $\tilde{\boldsymbol{R}}_{\mathrm{FU}}$ and using $\boldsymbol{U} = \frac{d\boldsymbol{x}}{dt}$ from (17) leads to a differential equation in position space. One more integration of (24) yields the change of the particle positions with Peclet number dependency; see the dimensional analysis in Refs. [18,21]. The position space differential equation in turn may be reformulated to an N-particle Fokker-Planck equation in position space as the positions of all particles are Markovian; see also Refs. [18,21].

In the following, we may shortly outline the effects of a position-dependent external force field $\boldsymbol{F}(\boldsymbol{x})$ added on the left-hand side of the Langevin equation (1), because in the non-Brownian case we will use a similar argumentation for the use of an alternative Fokker-Planck equation. Transferred to our terminology, such an additional external force $\boldsymbol{F}(\boldsymbol{x})$ could be nondimensionalized by the following:

$$\boldsymbol{F}(\boldsymbol{x}) = \frac{m}{\tau_k\tau_b}\,(l\ \tilde{\boldsymbol{F}}_1(\boldsymbol{x}/l) + a\ \tilde{\boldsymbol{F}}_2(\boldsymbol{x}/a)) = \frac{m}{\tau_k\tau_b}\,(l\ \tilde{\boldsymbol{F}}_1(\tilde{\boldsymbol{x}}_1) + a\ \tilde{\boldsymbol{F}}_2(\tilde{\boldsymbol{x}}_2)), \tag{26}$$

while the time scales τ_k and τ_b will subsequently be identified with some of the given time scales τ_p, τ_D, or τ_f, where we will distinguish two cases below. In the case that τ_k and τ_b do not equal τ_p, τ_D, or τ_f, additional time scales have to be introduced in the asymptotics, including the dimensionless analysis for $\boldsymbol{x}$ [Eq. (16)] and $\boldsymbol{U}$ [Eq. (17)]. For the explanations to follow we stick to the simpler cases that τ_k and τ_b equal either τ_p or τ_D with $\tau_k\tau_b = \tau_p\tau_D$ or $\tau_k\tau_b = \tau_p^2$, whereas the case that $\tau_k\tau_b = \tau_p^2$ prohibits the use of the Fokker-Planck equation in position space but necessitates another type of Fokker-Planck equation, namely the Kramers equation (see, e.g., Refs. [23,25,26,39]).

Wilemski [25] (among others like, e.g., Ref. [26]) gives a rule for the use of the Fokker-Planck equation in position,

$$\left|\frac{\tau_p^2}{m}\frac{\partial \boldsymbol{F}(\boldsymbol{x})}{\partial x}\right| \ll 1. \tag{27}$$

Inserting $\boldsymbol{F}(\boldsymbol{x})$ with $\tau_k\tau_b = \tau_p\tau_D$ into Eq. (27) does not pose any problems as the condition is fulfilled. The corresponding Fokker-Planck equation is an equation in position space, as for Eq. (24). For the second case that $\tau_k\tau_b = \tau_p^2$, Eq. (27) is not fulfilled. Adding the force $\boldsymbol{F}(\boldsymbol{x})$ from Eq. (26) with $\tau_k\tau_b = \tau_p^2$ to the right-hand side of Eq. (21), we see that these terms dominate on the τ_D time scale, i.e. in Eq. (24). The approach is as follows [23]: The regarded time scale $\tau_a = \tau_D$ has to be scaled down to $\tau_a = \tau_p$. The equation of interest for this case is the equation of motion on the smaller time scale τ_p, i.e., Eq. (23). The corresponding Fokker-Planck equation to (23) is a Fokker-Planck equation in velocity (Rayleigh‘s equation) which does not take into account the position dependency of $\boldsymbol{F}(\boldsymbol{x})$. So, in this case, the corresponding Fokker-Planck equation to the equation of motion on τ_p, i.e., (23), has to be a Fokker-Planck equation in position and velocity space, the so-called Kramers equation, where neither the velocity nor the position alone is Markovian but only a coupled variable in position-velocity space.

Finally,

$$\mathrm{Pe}\ \epsilon_1^{3/2}\ \text{terms:}\quad 0 = \tilde{\boldsymbol{R}}_{\mathrm{FU}}\tilde{\boldsymbol{U}}_1^\infty + (\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^\infty)_1, \tag{28}$$

$$\text{and } \epsilon_1^2\ \text{terms:}\quad \frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2} = 0. \tag{29}$$

Equations (23) and (28) are equations on τ_p. Equation (28) with $\epsilon_1^{3/2}$ components can be neglected when compared to Eq. (23) with the $\epsilon_1^{1/2}$ components. The same argumentation applies for Eqs. (24) and (29), which are both equations on τ_D. Therefore, Eq. (29) with ϵ_1^2 components can be neglected when compared to Eq. (24) with ϵ_1 components. Thus, in the following, Eqs. (28) and (29) are not considered.

2. Non-Brownian particles

For non-Brownian particles, $\tau_c = \tau_f$. Setting this into Eq. (21) above and multiplying by $\frac{\tau_p \tau_p}{ma}$ yields with $\frac{\tau_p}{\tau_D} = \frac{1}{\mathrm{Pe}} \frac{\tau_p}{\tau_f}$ and $\frac{l}{a} = \sqrt{\frac{1}{\mathrm{Pe}} \frac{\tau_p}{\tau_f}} = \sqrt{\frac{1}{\mathrm{Pe}}} \epsilon_2^{1/2}$ the following:

$$\left(\sqrt{\frac{1}{\mathrm{Pe}}}\, \epsilon_2^{1/2} \frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} + \epsilon_2^2 \frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2} \right) = -\sqrt{\frac{1}{\mathrm{Pe}}}\, \epsilon_2^{1/2} \tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_1 - \epsilon_2 \tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_2 + \sqrt{\frac{1}{\mathrm{Pe}}}\, \epsilon_2^{3/2} \tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_1^{\infty} + \epsilon_2 \tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_2^{\infty} + \sqrt{\frac{1}{\mathrm{Pe}}}\, \epsilon_2^{3/2} (\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^{\infty})_1 + \epsilon_2 (\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^{\infty})_2 + \sqrt{\frac{1}{\mathrm{Pe}}}\, \epsilon_2^{1/2} \tilde{\boldsymbol{F}}_1^B + \frac{1}{\mathrm{Pe}} \epsilon_2 \tilde{\boldsymbol{F}}_2^B. \tag{30}$$

Ordering according to small $\epsilon_2 = \frac{\tau_p}{\tau_f} \ll 1$ (which means a small Stokes number) yields the separate equations (31)–(35),

$$\sqrt{\frac{1}{\mathrm{Pe}}}\, \epsilon_2^{1/2} \text{ terms:} \quad \frac{d\tilde{\boldsymbol{U}}_1}{d\tilde{t}_1} = -\tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_1 + \tilde{\boldsymbol{F}}_1^B. \tag{31}$$

Equation (31) is the equation of motion on τ_p, where the hydrodynamic shear forces have no influence since they are of order $O(\epsilon_2^{3/2})$ in Eq. (34). Pe tends to ∞, thus (31) and (34) do not have the same significance as (23) and (28). As mentioned above, in the non-Brownian case, $\tau_{ac} = \tau_f$, so the time scale τ_p is not in the scope of interest for the shear-induced diffusion.

Further,

$$\epsilon_2 \text{ terms:} \quad 0 = -\tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_2 + \tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_2^{\infty} + (\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^{\infty})_2 + \frac{1}{\mathrm{Pe}} \tilde{\boldsymbol{F}}_2^B, \tag{32}$$

$$\text{thus, for } \mathrm{Pe} \to \infty: \quad 0 = -\tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_2 + \tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_2^{\infty} + (\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^{\infty})_2. \tag{33}$$

Equations (32) and (33) give the change of position on τ_f. It is important to note that the present approach also covers existing special approaches, as, for example, Eq. (33) can also be found as part of a Stokes number expansion in $\boldsymbol{U}$ in Ref. [11].

Finally,

$$\sqrt{\frac{1}{\mathrm{Pe}}}\, \epsilon_2^{3/2} \text{ terms:} \quad 0 = \tilde{\boldsymbol{R}}_{\mathrm{FU}} \tilde{\boldsymbol{U}}_1^{\infty} + (\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^{\infty})_1, \tag{34}$$

$$\text{and } \epsilon_2^2 \text{ terms:} \quad \frac{d\tilde{\boldsymbol{U}}_2}{d\tilde{t}_2} = 0. \tag{35}$$

For the description on τ_p, Eq. (34) with $\epsilon_2^{3/2}$ components can be neglected when compared to Eq. (31) with $\epsilon_2^{1/2}$ components. Also, the ϵ_2^2 components of Eq. (35) can be neglected in comparison to the ϵ_2 components of Eq. (32).

As we are interested in investigating the Fokker-Planck equation for non-Brownian particles for the case of shear-induced diffusion which occurs on τ_f, we analyze Eq. (33) in the following. After returning to the dimensional representation and rearranging terms in Eq. (33), it follows for a single particle α that:

$$0 = -f_r(\boldsymbol{U}_\alpha - \boldsymbol{U}_\alpha^{\infty}) + \boldsymbol{F}_\alpha^{\mathrm{op}} \tag{36}$$

$$\text{with} \quad \boldsymbol{F}_\alpha^{\mathrm{op}} = -(\boldsymbol{R}_{\mathrm{FU}}^{*}(\boldsymbol{U} - \boldsymbol{U}^{\infty}))_\alpha + (\boldsymbol{R}_{\mathrm{FE}} : \boldsymbol{E}^{\infty})_\alpha, \tag{37}$$

where $*$ indicates the hydrodynamic influence of all the other particles on a particle α resulting from $-\boldsymbol{R}_{\mathrm{FU}}(\boldsymbol{U} - \boldsymbol{U}^{\infty})$ in Eq. (33). The α components are still bold, as they denote a vector with the three components for the x, y, and z directions. Note that the diagonal components of $\boldsymbol{R}_{\mathrm{FU}}$ include both the friction f_r of an isolated particle and the hydrodynamic influence of the other particles [34]; therefore, a decomposition according to (36) is possible with nonzero diagonal elements of $\boldsymbol{R}_{\mathrm{FU}}^{*}$.

In Ref. [1], an analog to Eq. (36) is presented for the equation of motion of a non-Brownian particle with the hydrodynamic influence of the other particles expressed in a colored-noise force. We declare $\boldsymbol{F}_\alpha^{\mathrm{op}}$ also as a colored-noise force. In contrast to the Langevin equations, cf. Eq. (1), equations of motion with a colored-noise force, as (36), are called Langevin-*like* equations [23].

For our starting point, we rewrite Eq. (36) according to the following:

$$\boldsymbol{U}_\alpha = \boldsymbol{U}_\alpha^{\infty} + \boldsymbol{V}_\alpha \quad \text{with} \quad \boldsymbol{V}_\alpha = \frac{1}{f_r} \boldsymbol{F}_\alpha^{\mathrm{op}}. \tag{38}$$

We model $\frac{1}{f_r} \boldsymbol{F}_\alpha^{\mathrm{op}}$ as a colored-noise velocity $\boldsymbol{V}_\alpha$. In contrast to Eq. (1) which actually is an N-particle system of coupled equations (38) belongs to a system of $3N$ independent equations where the velocity $\boldsymbol{V}$ has to be modeled separately. The model for the colored-noise velocity will be presented in Sec. III.

It will be pointed out below that the derivation of a Fokker-Planck equation strongly depends on the appropriate choice of the regarded time scale τ_a and the time step size $\triangle t$, which is an important issue of the present work. Most importantly, we derive a possible alternative approach for the Fokker-Planck equation based on (38).

C. Shear-induced diffusion in comparison to Brownian diffusion

The basic approach may be taken from Refs. [23] and [42]. The Fokker-Planck equation is a parabolic differential equation for the probability density $\mathcal{P}(\boldsymbol{s},t)$, see Ref. [23],

$$\frac{\partial \mathcal{P}(\boldsymbol{s},t)}{\partial t} = -\sum_{i=1}^{r} \frac{\partial}{\partial s_i} A_i(\boldsymbol{s}) \mathcal{P} + \frac{1}{2} \sum_{i,j=1}^{r} \frac{\partial^2}{\partial s_i \partial s_j} \mathcal{D}_{ij}(\boldsymbol{s}) \mathcal{P}, \tag{39}$$

and can be derived by the differential quotient $\triangle \mathcal{P} / \triangle t$, with $\triangle \mathcal{P}(\boldsymbol{s},t) = \mathcal{P}(\boldsymbol{s},t + \triangle t) - \mathcal{P}(\boldsymbol{s},t)$ in the limit $\triangle t \to 0$. $\boldsymbol{s}$ is the set of variables of interest s_i with dimension r, here, e.g., position or velocity.

The drift A_i and diffusion terms $\mathcal{D}_{ij}$ of the Fokker-Planck equation can be found by evaluating $\boldsymbol{s}$ in the Langevin equation (1) during the characteristic time step $\triangle t$ on the regarded time scale τ_a,

$$A_i(\boldsymbol{s}) = \frac{\langle \triangle s_i \rangle_s}{\triangle t}, \tag{40}$$

$$\mathcal{D}_{ij}(\boldsymbol{s}) = \frac{\langle \triangle s_i \triangle s_j \rangle_s}{\triangle t}, \tag{41}$$

for $\triangle t \to 0$, whereas $\triangle s_i$ is the change of the variable of interest during $\triangle t$, i.e., $\triangle s_i = s_i(t + \triangle t) - s_i(t)$. The $\langle\rangle_s$ represents a conditional average with a known constant reference value, whereby in our paper an index $\boldsymbol{s}$ denotes the reference to the current $\boldsymbol{s}(t)$. The time step $\triangle t$ for integrating the equations of motion, e.g., (1) on the time scale τ_a, has to be small so $\boldsymbol{s}$ does not change much during $\triangle t$. Due to the smallness of $\triangle t$, all terms with O($\triangle t$) in the calculation of (40) and (41) can be neglected. In the following, we consider linear Fokker-Planck equations which, following the definition from Ref. [23], limits us to drift coefficients which are linear functions of $\boldsymbol{s}$ and constant diffusion terms $\mathcal{D}_{ij}$. Further, $\boldsymbol{s}$ is supposed to be a Markov variable. Then the change of $\boldsymbol{s}$ during $\triangle t$ serves to find $\triangle\mathcal{P}/\triangle t$, or $\partial\mathcal{P}/\partial t$ in the limit $\triangle t \to 0$.

1. Brownian particles

At first we will briefly discuss details for Brownian particles because their diffusive behavior shows analogies to the non-Brownian case. For times $\tau_p \ll t < \tau_D$ there is a so -called short-time self-diffusivity and for times $\tau_D < t$ there is a so-called long-time self-diffusivity [22,43]. For arbitrary times t, both in the short-time diffusive regime and in the long-time diffusive regime, use of Eq. (41) requires time steps $\tau_p \ll \triangle t < \tau_D$ to capture the short-time diffusivity of position. Since the configuration is unchanged during this time-step size (and thus forces depending on the configuration), Brownian particles with hydrodynamic interactions can be described by a Fokker-Planck equation in position space, cf. Refs. [21,24]. Here, the N-particle diffusion tensor is $\boldsymbol{D} = kT\boldsymbol{R}_{\mathrm{FU}}^{-1}$, cf. Ref. [18].

For time steps $\tau_D < \triangle t$ to capture the long-time diffusion, the configuration is not constant during $\triangle t$ and thus forces depending on the configuration are not constant either, and $O(\triangle t)$ terms cannot be neglected. Hence, in the present context, time steps $\tau_D < \triangle t$ are not allowed for the derivation of a corresponding Fokker-Planck equation in position space [see also Sec. III, Eq. (60)], cf. Ref. [21].

On $\tau_a = \tau_p$ with $\triangle t < \tau_p$ or $t < \tau_p$, Brownian particle velocities are correlated while the mean-square displacements ($\langle\triangle x^2\rangle$, $\langle\triangle y^2\rangle$, $\langle\triangle z^2\rangle$, and $\langle xx\rangle$, $\langle yy\rangle$, $\langle zz\rangle$) show quadratic behavior in $\triangle t$, respectively, in time t [23,34,40]. Hence, the position is not diffusive on $\tau_a = \tau_p$ nor even a Markovian variable, since the velocity from the last time step is not fully relaxed and thus needed to update the particle‘s position [23].

In contrast, on τ_p, the velocity variable for one particle α exhibits diffusive behavior with a linear in time mean-square velocity $\langle U_i(t)U_j(t)\rangle$ for $i,j = x,y,z$ [39]. In the previous Sec. II B, the underlying equation of motion for $\tau_a = \tau_p$ is given in Eq. (23) whereby the corresponding Fokker-Planck equation is built up in velocity space, named Rayleigh‘s equation, cf. Ref. [23]. It can be shown that on $\tau_a = \tau_p$ for times $t \to \infty$, the mean-square velocity $\langle U_i(t \to \infty)U_j(t \to \infty)\rangle$ is no longer linear in time, but a constant, i.e. the equilibrium value [23], cf. Appendix A, Eq. (A3) with $t \to \infty$.

2. Non-Brownian particles

For non-Brownian particles under shear flow in x direction, Sierou and Brady show simulation results with a linear behavior in time for times $t \to \infty$, i.e., $t > \tau_f$, for the mean-square displacements in the y and z directions, with initial positions of the particles in the origin [7],

$$\langle yy\rangle \sim 2D_{yy}t, \tag{42}$$

$$\langle zz\rangle \sim 2D_{zz}t, \tag{43}$$

with the angle brackets here and in the rest of the paper as an average over all particles in the system. As the D_{ij} coefficients in the non-Brownian context include averages over all particles they do not present the positions of all particles in the system anymore. They are not comparable to the Brownian N-particle diffusion tensor $\boldsymbol{D} = kT\boldsymbol{R}_{\mathrm{FU}}^{-1}$ mentioned above. The exact time t for shear-induced diffusion to arise also depends on the particle volume fraction and may not be exactly $t = \tau_f$, see, e.g., Ref. [7]. For short times, $t < \tau_f$, Sierou and Brady illustrate quadratic behavior in time. In contrast to Brownian diffusion, the shear-induced diffusion of non-Brownian particles exhibits a difficulty as it appears only for $t \to \infty$ on $\tau_a = \tau_f$, see, e.g., Refs. [1,7,15,28]. This corresponds to the long-time diffusivity in the Brownian case. The authors in Ref. [7] explain that the beginning of the linear behavior at times $t > \tau_f$ marks the beginning of the diffusive behavior of position with a constant diffusion coefficient after enough particle-particle interactions have taken place and the velocity is not correlated anymore. So at arbitrary times t (including times $t > \tau_f$), time steps of $\triangle t > \tau_f$ capture the diffusive behavior in Eq. (41) while time steps $\triangle t < \tau_f$ do not. A summary on the time scales can be taken from Table II in Sec. II B.

In contrast to Eqs. (42) and (43) for the diffusive component in the y and z directions, the diffusion components in the x direction, i.e., D_{xx} and D_{xy}, are more complicated because of the shear flow in the x direction (Ref. [7] which refers to Ref. [44]),

$$\langle xx\rangle = 2D_{xx}t + 2D_{xy}\dot{\gamma}t^2 + \tfrac{2}{3}D_{yy}\dot{\gamma}^2t^3, \tag{44}$$

$$\langle xy\rangle = 2D_{xy}t + D_{yy}\dot{\gamma}t^2, \tag{45}$$

where $D_{xy} = D_{yx}$ is the only off-diagonal component [5,7]. Extracting D_{xx} and D_{xy} via the mean-square displacements in Eqs. (44) and (45) in analogy to gaining D_{yy} and D_{zz} from (42) and (43) causes a difficulty, as the authors in Ref. [7] point out. Namely, for very large times t in the non-Brownian case, it is difficult to extract D_{xx} and D_{xy} from Eqs. (44) and (45) because the terms of order $O(t^2)$ and $O(t^3)$ dominate. So, instead, they derive D_{xx} and D_{xy} by calculating $\langle xx\rangle$ and $\langle xy\rangle$ from $\int_0^t \boldsymbol{U}_\alpha(t')dt' = \int_0^t \boldsymbol{U}_\alpha^\infty(t') + \boldsymbol{U}_\alpha^h(t')dt' = \boldsymbol{x}^\infty + \boldsymbol{x}^h$ with $\boldsymbol{x}^h$ as the hydrodynamic displacement due to the hydrodynamic velocity component $\boldsymbol{U}^h$, resulting from a time derivative of Eqs. (44) and (45), which also can be found in general form in Ref. [23], as follows:

$$\partial_t \langle xx \rangle - 2\langle x\,(\dot{\gamma} y)\rangle = 2D_{xx}, \tag{46}$$

$$\partial_t \langle xy \rangle - \langle y\,(\dot{\gamma} y)\rangle = 2D_{xy}. \tag{47}$$

Finally, Sierou and Brady [7] receive diffusion coefficients with a coupling term,

$$D_{xx}(t) = \frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle + \left\langle \frac{dx^h}{dt}\int_0^t \dot{\gamma} y(t')dt'\right\rangle, \tag{48}$$

$$D_{xy}(t) = \frac{1}{2}\frac{d}{dt}\langle x^h y\rangle + \frac{1}{2}\left\langle \frac{dy}{dt}\int_0^t \dot{\gamma} y(t')dt'\right\rangle. \tag{49}$$

They justify the dependence on t in order to use the diffusion tensors also for short times t where the diffusive behavior has not started yet and claim that both the diffusion tensors become constant for large times t.

Still, we would like to point out that at arbitrary times t, even in the long-time limit $t \to \infty$, i.e., $t > \tau_f$, the behavior of position is nondiffusive for small time-step sizes $\triangle t < \tau_f$ on the time scale $\tau_a = \tau_f$. This implies that the description of the diffusive regime necessitates time-step sizes $\triangle t > \tau_f$. On the other hand, large time-step sizes $\triangle t > \tau_f$ cause a problem as we will point out in the next Sec. III. Our argumentation will be similar to problems that arise in the Brownian case either when an external force field $\boldsymbol{F}(\boldsymbol{x})$ changes too fast (see Sec. II B, the Kramers equation) or when $\triangle t > \tau_D$. So we do not intend to increase the time-step size to $\triangle t > \tau_f$ until the position is diffusive. In contrast, we model the colored-noise velocity $\boldsymbol{V}_\alpha$ in Eq. (38) such that it is diffusive for $\triangle t < \tau_f$ on $\tau_a = \tau_f$. The mean square of $\boldsymbol{V}_\alpha$ will be linear in time for times $t < \tau_f$. For times $t \to \infty$, $\langle \boldsymbol{V}^2\rangle$ is a constant, in analogy to the Brownian velocity $\langle (U_i(t \to \infty))^2\rangle$ as mentioned above. A summary on the time scales can be found in Tables I and II in Sec. II B. We will need diffusion coefficients for the colored-noise velocities which we will derive from D_{xx}, D_{yy}, D_{zz}, and D_{xy}.

III. ALTERNATIVE APPROACH FOR THE FOKKER-PLANCK EQUATION BASED ON THE COLORED-NOISE ASSUMPTION

In the present work, Eq. (38) is the basis for the derivation of the Fokker-Planck equation of the shear-induced self-diffusion process, whereby from now on we explicitly write the dependence on time t to account for the correlation of varying times. The hydrodynamic velocity $V_i(t)$ results from the hydrodynamic interaction on this particle due to all other particles. Here, $V_i(t)$, exhibits a colored-noise property as follows:

$$U_x(t) = U_x^\infty(t) + V_x(t), \tag{50}$$

$$U_y(t) = V_y(t), \tag{51}$$

$$U_z(t) = V_z(t), \tag{52}$$

for time $t > \tau_f$ in order to map the diffusive regime. Note that this is the equation for one particle α in the full system. The whole system for all N particles consists of $3N$ equations. These $3N$ equations all are independent since the influence of the particles onto each other is going to be modeled separately in $\boldsymbol{V}$. Thus it makes no difference if the averaging procedures are for a single particle over different configurations, i.e., ensemble averages, or if averaging procedures are performed over all particles in the system. We will use the latter case for the purpose of manageability and we will skip the α index in the following. The important question is to find the corresponding Fokker-Planck equation. Hence, which time scale shall be taken as the underlying time scale τ_a and which variable is a Markov variable on this time scale must be determined.

We know from above that in the non-Brownian case, on $\tau_a = \tau_f$, for $\triangle t > \tau_f$ the position is diffusive and as the velocity changes with the configuration, the velocity is uncorrelated for this time-step size. However, the configuration will only be constant for $\triangle t \to 0$, respectively, $\triangle t < \tau_f$. So whether $\triangle t$ should be larger or smaller than τ_f must be determined. At first, we have to develop a model for V_i. In a second step, the drift and diffusion terms in position for both $\triangle t < \tau_f$ and $\triangle t > \tau_f$ will be considered separately to show that time-step sizes $\triangle t < \tau_f$ are necessary.

Presently, we assume that the colored-noise velocity $V_i(t)$ itself is a Markov process and can be mimicked by an Ornstein-Uhlenbeck process (cf. Refs. [23,45]) represented by the following Langevin equation:

$$\frac{dV_i(t)}{dt} = -\frac{1}{\tau_{\text{corr}}}V_i(t) + L_i(t), \tag{53}$$

for $i = x,y,z$ with white noise $L_i(t)$. The denotation as a colored-noise velocity also has its origin in the fact that this velocity component is correlated on the same time scale as the configuration changes. This is in stark contrast to the velocity in the Brownian case, which is correlated on τ_p and thus on a much smaller time scale than the change of position. Due to the Markov property, the value of $V_i(t)$ depends on the value from the previous time step but not from earlier time steps. $\boldsymbol{V}$ is defined on τ_f and not on τ_p as it results from the equation of motion (36) for non-Brownian particles on τ_f. τ_{corr} is the correlation time of the colored-noise velocity. According to Ref. [7] the correlation time of the velocity in the non-Brownian case is τ_f. Since in our colored-noise velocity model (53) we have to account for these long correlation times, τ_{corr} is set to τ_f. The white noise $L_i(t)$ introduces a randomness which results in the diffusive behavior in the position space for times $t > \tau_f$, $t \to \infty$.

For an Ornstein-Uhlenbeck process the following results apply in the limit $t \to \infty$ [23,39,45]:

$$\langle L_i(t+t')L_j(t)\rangle = B_{ij}\delta(t'), \tag{54}$$

$$\langle V_i(t+t')V_j(t)\rangle = \frac{B_{ij}\tau_f}{2}\exp\left(\frac{-|t'|}{\tau_f}\right), \tag{55}$$

$$\langle V_i(t)\rangle = 0, \tag{56}$$

where $\boldsymbol{V}$ fulfills these equations for $t \to \infty$, which is the equilibrium state. The large t guarantees that there is no dependency on the initial value $V_i(0)$, cf. Appendix A, Eq. (A3). Further, for our case the large time t guarantees that it is in the regime where shear-induced diffusion has already started. We will show how to gain the B_{ij} coefficients in Sec. III B. Later, the B_{ij} coefficients will appear in our new colored-noise Fokker-Planck equation. Equation (55) accounts for the long correlation times which result in colored noise in contrast to the white noise from the Brownian force $\boldsymbol{F}^B$, cf. Eq. (1).

A. Drift and diffusion terms in position space

In the following, the drift and diffusion terms in position space are analyzed with respect to the time-step size Δt. We will show that a time step $\Delta t < \tau_f$ is necessary. Note that the present derivation for the drift and diffusion terms will be done for the x direction only as this is also the direction of shear flow. The y and z directions work analogously. First, we regard the drift term which consists of a term due to the shear flow Δx^∞ and a term due to the hydrodynamic interactions Δx^h. Here we show the full drift coefficient for this work (we use the rules for these coefficients from, e.g., Refs. [39] and [23]). From Eqs. (50) and (53) we have:

$$\langle \Delta x\rangle = \langle \Delta x^\infty\rangle + \langle \Delta x^h\rangle, \tag{57}$$

$$\text{with}\quad \langle \Delta x^\infty\rangle = \left\langle \int_{t_n}^{t_{n+1}} y(t)\dot{\gamma}\,dt\right\rangle,\ \langle \Delta x^h\rangle = \int_{t_n}^{t_{n+1}} \langle V_x(t)\rangle dt, \tag{58}$$

$$\text{and}\quad \langle V_x(t_{n+1})\rangle = \langle V_x(t_n)\rangle \exp\left(-\frac{\Delta t}{\tau_f}\right). \tag{59}$$

Note that the order of the averaging and the integration can be interchanged, cf. Ref. [42]. Now we regard the first and second parts of Eq. (57) separately for $\Delta t > \tau_f$ and $\Delta t < \tau_f$.

In the first part of Eq. (57), y is expanded in a Taylor series around t_n, cf. the derivation of the Fokker-Planck equation in Refs. [23,42],

$$\begin{aligned}\langle \Delta x^\infty\rangle &= \left\langle \int_{t_n}^{t_{n+1}} y(t_n)\dot{\gamma}\,dt\right\rangle + \left\langle \int_{t_n}^{t_{n+1}} (t-t_n)\frac{dy(t_n)}{dt}\dot{\gamma}\,dt\right\rangle \\ &\quad + \left\langle \int_{t_n}^{t_{n+1}} O((t-t_n)^2)dt\right\rangle \\ &= \langle y(t_n)\rangle\dot{\gamma}\Delta t + \left[\frac{(t-t_n)^2}{2}\left\langle\frac{dy(t_n)}{dt}\right\rangle\dot{\gamma}\right]_{t_n}^{t_{n+1}} + O(\Delta t^3)\end{aligned}$$

$$\frac{\langle \Delta x^\infty\rangle_{\boldsymbol{x},\boldsymbol{V}}}{\Delta t} = \dot{\gamma}\left(y(t_n) + \frac{\Delta t}{2}\frac{dy(t_n)}{dt}\right) + O(\Delta t^2). \tag{60}$$

Note that in Eq. (60) we added the $\boldsymbol{x},\boldsymbol{V}$ index to account for the conditional average from Eq. (41) with constant $\boldsymbol{x},\boldsymbol{V}$ at time t_n. As a result, the $\langle\rangle$ brackets for the constant values at t_n can be removed. In the following, the dimensionless form is regarded. Since we are only interested in the question if Δt should be smaller or larger than τ_f, τ_a is set to τ_f and thus only the components on τ_f are regarded (and not possible components on τ_p) as follows:

$$\frac{a}{\tau_f}\frac{\langle \Delta \tilde{x}^\infty\rangle_{\boldsymbol{x},\boldsymbol{V}}}{\Delta \tilde{t}} = \frac{1}{\tau_f}\left(a\ \tilde{y}(t_n) + \frac{\tau_f \Delta\tilde{t}}{2}\frac{a}{\tau_f}\frac{d\tilde{y}(t_n)}{d\tilde{t}} + \cdots\right) \tag{61}$$

with $\Delta t = \tau_f\,\Delta\tilde{t}$, $\frac{dy}{dt} = \frac{a}{\tau_f}\frac{d\tilde{y}}{d\tilde{t}}$ [see Eq. (17)] and $\dot{\gamma} = \frac{1}{\tau_f}$. Note, that $\frac{dy(t_n)}{dt} = V_y(t_n)$, see Eq. (51). For $\Delta t \to 0$, i.e., $\Delta t < \tau_f$, terms of order $O(\Delta\tilde{t})$ can be neglected since $\Delta\tilde{t} \to 0$. For $\Delta t > \tau_f$, the higher-order terms cannot be neglected since $\Delta\tilde{t} > 1$.

This means that the update of the position cannot be realized with only the position of the last time step. $\boldsymbol{U}^\infty$, depending also on the position, cannot be taken as constant over the time step. For an alternative argumentation concerning the problem of too-large time-step sizes in analogy to the Kramers equation, see Appendix D. We come to the conclusion that, for time steps $\Delta t > \tau_f$, a Fokker-Planck equation in the position is not valid here as the drift term $\boldsymbol{U}^\infty$ (corresponding to an inhomogeneous force field) changes too fast for this time step, cf. (the Kramers equation and the condition for constant forces over the time step).

We also analyze the $\langle \Delta x^h\rangle$ part of Eq. (57), where for $\Delta t < \tau_f$, $\Delta t \to 0$, cf. Eq. (59), we have the following:

$$\langle V_x(t_{n+1})\rangle_{\boldsymbol{V}} = \langle V_x(t_n)\rangle_{\boldsymbol{V}} + O(\Delta t), \tag{62}$$

$$\langle \Delta x^h\rangle = \int_{t_n}^{t_{n+1}} \langle V_x(t)\rangle dt = \langle V_x(t_n)\rangle\Delta t + O(\Delta t^2), \tag{63}$$

$$\frac{\langle \Delta x^h\rangle_{\boldsymbol{x},\boldsymbol{V}}}{\Delta t} = V_x(t_n) + O(\Delta t), \tag{64}$$

where terms of order $O(\Delta t)$ can be neglected. The $O(\Delta t)$ terms in (62) come from the series representation of the exponential function in (59).

For the purpose of completeness, we also analyze $\langle \Delta x^h\rangle$ for $\Delta t > \tau_f$. We see from Eq. (59) that $\langle V_x(t_{n+1})\rangle_{\boldsymbol{V}} \approx 0$ for $\Delta t \to \infty$. The time-step size is longer than the correlation length of the colored-noise velocity and thus, using Eq. (59),

$$\langle \Delta x^h\rangle = \int_{t_n}^{t_{n+1}} \langle V_x(t)\rangle dt \tag{65}$$

$$= \int_{t_n}^{t_{n+1}} \langle V_x(t_n)\rangle \exp\left[-\frac{(t-t_n)}{\tau_f}\right]dt, \tag{66}$$

$$\langle \Delta x^h\rangle_{\boldsymbol{x},\boldsymbol{V}} = \langle V_x(t_n)\rangle_{\boldsymbol{V}}\tau_f = V_x(t_n)\tau_f, \tag{67}$$

and thus

$$\frac{\langle \Delta x^h\rangle_{\boldsymbol{x},\boldsymbol{V}}}{\Delta t} \approx 0. \tag{68}$$

Due to the problems arising for large Δt in the context of $\boldsymbol{U}^\infty$, in the present work, the time step Δt is considered to be much smaller than the time scale of configurational changes, τ_f. For this case, the bulk velocity is considered to be constant over the time step and the drift term for the position in our alternative Fokker-Planck equation later is achieved by combining Eqs. (60) and (64) into $\frac{\langle \Delta x\rangle_{\boldsymbol{x},\boldsymbol{V}}}{\Delta t} = \dot{\gamma}\,y(t_n) + V_x(t_n)$. Hence, the correlation time τ_f of V_i is larger than the time step employed in the present work.

This colored-noise property destroys the Markov property of the position (for a mathematical reason, see, e.g., Ref. [23]). We conclude that also for the small time-step size it is not possible to derive a consistent Fokker-Planck equation in position space.

The same analysis can be conducted for the diffusion terms. Again the rules for the diffusion terms for Ornstein-Uhlenbeck processes and Fokker-Planck equations can be found, e.g., in Refs. [34,39], or [23]. The diffusion coefficients can be achieved by using the following equation (already mentioned in Sec. II C):

$$\langle \triangle x \triangle x \rangle = \left\langle \left(\int_{t_n}^{t_{n+1}} y(t)\dot{\gamma} dt \right)^2 \right\rangle + 2 \left\langle \int_{t_n}^{t_{n+1}} \int_{t_n}^{t_{n+1}} U_x^{\infty}(t') V_x(t'') dt' dt'' \right\rangle + \left\langle \int_{t_n}^{t_{n+1}} \int_{t_n}^{t_{n+1}} V_x(t') V_x(t'') dt' dt'' \right\rangle. \quad (69)$$

For $\triangle t \to 0$, i.e., $\triangle t < \tau_f$,

$$\langle \triangle x \triangle x \rangle_{\mathbf{x},\mathbf{V}} = \langle U_x^{\infty}(t_n)^2 \rangle_{\mathbf{x},\mathbf{V}} \triangle t^2 + 2 \langle U_x^{\infty}(t_n) V_x(t_n) \rangle_{\mathbf{x},\mathbf{V}} \triangle t^2 + \langle V_x^2(t_n) \rangle_{\mathbf{V}} \triangle t^2 = 0, \quad (70)$$

with $\langle V_x^2(t_n) \rangle_{\mathbf{V}} = V_x^2(t_n)$; see also Appendix B. We see here the quadratic component that arises for small times (compare simulation results from Sierou and Brady [7], mentioned above).

The calculation of the first and second parts of Eq. (69) poses further difficulties. But, at least for the diffusivity of the position in the y and z directions, where there are no such terms as the first and second parts of Eq. (69), we see that the time-step size should be larger than τ_f in order to reach the linear regime. For $\triangle t \to \infty$, i.e., $\triangle t > \tau_f$, in Appendix B in Eq. (B1) is shown that $\langle \triangle x^h \triangle x^h \rangle_{\mathbf{x},\mathbf{V}} = V_x^2(t_n)\tau_f^2 + B_{xx}\tau_f^2 \triangle t - \frac{3}{2} B_{xx}\tau_f^3$, thus linear behavior in $\triangle t$.

B. Colored-noise Fokker-Planck equation

Due to this conflict, the traditional Fokker-Planck equation in position space has to be extended. On the time scale $\tau_a = \tau_f$ with $\triangle t < \tau_f$, the colored-noise velocity is a diffusive process. This is analog to the Brownian case, where for $\tau_a = \tau_p$ not the position but the velocity is assumed to be diffusive. In the present work, it is not the whole velocity but only V_i,

$$\text{for } \triangle t < \tau_f \text{ and } \triangle t \to 0: \quad \langle \triangle V_i \triangle V_j \rangle_{\mathbf{V}} = B_{ij} \triangle t, \quad (71)$$

$$\text{for } \triangle t > \tau_f \text{ and } \triangle t \to \infty: \quad \langle \triangle V_i \triangle V_j \rangle_{\mathbf{V}} = \frac{B_{ij}\tau_f}{2} + V_i(t_n)V_j(t_n) = \text{const.} \quad (72)$$

For a proof see Appendix A and for an overview see Table II in Sec. II B.

In order to avoid the violation of the Markov property of the position variables, we use a colored-noise approach for the Fokker-Planck equation. Van Kampen [23] gives a general mathematical description for colored-noise problems in 1D under the assumption of a composite Markov process which means that the behavior of the one variable space can be decomposed from the other variable space and thus is added separately in the Fokker-Planck equation which yields a modified form of Eq. (39). We extend this approach to the 3D shear-induced diffusion problem. This means that by using Eq. (39) there are no diffusion coefficients in position space [as for small time-step sizes, the mean-square displacements are of order $O(\triangle t^2)$, cf. Eq. (70)] and due to the composite Markov process assumption, no coupled diffusion coefficients of position and the colored-noise velocity. The diffusion coefficients for V_i according to Eq. (41) are obtained from (71) as $\frac{\langle \triangle V_i \triangle V_j \rangle_{\mathbf{V}}}{\triangle t} = B_{ij}$. Thus, we only need the coefficients B_{ij}.

Integrating Eq. (55) for $t \to \infty$ yields for B_{ij} the following:

$$B_{ij} = \int_0^{\infty} \lim_{t\to\infty} \langle V_i(t+t')V_j(t) \rangle dt' \frac{2}{\tau_f} \left[\int_0^{\infty} \exp\left(\frac{-t'}{\tau_f} \right) dt' \right]^{-1} \quad (73)$$

$$= \int_0^{\infty} \lim_{t\to\infty} \langle V_i(t+t')V_j(t) \rangle dt' \; \frac{2}{\tau_f^2}. \quad (74)$$

The determination of B_{ij} requires knowledge of $\int_0^{\infty} \lim_{t\to\infty} \langle V_i(t+t')V_j(t) \rangle dt'$.

The B_{ij} coefficients are not the same as the diffusion coefficients D_{ij}. B_{yy} is connected to D_{yy} using $\langle U_y(t+t')U_y(t) \rangle = \langle V_y(t+t')V_y(t) \rangle$ with Eq. (51).

From the known rule $D_{yy} = \int_0^{\infty} \lim_{t\to\infty} \langle U_y(t+t')U_y(t) \rangle dt'$ (cf. Refs. [7,46] or with a colored-noise force in Ref. [1]) we find that

$$D_{yy} = \int_0^{\infty} \lim_{t\to\infty} \langle V_y(t+t')V_y(t) \rangle dt'. \quad (75)$$

For D_{zz} the procedure is fully analogous.

Equation (73) results in:

$$B_{ij} = \frac{2D_{ij}}{\tau_f^2}, \quad \text{for} \quad i,j = y,z. \quad (76)$$

A mathematically analog form to Eq. (76) can also be found in Ref. [46]. In contrast to D_{ij}, which scales as $a^2\dot{\gamma} = a^2/\tau_f$, cf. e.g., Ref. [7], our coefficients B_{ij} scale as a^2/τ_f^3, as can also be seen by regarding the dimensions in Eq. (71). Under use of the exponentially decaying correlation for long times t, i.e., Eq. (55), we find for the x direction the following:

$$\int_0^{\infty} \lim_{t\to\infty} \langle V_x(t+t')V_x(t) \rangle dt' = D_{xx} - \left\langle \int_0^{\infty} \lim_{t\to\infty} U_x^{\infty}(t')V_x(t)dt' \right\rangle = \frac{1}{2}\frac{d}{dt}\langle x^h x^h \rangle, \quad (77)$$

for proof, see Appendix C. The coupled U_x^{∞}, V_x term corresponds to the coupled term of Sierou and Brady [7] in Eq. (48). Consequently, we obtain:

$$B_{xx} = \frac{2}{\tau_f^2} \frac{1}{2} \frac{d}{dt} \langle x^h x^h \rangle. \quad (78)$$

For the xy component the procedure is analogous (compare proof for the xx component in Appendix C) resulting in the following:

$$\int_0^{\infty} \lim_{t\to\infty} \langle V_x(t+t')V_y(t) \rangle dt' = \frac{1}{2}\frac{d}{dt}\langle x^h y \rangle, \quad (79)$$

and, hence, we obtain

$$B_{xy} = \frac{2}{\tau_f^2} \frac{1}{2} \frac{d}{dt} \langle x^h y \rangle. \quad (80)$$

As shown above, the B_{ij} coefficients can be derived via the position diffusion coefficients, i.e., the mean-square displacements. As already mentioned in the context of the non-Brownian position diffusion tensors D_{ij} in Sec. II C, also the B_{ij} coefficients do not incorporate any position dependency since these are only one-particle velocity diffusion tensors. The mean-square displacements can be achieved either by experiments, see, e.g., the work of Breedveld *et al.* [1], or by numerical simulation, e.g., via the accelerated Stokesian dynamics method [19] as done by Sierou and Brady [7]. Hence, all terms necessary to determine $\langle V_i(t+t')V_j(t)\rangle$, and thus B_{ij}, are available. The drift for the colored-noise velocity results from Eq. (40) by calculating $\triangle V_i = V_i(t_{n+1}) - V_i(t_n)$ with Eq. (59) for small time-step sizes $\triangle t$.

Inserting the terms into Eq. (39) under the assumption of a composite Markov process yields

$$\begin{aligned}\frac{\partial \mathcal{P}(x,y,z,V_x,V_y,V_z,t)}{\partial t} &= -\frac{\partial}{\partial x}(U_x^\infty + V_x)\mathcal{P} - \frac{\partial}{\partial y}(V_y)\mathcal{P} - \frac{\partial}{\partial z}(V_z)\mathcal{P} \\ &+ \frac{1}{\tau_f}\frac{\partial}{\partial V_x}V_x\mathcal{P} + \frac{1}{\tau_f}\frac{\partial}{\partial V_y}V_y\mathcal{P} + \frac{1}{\tau_f}\frac{\partial}{\partial V_z}V_z\mathcal{P} \\ &+ \frac{1}{2}B_{xx}\frac{\partial^2\mathcal{P}}{\partial V_x\partial V_x} + \frac{1}{2}B_{yy}\frac{\partial^2\mathcal{P}}{\partial V_y\partial V_y} + \frac{1}{2}B_{zz}\frac{\partial^2\mathcal{P}}{\partial V_z\partial V_z} \\ &+ 2\frac{1}{2}B_{xy}\frac{\partial^2\mathcal{P}}{\partial V_x\partial V_y}. \end{aligned} \tag{81}$$

We observe the separated behavior in position terms and colored-noise velocity terms due to the composite Markov process. The third and fourth lines of Eq. (81) correspond to the Fokker-Planck equation of the Ornstein-Uhlenbeck process of $\boldsymbol{V}$. Altogether, Eq. (81) is a Fokker-Planck equation describing the shear-induced self-diffusion of non-Brownian particles taking into account long correlation times. The analysis of the different time-scale phenomena gives rise to the assumption that a coupled variable of position and colored-noise velocity is necessary in order to fulfill the Markov property.

IV. SOLUTION OF THIS FOKKER-PLANCK EQUATION: THE PROBABILITY DISTRIBUTION $\mathcal{P}$

Fokker-Planck equations of type of (39) can be solved according to Ref. [23] by a Gaussian distribution of the form

$$\mathcal{P}(\boldsymbol{s},t) = (2\pi)^{-\frac{1}{2}r}(Det\,\Xi)^{-\frac{1}{2}}\exp\left[-\frac{1}{2}(\boldsymbol{s}^T - \langle \boldsymbol{s}^T\rangle) \times \Xi^{-1}(\boldsymbol{s} - \langle \boldsymbol{s}\rangle)\right], \tag{82}$$

$$\text{with}\quad \Xi(t) = \int_0^t e^{(t-t')\boldsymbol{A}}\boldsymbol{D}e^{(t-t')\boldsymbol{A}^T}dt', \tag{83}$$

$$\langle \boldsymbol{s}\rangle = e^{t\boldsymbol{A}}\boldsymbol{s}(0), \tag{84}$$

and initial condition $\mathcal{P}(\boldsymbol{s},0) = \prod_{i=1}^6 \delta(s_i - s_i(0))$. The superscript T indicates the transposed terms.

In accordance to the algorithm of solution in Ref. [23], we set our initial variables $x(0)$, $y(0)$, $z(0)$, $V_x(0)$, $V_y(0)$, $V_z(0)$ to zero. Setting this into Eq. (84) shows that the variables $\langle x\rangle$, $\langle y\rangle$, $\langle z\rangle$, $\langle V_x\rangle$, $\langle V_y\rangle$, and $\langle V_z\rangle$ are zero in Eq. (82). For the actual evaluation of the analytic solution and an asymptotic expansion for large t, MAPLE16 (Maplesoft, Waterloo Maple Inc.) was employed. With the assumption $t \to \infty$ it is implied that the diffusive regime has started. Therefore, in the present work, terms of order $O(\exp(-t/\tau_f))$ are neglected, when summed with terms of order $O(t^0)$, and, respectively, terms of order $O(t^n)$ are neglected when summed with terms of order $O(t^{n+1})$, for $n \geqslant 0$. We have to keep in mind that the displacement in the x direction grows as $\sqrt{t^3}$ in contrast to the displacements in y and z directions which grow as $\sqrt{t}$, see Eqs. (42), (43), and (44). In the limit $t \to \infty$ to leading order the approximated probability distribution is given by:

$$\begin{aligned}\mathcal{P}(x,y,z,V_x,V_y,V_z,t) = (2\pi)^{-\frac{6}{2}}\left(\frac{(B_{xx}B_{yy} - B_{xy}^2)\dot{\gamma}^2B_{yy}^2B_{zz}^2t^5\tau_f^9}{96}\right)^{-1/2}\exp\Bigg\{-\frac{1}{2}\Bigg[\frac{(x - 1/2\dot{\gamma}yt)^2}{\frac{1}{12}B_{yy}t^3\dot{\gamma}^2\tau_f^2} + \frac{y^2}{B_{yy}t\tau_f^2} + \frac{z^2}{B_{zz}t\tau_f^2} \\ + \frac{2{V_x}^2}{(B_{xx} - B_{xy}^2/B_{yy})\tau_f} + \frac{2{V_y}^2}{(B_{yy} - B_{xy}^2/B_{xx})\tau_f} + \frac{4V_xV_y}{(B_{xy} - B_{xx}B_{yy}/B_{yy})\tau_f} + \frac{2{V_z}^2}{B_{zz}\tau_f} \\ + \frac{V_y\dot{\gamma}t(x - 1/2yt\dot{\gamma})}{1/12\dot{\gamma}^2t^3B_{yy}\tau_f} - \frac{2yV_y}{B_{yy}t\tau_f} - \frac{2zV_z}{B_{zz}t\tau_f}\Bigg]\Bigg\}. \end{aligned} \tag{85}$$

The prefactor of the Gaussian distribution can be decomposed into

$$\left(\frac{(B_{xx}B_{yy} - B_{xy}^2)\dot{\gamma}^2B_{yy}^2B_{zz}^2t^5\tau_f^9}{96}\right)^{-1/2} = \left(\frac{(B_{xx}B_{yy} - B_{xy}^2)B_{zz}\tau_f^3}{8}\right)^{-1/2}\left(\frac{\dot{\gamma}^2B_{yy}^2t^4\tau_f^4\,B_{zz}t\tau_f^2}{12}\right)^{-1/2}. \tag{86}$$

The highest order of the prefactor, i.e., Eq. (86) agrees with the composite Markov process, as it can be separated. The first part of the product on the right-hand side of (86) equals the prefactor of a process corresponding to a Fokker-Planck equation for an Ornstein-Uhlenbeck process in $\boldsymbol{V}$. From Eq. (55) with $t' = 0$, it follows that $\langle V_i(t)V_j(t)\rangle = \frac{B_{ij}\tau_f}{2}$; hence, the colored-noise velocity is in equilibrium for large times. Further, we want to compare our resulting probability distribution $\mathcal{P}(x,y,z,V_x,V_y,V_z,t)$ with the solution of Breedveld *et al.* [32], who use a Fokker-Planck equation solely in position space and gain a corresponding $\mathcal{P}(x,y,z,t)$. The second part of our prefactor in Eq. (86) equals the highest order of the prefactor of $\mathcal{P}(x,y,z,t)$ corresponding to their Fokker-Planck equation in position space, see Ref. [32]. The τ_f^2 terms in the second part arise due to the connection between B_{ij} and D_{ij}, cf. Eq. (76).

The first three terms of the exponent of our $\mathcal{P}(x,y,z,V_x,V_y,V_z,t)$ in (85) are position-related terms and agree with the highest-order terms of the $\mathcal{P}(x,y,z,t)$ from the position Fokker-Planck equation in Ref. [32], whereby the term $(x - 1/2\dot{\gamma}yt)^2$ implies the increasing affine motion of the particle from zero (due to the starting position $y = 0$) to the total velocity of $y\dot{\gamma}t$. The colored-noise terms in the exponent of our $\mathcal{P}(x,y,z,V_x,V_y,V_z,t)$, i.e., third and fourth lines of (85), correspond to the terms from a Fokker-Planck equation for the Ornstein-Uhlenbeck process $\boldsymbol{V}$. The terms in the fourth line of Eq. (85) account for a coupling effect of the displacement and the colored-noise velocity.

When the probability distribution $\mathcal{P}(x,y,z,V_x,V_y,V_z,t)$ from Eq. (85) is inserted into our Fokker-Planck colored-noise equation (81), it fulfills the Fokker-Planck equation to an accuracy in order $O((1/t)^{\frac{7}{2}})$. The time is supposed to be large in order to come into the shear-induced diffusion regime.

V. SUMMARY

The comparison of Brownian to non-Brownian diffusion behavior reveals that the short-time diffusivity of Brownian particles, captured by Fokker-Planck equations in position space [23], is not existent in the non-Brownian case. As shown, e.g., in Ref. [7], the non-Brownian particles only show long-time diffusive behavior. Other works in that context focus on modifying the diffusion tensor in the position space Fokker-Planck equation, e.g., Refs. [7,32], in order to describe non-Brownian particles. In contrast, our intention was to investigate whether generally the Fokker-Planck equation restricted to position space still is a suitable means to also describe the non-Brownian suspensions. For that, we started with an analysis of the Brownian particles' equation of motion in comparison to the non-Brownian case by using a new compact formulation. By means of the Kramers equation [23] we outlined a special situation which necessitates the use of a Fokker-Planck equation in coupled position-velocity space in the Brownian case. In that context, we showed that the non-Brownian case creates a similar situation and thus also requires an alternative Fokker-Planck equation. We adapted the approach of Ref. [1] where they assume that the hydrodynamic influence of the other particles results in a force with long correlation time, i.e., a colored-noise force. We reformulated the problem with a colored-noise velocity and assumed that this colored-noise velocity can be modeled by an Ornstein-Uhlenbeck process. Then we applied the strict rules of the mathematical framework of Markov processes and validity ranges of the Fokker-Planck equation described in Ref. [23] to show that the Markov property of the position in the present case is violated. Furthermore, we showed that the Fokker-Planck equation in position can be used neither for short time-step sizes nor for long time-step sizes. Our approach suggests that in the present case a coupled variable has to be introduced which then is a Markov variable again. This leads to the conclusion that the Fokker-Planck equation has to be built up in a coupled position and colored-noise velocity space. The method used to obtain all coefficients necessary to determine the new Fokker-Planck equation is shown. Furthermore, the resulting probability distribution is derived, which is an extension of the probability distributions derived from position space Fokker-Planck equations for non-Brownian particles [32] in combination with the probability distributions according to Ornstein-Uhlenbeck processes.

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APPENDIX A

We will prove the relations (71) and (72), i.e.,

$$\text{for} \quad \triangle t < \tau_f \text{ and } \triangle t \to 0\text{: } \langle \triangle V_i \triangle V_j \rangle_V = B_{ij}\triangle t + O(\triangle t^2), \tag{A1}$$

$$\text{for} \quad \triangle t > \tau_f \quad \text{and} \quad \triangle t \to \infty\text{:}$$
$$\langle \triangle V_i \triangle V_j \rangle_V = \frac{B_{ij}\tau_f}{2} + V_i(t_n)V_j(t_n), \tag{A2}$$

whereby the $\langle\rangle$ brackets are supposed to be related to t_n instead of $t = 0$.

From $\triangle V_i = V_i(t_{n+1}) - V_i(t_n)$ we have:
$\langle \triangle V_i \triangle V_j \rangle_V = \langle V_i(t_{n+1})V_j(t_{n+1})\rangle_V - \langle V_i(t_{n+1})V_j(t_n)\rangle_V - \langle V_i(t_n)V_j(t_{n+1})\rangle_V + \langle V_i(t_n)V_j(t_n)\rangle_V$.

The following rule from Ref. [39] for a general equation for the velocity autocorrelation according to an Ornstein-Uhlenbeck process is transferred to our colored-noise velocity as follows:

$$\begin{aligned}\langle V_i(t')V_j(t'')\rangle_V = {} & V_i(0)V_j(0)\exp\left[\frac{-(t'+t'')}{\tau_f}\right] \\ & + \frac{B_{ij}\tau_f}{2}\left\{\exp\left(\frac{-|t'-t''|}{\tau_f}\right)\right. \\ & \left. - \exp\left(\frac{-(t'+t'')}{\tau_f}\right)\right\}. \end{aligned} \tag{A3}$$

Transferring Eq. (A3) to $\langle V_i(t')V_j(t'')\rangle_V$ yields:

$$\begin{aligned}\langle V_i(t')V_j(t'')\rangle_V = {} & V_i(t_n)V_j(t_n)\exp\left[\frac{-(t'+t''-2t_n)}{\tau_f}\right] \\ & + \frac{B_{ij}\tau_f}{2}\left\{\exp\left(\frac{-|t''-t'|}{\tau_f}\right)\right. \\ & \left. - \exp\left[\frac{-(t'+t''-2t_n)}{\tau_f}\right]\right\}. \end{aligned} \tag{A4}$$

Therefore, we have the following:

$$\begin{aligned}\langle \triangle V_i \triangle V_j \rangle_V &= V_i(t_n)V_j(t_n)\exp(-2\triangle t/\tau_f) \\ &\quad + \frac{B_{ij}\tau_f}{2}[\exp(0) - \exp(-2\triangle t/\tau_f)] \\ &\quad - 2\bigg\{V_i(t_n)V_j(t_n)\exp(-\triangle t/\tau_f) \\ &\quad + \frac{B_{ij}\tau_f}{2}[\exp(-\triangle t/\tau_f) - \exp(-\triangle t/\tau_f)]\bigg\} \\ &\quad + V_i(t_n)V_j(t_n) \\ &= V_i(t_n)V_j(t_n)\exp(-2\triangle t/\tau_f) \\ &\quad + \frac{B_{ij}\tau_f}{2}[1 - \exp(-2\triangle t/\tau_f)] \\ &\quad - 2V_i(t_n)V_j(t_n)\exp(-\triangle t/\tau_f) + V_i(t_n)V_j(t_n).\end{aligned} \tag{A5}$$

For $\triangle t \to \infty$ the following applies:

$$\langle \triangle V_i \triangle V_j \rangle_V = \frac{B_{ij}\tau_f}{2} + V_i(t_n)V_j(t_n). \tag{A6}$$

Note that in the case where the $\langle\rangle$ brackets are related to $t = 0$ (instead of $\boldsymbol{V}$ in the index) with $t > \tau_f, t \to \infty$, it follows that $\langle V_i(t)V_j(t)\rangle = B_{ij}\tau_f/2$.

For $\triangle t \to 0$ the following applies:

$$\langle \triangle V_i \triangle V_j \rangle_V = B_{ij}\triangle t + O(\triangle t^2). \tag{A7}$$

APPENDIX B

Here we will show the $\triangle t^2$ dependence of $\triangle x^2$ in Eq. (70). Integrating Eq. (A4) yields:

$$\begin{aligned}&\int_{t_n}^{t}\int_{t_n}^{t}\langle V_i(t')V_j(t'')\rangle_V dt'dt'' \\ &= V_i(t_n)V_j(t_n)\tau_f^2\left\{1 - \exp\left[\frac{-(t-t_n)}{\tau_f}\right]\right\}^2 \\ &\quad + B_{ij}\tau_f\left\{\tau_f(t-t_n) + \tau_f^2\exp\left[\frac{-(t-t_n)}{\tau_f}\right] - \tau_f^2\right\} \\ &\quad - \frac{1}{2}B_{ij}\tau_f^3\left\{1 - \exp\left[\frac{-(t-t_n)}{\tau_f}\right]\right\}^2.\end{aligned} \tag{B1}$$

For $t = t_{n+1}$, in the limit $\triangle t = (t_{n+1} - t_n) \to 0$ we have the following: $\int_{t_n}^{t}\int_{t_n}^{t}\langle V_i(t')V_j(t'')\rangle_V dt'dt'' = V_i(t_n)V_j(t_n)\triangle t^2$.

APPENDIX C

We need to define $\int_0^\infty \lim_{t\to\infty}\langle V_x(t+t')V_x(t)\rangle dt'$ in order to find B_{xx}.

We will show the relation (77), i.e., $\int_0^t \langle V_x(t+t')V_x(t)\rangle dt' = \frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle$ in the limit $t \to \infty$. The case for B_{xy} is analog.

D_{xx} can be written as shown in Eq. (45): $\partial_t\langle xx\rangle - 2\langle x\,(\dot\gamma y)\rangle = 2D_{xx}$.

With $x(t) = \int_0^t (V_x(t') + U_x^\infty(t'))dt'$, $x(0) = 0$ we find:

$$\langle xx\rangle = \left\langle\left(\int_0^t \left[V_x(t') + U_x^\infty(t')\right]dt'\right)^2\right\rangle, \tag{C1}$$

$$\partial_t\langle xx\rangle = 2\left\langle\left(\int_0^t \left[V_x(t') + U_x^\infty(t')\right]dt'\right)\left[V_x(t) + U_x^\infty(t)\right]\right\rangle, \tag{C2}$$

with $\langle\int_0^t V_x(t)V_x(t')dt'\rangle = \langle\int_0^t V_x(t)V_x(t+t')dt'\rangle$ as $t \to \infty$. This can be shown by integrating Eq. (55) for both cases, t' and $t + t'$.

Inserting this into the equation for D_{xx} yields:

$$\begin{aligned}D_{xx} &= \frac{1}{2}\frac{\partial}{\partial t}\langle xx\rangle - \langle x(\dot\gamma y)\rangle \\ &= \left\langle\int_0^t V_x(t)V_x(t+t')dt'\right\rangle + \left\langle\int_0^t U_x^\infty(t)V_x(t+t')dt'\right\rangle \\ &\quad + \left\langle\int_0^t V_x(t)U_x^\infty(t')dt'\right\rangle + \left\langle\int_0^t U_x^\infty(t)U_x^\infty(t')dt'\right\rangle \\ &\quad - \left\langle\int_0^t U_x^\infty(t)V_x(t+t')dt'\right\rangle - \left\langle\int_0^t U_x^\infty(t)U_x^\infty(t')dt'\right\rangle,\end{aligned} \tag{C3}$$

$$D_{xx} = \left\langle\int_0^t V_x(t)V_x(t+t')dt'\right\rangle + \left\langle\int_0^t V_x(t)U_x^\infty(t')dt'\right\rangle. \tag{C4}$$

Thus,

$$\left\langle\int_0^t V_x(t)V_x(t+t')dt'\right\rangle = D_{xx} - \left\langle\int_0^t V_x(t)U_x^\infty(t')dt'\right\rangle \tag{C5}$$

It can be shown that $\langle\int_0^t V_x(t)U_x^\infty(t')dt'\rangle = \langle V_x(t)\int_0^t U_x^\infty(t')dt'\rangle$. So it follows that $\langle\int_0^t V_x(t)U_x^\infty(t')dt'\rangle = \langle\frac{dx^h}{dt}\int_0^t \dot\gamma y(t')dt'\rangle$. With the diffusion coefficient in the x direction from Ref. [7], see also Eq. (48) in Sec. II C, Eq. (C5) yields the following:

$$\begin{aligned}\left\langle\int_0^t V_x(t)V_x(t+t')dt'\right\rangle &= \int_0^t \langle V_x(t)V_x(t+t')\rangle dt' \\ &= \frac{1}{2}\frac{d}{dt}\langle x^h x^h\rangle.\end{aligned} \tag{C6}$$

APPENDIX D

The derivation of the drift coefficient can also be compared to the explanation of the Kramers equation. In case that the time step on which we regard the system is larger than τ_f, we can also define a new time scale based on the length that a particle would travel in the time $\triangle t$. We call this new length L which actually is of the same magnitude as the particle radius a, but here we just want to show the matter of principle. The corresponding time scale is called τ_k with $\tau_k \gg \tau_f$ with the $\triangle t$ now small on τ_k. Now we go back to Eq. (1) with

$\boldsymbol{F}^P = 0$, $\boldsymbol{F}^B = 0$ and nondimensionalize on the new time scale τ_k,

$$\boldsymbol{m}\frac{d\boldsymbol{U}}{dt} = -\boldsymbol{R}_{\mathrm{FU}}(\boldsymbol{U} - \boldsymbol{U}^\infty) + \boldsymbol{R}_{\mathrm{FE}} : \boldsymbol{E}^\infty, \tag{D1}$$

$$\tau_p \frac{L}{\tau_k \tau_k}\tilde{\boldsymbol{m}}\frac{d\tilde{\boldsymbol{U}}}{d\tilde{t}} = -\tilde{\boldsymbol{R}}_{\mathrm{FU}}\left(\tfrac{L}{\tau_k}\tilde{\boldsymbol{U}} - \tfrac{L}{\tau_f}\tilde{\boldsymbol{U}}^\infty\right) + \tfrac{L}{\tau_f}\tilde{\boldsymbol{R}}_{\mathrm{FE}} : \tilde{\boldsymbol{E}}^\infty. \tag{D2}$$

We see that the term including the shear flow dominates over all other terms; this is the same situation as described in the context of the Kramers equation where the time scale has to be scaled down from τ_D to τ_p, which finally required a coupled variable of position and velocity.

[1] V. Breedveld, D. van den Ende, M. Bosscher, R. J. J. Jongschaap, and J. Mellema, J. Chem. Phys. **116**, 10529 (2002).
[2] G. K. Batchelor and J. T. Green, J. Fluid Mech. **56**, 401 (1972).
[3] G. K. Batchelor and J. T. Green, J. Fluid Mech. **56**, 375 (1972).
[4] J. Morris, Rheol. Acta **48**, 909 (2009).
[5] J. F. Brady and J. F. Morris, J. Fluid Mech. **348**, 103 (1997).
[6] A. Acrivos, G. K. Batchelor, E. J. Hinch, D. L. Koch, and R. Mauri, J. Fluid Mech. **240**, 651 (1992).
[7] A. Sierou and J. F. Brady, J. Fluid Mech. **506**, 285 (2004).
[8] Y. Wang, R. Mauri, and A. Acrivos, J. Fluid Mech. **327**, 255 (1996).
[9] F. R. Da Cunha and E. J. Hinch, J. Fluid Mech. **309**, 211 (1996).
[10] I. Rampall, J. R. Smart, and D. T. Leighton, J. Fluid Mech. **339**, 1 (1997).
[11] G. Subramanian and J. F. Brady, J. Fluid Mech. **559**, 151 (2006).
[12] E. C. Eckstein, D. G. Bailey, and A. H. Shapiro, J. Fluid Mech. **79**, 191 (1977).
[13] D. Leighton and A. Acrivos, J. Fluid Mech. **177**, 109 (1987).
[14] G. Bossis and J. F. Brady, J. Chem. Phys. **87**, 5437 (1987).
[15] G. Drazer, J. Koplik, B. Khusid, and A. Acrivos, J. Fluid Mech. **460**, 307 (2002).
[16] M. Marchioro and A. Acrivos, J. Fluid Mech. **443**, 101 (2001).
[17] T. N. Phung, J. F. Brady, and G. Bossis, J. Fluid Mech. **313**, 181 (1996).
[18] J. F. Brady and G. Bossis, Annu. Rev. Fluid Mech. **20**, 111 (1988).
[19] A. Sierou and J. F. Brady, J. Fluid Mech. **448**, 115 (2001).
[20] D. J. Pine, J. P. Gollub, J. F. Brady, and A. M. Leshansky, Nature **438**, 997 (2005).
[21] D. L. Ermak and J. A. McCammon, J. Chem. Phys. **69**, 1352 (1978).
[22] D. R. Foss and J. F. Brady, J. Fluid Mech. **401**, 243 (1999).
[23] N. G. van Kampen, *Stochastic Processes in Physics and Chemistry*, 3rd ed. (Elsevier, Amsterdam, 2007).
[24] T. J. Murphy and J. L. Aguirre, J. Chem. Phys. **57**, 2098 (1972).
[25] G. Wilemski, J. Stat. Phys. **14**, 153 (1976).
[26] D. Wycoff and N. L. Balazs, Physica A **146**, 175 (1987).
[27] G. Subramanian and J. F. Brady, Physica A **334**, 343 (2004).
[28] V. Breedveld, D. van den Ende, R. J. J. Jongschaap, and J. Mellema, J. Chem. Phys. **114**, 5923 (2001).
[29] I. Santamaría-Holek, G. Barrios, and J. M. Rubi, Rev. Mex. Fís. **55**, 77 (2009).
[30] I. Santamaría-Holek, G. Barrios, and J. M. Rubi, Phys. Rev. E **79**, 031201 (2009).
[31] R. Mauri and D. Leporini, Europhys. Lett. **76**, 1022 (2006).
[32] V. Breedveld, D. van den Ende, A. Tripathi, and A. Acrivos, J. Fluid Mech. **375**, 297 (1998).
[33] H. Nicolai, B. Herzhaft, E. J. Hinch, L. Oger, and E. Guazzelli, Phys. Fluids **7**, 12 (1995).
[34] J. K. G. Dhont, *An Introduction to Dynamics of Colloids* (Elsevier, Amsterdam, 1996).
[35] G. Bossis and J. F. Brady, J. Chem. Phys. **80**, 5141 (1984).
[36] H. Brenner and M. E. O'Neill, Chem. Eng. Sci. **27**, 1421 (1972).
[37] S. Kim and R. T. Mifflin, Phys. Fluids **28**, 2033 (1985).
[38] L. Durlofsky, J. F. Brady, and G. Bossis, J. Fluid Mech. **180**, 21 (1987).
[39] H. Risken, *The Fokker-Planck Equation* (Springer, Berlin, 1996).
[40] O. G. Bakunin, *Chaotic Flows Correlation Effects, Transport and Structures* (Springer, Berlin, 2011).
[41] Y. Drossinos and M. W. Reeks, Phys. Rev. E **71**, 031113 (2005).
[42] H. Haken, *Synergetics An Introduction Nonequilibrium Phase Transitions and Self-Organization in Physics, Chemistry and Biology* (Springer-Verlag, Berlin, 1977).
[43] J. F. Morris and J. F. Brady, J. Fluid Mech. **312**, 223 (1996).
[44] D. E. Elrick, Aus. J. Phys. **15**, 283 (1962).
[45] G. E. Uhlenbeck and L. S. Ornstein, Phys. Rev. **36**, 823 (1930).
[46] N. G. van Kampen, J. Stat. Phys. **54**, 1289 (1989).