1 Introduction and motivation

A suspension may be defined as a disperse multi-phase system wherein the disperse phase consisting of solid particles, Brownian or non-Brownian, is suspended in a continuous phase that may be a liquid or a gas. The definition may be extended to include cases where the disperse phase consists of liquid drops (emulsions) or bubbles, provided the deformation of the drops (or bubbles) and processes of break-up and coalescence do not play a major role in the dynamics. The importance of suspensions can hardly be overemphasized. Suspensions are ubiquitous, and we use/consume many of the on a daily basis:

2. Milk is itself a colloidal suspension containing micelles (of primarily the protein Casein) in the size range 0.04 – 3\(\mu\)m. The larger of these micelles scatter visible light rendering milk opaque.
3. Ice cream - a suspension of microscopic ice crystals and sugar dispersed in foamed cream.
4. Aerosols or hydrosols - suspensions of fine liquid droplets or fine solid particles in a liquid or a gas. Here, the shelf life of the final concoction is of critical importance, and this is directly related to the question of aerosol/hydrosol stability.
5. Toothpaste - a concentrated paste of hydrogen peroxide and baking soda that behaves as a soft solid, and can therefore support a yield stress. The latter ensures that a toothpaste does not flow out of the tube under its own weight.

6. Mayonnaise - a stable emulsion containing vegetable oil, vinegar and egg yolk. Lecithin, a natural surfactant contained in egg yolk stabilizes the droplets of vegetable oil.

7. Paints - latex paints, for instance, are stable water-based emulsions of solid monomers (the pigment particles).

8. Biological fluids, for instance, blood. Blood is a concentrated suspension (suspended constituents make up about 45% of the available volume) of predominantly RBC’s, and in addition, WBC’s and platelets in a plasma medium (itself containing dissolved proteins, and therefore non-Newtonian). Blood flow through arteries is an interesting but complex fluid dynamical problem. The flow occurs under the action of an oscillatory pressure gradient, through an elastic walled tube, where the tube diameter changes from about an inch for the aorta (the largest artery) to less than the size of the suspended constituents for the smallest capillaries.

9. Nano-particle suspensions, popularly known as nanofluids, are suspensions of nano-metre sized metal or metal oxide particles (Au, Ag, Fe, Al₂O₃, etc.) suspended in aqueous or polar organic solvents (for instance, ethylene glycol). These suspensions have been touted as the future transfer agents. Preliminary experiments appear to indicate a considerable enhancement in the effective thermal conductivity; an enhancement seemingly incommensurate with the small volume fraction of the suspended particles.

10. Bacterial suspensions - The fluid mechanics of suspensions of micro-organisms (for instance, algae like C. Nivalis, or bacteria including E.Coli and B. Subtilis) is becoming an increasingly interesting topic for study. They are usually termed as ‘active’ particle suspensions, and differ fundamentally from suspensions of passive particles. Indeed, the swimming of micro-organisms, and the energy metabolized in this process, implies that such systems are inherently out of equilibrium even in the absence of an imposed flow. Hydrodynamic interactions in groups of microbes may play a crucial role in the formation of bio-films, and therefore in the initiation of infection processes. Modelling the dynamics of micro-organisms found in seawater, such as phytoplankton and algae which constitute the base of the sea-food chain, evidently has implications for the oceanic ecosystem. Studying the swimming mechanism of a single micro-organism offers insight into possible evolutionary processes at work. From a fundamental point of view, suspensions of micro-organisms (and those of passive sedimenting particles) are also of interest to physicists, since they serve as a model non-equilibrium system.

11. Paper-pulp industry - The process of paper manufacture involves the processing of cellulose fibre suspensions in an aqueous medium. The orientation of fibres under flow conditions decides, in part, the property of the final paper product.

12. An understanding of the dynamics and stability of agitated gas-solid suspensions is fundamental to understanding the operation of fluidized bed catalytic reactors employed in the chemical industry.

Vast quantities of some of the aforementioned products, and others in a raw material form, are handled by the chemical, bio-chemical and pharmaceutical industries. Thus, there is a clear need for an understanding of the flow behavior or rheology of these systems; understanding that would then translate to better and more efficient handling of such materials in an industrial scenario. The difficulty in analyzing suspension flow behavior arises because suspensions are quite unlike simple fluids like water or air (Newtonian fluids). The latter, in the incompressible limit, are entirely characterized by a single material property - the shear viscosity. On the other hand, the above examples suggest that suspensions may exhibit a wide range of flow characteristics ranging from solid-like (squeezing toothpaste out of a tube) to even gas-like (the bed of particles in a gas-solid fluidized beds). In fact, complex flow behavior must be the rule rather than the exception; in particular, because the size of the suspended particles may range over more than six orders of magnitude, and the governing physical phenomena, relevant to macroscopic flow behavior, are quite radically different at the different length scales. For instance, one has colloidal gold particles, with a diameter of a few nanometers, constituting nanofluids at one end of the spectrum, through disk-shaped, charged, clay particles
a fraction of a micron in diameter, bi-concave RBC’s with a diameter of $1 - 10\mu$, to finally sand particles and cellulose fibres with a size on the order of millimeters. The resulting extreme variation in flow characteristics suggests that one would need more than a mere viscosity in order to characterize the flow behavior of such materials.

That the rheology of suspensions is likely to be complicated may also be understood from the fundamental inter-play between the relaxation and deformation time scales. A rather naive estimate of the structural relaxation time scale ($\tau_{\text{rel}}$) in a molecular liquid, assuming the inter-molecular spacing (of the order of angstroms) to be the relevant length scale, and the speed of sound ($300m/s$) to be the relevant velocity scale, yields $\tau_{\text{rel}} \approx 10^{-13} \text{sec}$. The relaxation time scale is larger for gases, where the mean free path ($\approx 0.1\mu$) provides the relevant length scale. In either instance, $\tau_{\text{rel}}$ still remains very small compared to the time scale of most imposed deformations (about $10^{-3}\text{sec}$ in rapid flows). Thus, the structure of molecular liquids or gases is hardly perturbed under normal conditions. The flow being a small perturbation, one expects a response linear in the rate of deformation; this, of course, leads to the well-known Navier-Stokes equations.

On the other hand, suspensions come under the general category of complex fluids, fluids that possess a meso-scale structure, that is to say, a structure on a length scale that is intermediate between the macroscopic and molecular length scales. Let us take an example: consider the flow of an aqueous suspension of cellulose fibres, materials that are routinely handled in the paper-pulp industry. The suspension typically flows through a pipe a metre in diameter, the macroscopic length scale. On the other hand, the suspended fibres themselves are of the order of a millimetre in length. The macroscopic flow behavior crucially depends on both the spatial arrangement and the orientation distribution of the suspended fibres on the millimetre length scale. An anisotropic orientation of the fibres may lead to a non-Newtonian bulk rheology. Thus, one needs to understand the dynamics of the suspended fibres on the millimetre length scale, the latter being the relevant mesoscopic length scale. A second example is that of a block copolymer. This is a material made of polymer chains, wherein each chain, as the name ‘co-polymer’ suggests, consists of two incompatible portions. Although the covalent linkage between the dissimilar parts prevents macroscopic phase separation, the mutual incompatibility leads to a whole variety of meso-scale structures, ‘meso’ here referring to the length scale of the polymer chain which for sufficiently high molecular weights is, of course, much larger than the molecular length scale. The flow behavior of such block co-polymers is again crucially related to the microscopic dynamics of the various phases when subjected to a flow. Figure 1 illustrates this aspect for the flow of a suspension of spherical particles. The meso-scale structure, in the context of suspensions, is usually referred to as the microstructure, since the disperse phase typically comprises micron-sized Brownian particles. For reasons of conveniences, we will continue to use the term ‘microstructure’ even when the particles do not inhabit this size range, and may, in fact, be large enough for Brownian motion to be negligible. In either of the aforementioned examples, the existence of a meso-scale structure implies the existence of relaxation time scales for the complex fluid that are much larger than those in molecular fluids. In turn, this implies that complex fluids such as suspensions are easily driven out of equilibrium, and the resulting non-equilibrium states with a significantly deformed micro-structure are characterized by a complicated non-linear rheology.

2 Disperse multi-phase systems

As implied in the previous section, an analysis of the flow of any complex fluid involves, in essence, a deduction of equations governing the macro-scale flow behavior from information obtained at the microstructural level. Specifically, one solves the governing equations describing the interaction of the two phases on the microstructural length scale, and then uses appropriate averaging techniques to calculate a bulk flow property such as the coefficient of viscosity. This would yield, in principle, the viscosity as a function of the parameters characterizing the constituent phases, and possibly, the flow. For instance, one would obtain the suspension

\[ \text{1That it is very difficult to drive molecular fluids out of equilibrium is evident from the extreme conditions required for the Navier-Stokes description to break down. An example is that of shock waves in rarefied gases, where one falls back on the more fundamental (and formidable) Boltzmann equation in order to calculate the structure of a shock on scales of the order of a few mean free paths.} \]
viscosity as a function of the particle shape and size, the viscosity of the suspending fluid, and the flow rate. Such an effective viscosity would then serve as an input in equations that model the suspension as an effective fluid, and therefore govern its motion on the macroscopic length scale.

2.1 Governing equations for fluid (continuous) phase

Assuming the suspending fluid to be Newtonian in nature, the equations governing its motion, on all length scales larger than molecular, are the well-known Navier-Stokes equations [1]. In the dimensionless form, these equations may be written as:

\[
SLRe \frac{\partial \mathbf{u}}{\partial t} + Re \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla \hat{p} + \nabla^2 \mathbf{u} + \frac{Re}{Fr} \hat{g},
\]

where \( Re = Ua / \mu \) is the Reynolds number, \( Sl = a / (U \tau) \) is the Strouhal number, \( Fr = U^2 / ga \) is the Froude number, and \( \hat{g} \) denotes the direction of gravity. Here, \( a \) is a length scale characteristic of the microstructure, say, the particle size, \( U \) is a relevant micro-structural velocity scale, and \( \rho \) and \( \mu \) are the density and viscosity of the suspending fluid. Thus, the Reynolds number \( Re \) is a dimensionless parameter that governs the relative importance of inertial and viscous forces at the micro-scale, while the Strouhal number \( Sl \) is the ratio of the convective time scale to any imposed time scale. The Froude number (\( Fr \)), in a similar manner, is the ratio of inertial forces to that of gravity. It is of relevance only in flows involving free surfaces, and will not be considered hereafter; indeed, in the absence of free surfaces, one may define a dynamic pressure, \( p = \hat{p} - \frac{Re}{Fr} \hat{g} \), and thereby eliminate the body force from the equations of motion.

It must be noted that the micro-scale dimensionless parameters may differ from their macro-scale analogues by orders of magnitude owing precisely to the disparity between the macroscopic and microstructural length and time scales. Thus, \( Re \) above is usually of order unity or smaller, indicating that inertial forces are usually of secondary importance at the micro-structural level. Returning to our favorite example of the flow of a suspension through a pipe, inertia is certainly important for the suspension flow on the length scale of a pipe diameter. However, the effective suspension viscosity needed, for instance, to calculate the velocity profile across the pipe, may be determined from an inertialess micro-scale problem; the latter would involve a homogeneous suspension of spherical particles subjected to a linear flow. Clearly, therefore, the physics at the macro- and micro-scales can be very different. A second example is the turbulent coagulation of aerosols.

\[\text{2}^2\text{That the suspending fluid is Newtonian is a simplification introduced only for the purposes of this write-up. One can, and does, encounter suspensions of particles in viscoelastic fluids (polymer solutions or melts). The rheological characteristics of such suspensions are of much importance in understanding the behavior of filled polymeric materials.} \]
The rate of coagulation in an aerosol depends on the relative velocity of particle-pairs. For sufficiently large particles, and in the absence of significant polydispersity, it is therefore controlled by the underlying turbulent motion, in particular, that on scales smaller than the Kolmogorov scale. This is because the shear rate in homogeneous isotropic turbulence increases with decreasing length scale, and is therefore the highest at the Kolmogorov scale. On this scale, the relevant micro-scale for the problem, the motion is laminar, and may again be modelled as a linear flow, but with a fluctuating velocity gradient tensor and an appropriate correlation time.

If one assumes $Re \ll 1$, and neglects the non-linear term representing convective inertia, one obtains the unsteady Stokes equations:

$$SlRe \frac{\partial u}{\partial t} = -\nabla p + \nabla^2 u,$$

which are a system of linear equations that govern unsteady viscous phenomena, typically involving high frequency, low amplitude oscillations in a viscous fluid. The restriction to low amplitude is so the rapid oscillation does not lead to separation. In these cases, the time scale characterizing the fluid motion is much smaller than the convective time scale, implying that $Sl \gg 1$; $\tau$ is, in fact, comparable to the time scale for the viscous diffusion of momentum, and the product $SlRe$ is therefore of order unity despite the smallness of the Reynolds number. That the effects of unsteady acceleration, $\partial u/\partial t$ cannot be neglected may be simply seen by a scaling analysis. If $A(<<a)$ be the small amplitude of oscillation, and $\omega$ its frequency, then

$$\frac{\partial u}{\partial t} \sim \frac{O(A\omega)}{\omega} \sim O(A\omega^2),$$

$$u \cdot \nabla u \sim O(A\omega), \frac{O(A\omega)}{a} \sim A^2 \omega^2 a.$$ (4)

The ratio of the non-linear to the unsteady terms therefore becomes $O(A/a) \ll 1$.

An example where the flow described by (2) might be of relevance is Atomic force micro-scopy (AFM). Specifically, the tapping mode in AFM, used for probing the surfaces of soft samples, typically consists of an oscillating cantilever (at frequencies $\sim MHz$) close to the test surface. Calibration usually involves modelling the probe motion as that of an oscillator moving in a potential well, and thereby determining the defining parameters of the oscillator - its mass, and damping coefficient. The proximity to the surface decides the nature of the potential energy profile, while the nature of the unsteady viscous flow around the cantilever contributes, in part, to both its effective inertia (on account of the added mass), and damping. Thus, determination of the response curve requires the solution of an unsteady flow problem. A second example is the Brownian motion, which is the random motion of particles, usually smaller than a micron in size, in a solvent. It is the result of the particle being constantly buffeted around by colliding solvent molecules. For Brownian particles not much heavier than the fluid, the particle velocity fluctuates sufficiently rapidly for unsteady viscous effects to become important.

On the other hand, for small-scale, slowly varying motions ($Sl \sim 1$), one may indeed neglect the Eulerian acceleration, $\partial u/\partial t$, to obtain the quasi-steady Stokes equations:

$$-\nabla p + \nabla^2 u = 0,$$

which represent an instantaneous balance between pressure and viscous forces. As will be seen later in section 4.1, both the linearity and the quasi-steady nature of (5) represent enormous simplifications. In fact, (5) is central to almost all of suspension mechanics!

### 2.2 Governing equations for disperse (particle) phase

The equations of motion for a single particle are, of course, just Newton’s equations. Thus, mass times the acceleration of the particle must equal the sum of the forces acting on it viz. $m \frac{dU}{dt} = \Sigma F$. Taking $F$ to be the well-known Stokes drag, one obtains a dimensionless measure for the importance of particle inertia - the Stokes number ($St$). For a sedimenting particle, one obtains $St = \frac{mU}{\frac{3}{2} \pi \mu a^2}$; if the velocity scale $U$ is determined
by an imposed shear rate ($\dot{\gamma}$), one has $St = \frac{m\dot{\gamma}}{6\pi\mu a}$. The Stokes number may be regarded as the ratio of a momentum relaxation time scale, $\tau_p = \frac{m}{6\pi\mu a}$, to an imposed flow time scale (say, $\dot{\gamma}^{-1}$). In particular, using $m = \frac{4\pi}{3}\rho_p a^3$ for a spherical particle, and neglecting numerical pre-factors, we have $St = \rho_p U a/\mu$, $\rho_p$ being the density of the particle. One now observes that $St$ is, in fact, $Re$ times the density ratio $\rho_p/\rho_f$. For cases where $St$ is small enough to be neglected, for instance, in the case of a sub-micron Brownian particle, the momentum balance is merely equivalent to an instantaneous balance of all forces involved.

Although, one may write down an equation of motion for each particle in the system, integration of which should, in principle, yield the particle trajectory, in most instances such detailed information on particle trajectories is not needed. Rather, one models the motion of the particles in a statistical sense. The statistical approach entails modelling the degrees of freedom of the disperse particulate phase using a probability density function. For example, if $\{F_i\} (i = 1, 2, \ldots, N$ denotes the particle number) denotes the relevant degrees of freedom associated with a single disperse phase element (particle), one may define a probability density function $P(\{F_i\}, t)$ such that $P(\{F_i\}, t)d\Pi(\{F_i\})$ denotes the probability of finding the disperse phase in a differential element of phase space. One then writes down a kinetic equation governing the evolution of the $P(\{F_i\}, t)$. Successive moments of the probability density function give information, in an averaged sense, about the dynamics of the disperse phase. We will briefly consider two such examples in order to illustrate the basic ideas.

The first is a Brownian particle which follows a random trajectory on account of repeated collisions with solvent molecules. The inherent uncertainty inherent in the latter process clearly suggests a statistical approach. The resulting stochastic differential equation, the Langevin equation, or the equivalent kinetic equation for the positional probability, the Smoluchowski equation, form the basis of all studies of Brownian suspensions (see equations (6), (18) and figure 2 below). A similar approach is necessary for the case where one deals with an ensemble of non-Brownian particles. Here, it is the random nature of the microstructure, either the spatial arrangement or the velocity distribution of the particles or both, and the resulting interactions, that is modelled via a probability density function. Thus, the second example we consider is that of the Boltzmann equation, again a kinetic equation governing the evolution of the probability density for the spatial arrangement or the velocity distribution of the particles or both, and the resulting interactions, as well as the dynamics of the disperse phase.

2.2.1 Brownian motion

As seen above, particles smaller than a micron respond perceptibly to collisions with solvent molecules, and consequently move along what appears to be a random trajectory even in a quiescent fluid. Rather than modelling Brownian motion by trying to solve the coupled equations of motion of the particle, and those governing the enormous number of solvent molecules ($\sim 10^{23}$) constituting the fluid bath, one models the effect of the collisions on the particle in a statistical sense by including a random force in the equation of motion. Thus, the solvent is still treated as a continuum. The total force acting on the particle is split into a deterministic viscous drag, modelled here by the quasi-steady Stokes drag\textsuperscript{3}, and the aforementioned random force with zero mean. The properties of the random force are specified via its moments, and reflect the solvent degrees of freedom; they may be determined from considerations involving equipartition of energy.

With the introduction of the random force, one obtains the Langevin equation for a spherical Brownian particle\textsuperscript{[2]}:

$$m\frac{dU}{dt} = -6\pi\mu a U + F^B,$$

or using the scales $a$ and $(a/U)$ for length and time,

$$St\frac{d\dot{U}}{dt} = -\dot{U} + \frac{F^B}{(6\pi\mu a)},$$

\textsuperscript{3}This drag may be thought of as a consequence of the asymmetry of solvent molecule collisions with a translating particle. The probability of collisions on the fore side becomes greater than that on the aft, the net momentum transfer is in a direction opposite to the particle velocity.
in dimensionless form; here, $St = mU/(6\pi\mu a^2)$, as before. It may be regarded as the ratio of the (small) momentum relaxation time of the particle, $\tau_p \equiv m/(6\pi\mu a)$, to the time $a/U$ required for the particle to move a distance of the order of its own size. In (7), $F^B$ satisfies the following properties:

$$\langle F^B(t) \rangle = 0,$$

$$\langle F^B(t) F^B(t') \rangle = 2kT(6\pi\mu a) \delta(t - t').$$

The latter property indicates an instantaneous de-correlation of the random force. This is because the random force remains correlated only for an interval of time comparable to that between successive uncorrelated collisions with solvent molecules. However, this latter time scale ($10^{-12}$ sec) is much smaller than any time scale characterizing particle motion, and therefore appears instantaneous on these time scales. Further, for times longer than $O(\tau_p)$, the momentum of the particle has relaxed, and inertia is unimportant ($St \ll 1$). Thus, neglecting the particle acceleration in (6), one obtains

$$\frac{dx}{dt} = \frac{F^B}{(6\pi\mu a)},$$

for the velocity of an inertialess Brownian particle. The averaged motion of the Brownian particle may readily be deduced from the solution of (10):

$$x(t) = \int_0^t \frac{F^B(t')}{(6\pi\mu a)} dt'.$$

Clearly, (8) implies $\langle x(t) \rangle = 0$; using (11), the mean displacement of the particle is zero. On the other hand, the mean square displacement of the particle is given by

$$\langle x(t)x(t) \rangle = \int_0^t \int_0^t \frac{F^B(t') F^B(t'')}{(6\pi\mu a)} dt' dt'',$$

$$= \frac{1}{(6\pi\mu a)^2} \int_0^t \int_0^t \langle F^B(t') F^B(t'') \rangle dt' dt'',$$

$$= \frac{2kT}{(6\pi\mu a)} \int_0^t dt' \int_0^t \delta(t' - t'') dt'',$$

$$= \frac{2kT}{(6\pi\mu a)} t,$$

and is therefore linear in time. Thus, for times much longer than $O(\tau_p)$ the Brownian particle starts to diffuse with a diffusion coefficient given by $D = kT/(6\pi\mu a)$. The time scale required for the particle to diffuse a distance of $O(a)$ is $O(a^2/D)$, and characterizes the micro-structural changes (particle rearrangement) in an equilibrium suspension of Brownian particles. In other words, $a^2/D$ is a good estimate for the configurational relaxation time in a dilute Brownian suspension. Evidently, the kinetic equation for the spatial probability, $P(x, t)$, corresponding to (10) is a diffusion equation given by:

$$\frac{\partial P}{\partial t} = \frac{kT}{6\pi\mu a} \nabla_x^2 P$$

In presence of flow, (10) takes the form

$$\frac{dx}{dt} = U^\infty + \frac{F^B}{(6\pi\mu a)},$$

where $U^\infty$ is the imposed external flow, and we have assumed that the single particle, but for its Brownian motion, would be convected by this flow. The rheology of Brownian suspensions is now determined by the competing effects of the deforming flow (with a time scale $a/U^\infty$, or $\dot{\gamma}^{-1}$, a characteristic shear rate), and the aforementioned relaxation time of $O(a^2/D)$ that acts to restore an isotropic microstructure. The ratio of the two time scales is known as the Peclet number, $Pe = 6\pi\mu a^2\dot{\gamma}/kT$, and is a dimensionless measure of the departure of the Brownian suspension from equilibrium. Again, it may be shown that equation (17) is
exactly equivalent to a probabilistic description, and the corresponding equation governing the evolution of the positional probability, the Smoluchowski equation, is given by:

$$\frac{\partial P}{\partial t} + U^\infty \cdot \nabla_x P = \frac{1}{\rho_c} \nabla^2 P$$  \hspace{1cm} \text{(18)}$$

Equation (18) is a convection-diffusion equation for a single particle, and corresponds to the physical picture where particles in a Brownian suspension diffuse relative to each other, while being simultaneously convected by the imposed flow. As the previous statement suggests, equation (18) may be generalized to describe a multi-particle system, where the variable $x$ now represents the spatial degrees of freedom for all particles. Thus, for a system of $N$ spherical particles, $x \equiv (x_1, x_2, \ldots, x_N)$. One would also have to account for the effects of hydrodynamic interactions between different particles in this case; this leads to both a tensorial Brownian diffusivity that is now a function of the particle configuration, and a deviation of the particle pathlines from the fluid streamlines; that is, $U^\infty$ in (18) is no longer the ambient fluid velocity at $x$ (see section 4.6.1).

We note that equation (18) is valid only on times scales much greater than $O(\tau_p)$, since it does not account for the momentum degrees of freedom associated with the Brownian particles. This is not really a limitation in most cases since the changes in the spatial configuration of a Brownian suspension are effected by particles displacements of $O(a)$; on the other hand, the distance travelled by a Brownian particle in a time of $O(\tau_p)$, the Brownian mean free path ($\lambda_B$), is $O(kT/m)^{1/2} \cdot O(\tau_p)$, and one finds $\lambda_B \ll a$. In other words, inertia plays no role in configurational displacements of $O(a)$. A kinetic equation for the probability, valid on time scales of $O(\tau_p)$ or smaller, will therefore mainly account for momentum relaxation processes. Thus, in contrast to the Smoluchowski equation, this kinetic equation, known as the Fokker-Planck equation, is one in phase space $(x, U)$. In addition, as indicated earlier in the discussion following (2), one needs to also account for the effects of unsteady fluid inertia on time scales of $O(\tau_p)$. This leads to the drag on a Brownian particle being history dependent, and the resulting governing equation for the probability is no longer an evolution (differential) equation. It is, in fact, an integro-differential appropriate to a history dependent non-Markovian process.

\subsection{2.2.2 Rapid granular flows (Granular gases)}

The particles or the ‘grains’ in a granular gas are much larger than Brownian particles, typically being of the order of a few millimetres in size. The suspending fluid is a gas, and owing to the large density ($\rho_f$) of the order of a few millimetres in size. The suspending fluid is a gas, and owing to the large density ratio($\rho_p/\rho_f \gg 1$), viscous forces in the fluid, although large compared to fluid inertial forces ($Re \ll 1$), are much smaller when compared to the inertia of the particles ($St \gg 1$). The grains are therefore hardly affected by the viscous drag exerted by the gaseous phase, and the dominant changes in the grain velocities occur due to collisions with other grains. Unlike molecular collisions, the collisions between the macroscopic grains are inelastic and dissipate energy. Similar to the Brownian motion example, it is these inter-grain collisions that necessitate a statistical approach to the dynamics of granular gases.

Since grain inertia is dominant in these systems, one must account for both the spatial and momentum degrees of freedom of the grains, and the probability density function, $P(x, U, t)$, is therefore defined in phase (position + velocity) space$^4$. The kinetic equation governing the evolution of the probability density function of a single particle, in a dilute granular gas, under the combined effect of a driving force (an external flow, for instance), and inelastic binary collisions, is the Boltzmann equation ([4], [3]):

$$\frac{\partial P}{\partial t} + U_1 \cdot \nabla_{x_1} P - \frac{(6\pi \mu a)}{m} \nabla_{U_1} \cdot [(U_1 - U^\infty)] = \int \int d\sigma dU_2 \left[ \frac{1}{e^2} P(U_1')P(U_2') - P(U_1)P(U_2) \right].$$  \hspace{1cm} \text{(19)}$$

where $P \equiv P(x_1, U_1, t)$ and $e (0 < e < 1)$ is the coefficient of restitution that measures the inelasticity of a collision; $e = 1$ implies perfectly elastic collisions. The second term on the left hand side represents the spatial convection of probability, similar to that in Brownian suspensions. The third term denotes the

$^4$In this sense, the probabilistic description is similar to that for Brownian suspensions on times scales smaller than $O(\tau_p)$ in section 2.2.1.
change in probability on account of the small viscous deceleration experienced by the grain as it moves through the gaseous phase. The effect of binary collisions with a second particle is represented by the integral on the right hand side, where \( \sigma \) denotes the collision cross-section. Herein, the term proportional to \( P(U_1)P(U_2) \) represents the effect of direct collisions, between the given grain with velocity \( U_1 \) and a second grain with velocity \( U_2 \), which act to convect probability out of the interval \((U_1, U_1 + dU_1)\), while the term proportional to \( P(U'_1)P(U'_2) \) represents the effect of inverse collisions between a pair of grains with velocities \( U'_1 \) and \( U'_2 \), and act to convect probability into the aforementioned velocity interval. In presence of inelasticity, the inverse collisions are relatively more intense (by a factor of \( O(1/e^2) \)), and correspondingly less probable (in the absence of a flow), in order to compensate for the loss in kinetic energy. For both direct and inverse collisions, one usually neglects any correlation between the velocities of colliding grains, an assumption that becomes increasingly accurate in the dilute limit. The difference between the direct and inverse collisions represents the net change in probability due to binary collisions. Note that appropriate collision rules uniquely define the pre-collisional velocities associated with an inverse collision in terms of those involved in a direct collision; thus, in (19), we have \( U'_i \equiv F_i(U_1, U_2) \). Equation (19), owing both to its non-linearity, and the non-locality in velocity space, is a formidable one to solve.

In rapid granular flows, collisions are responsible for the relaxation of the micro-structure. Thus, in a manner similar to Brownian suspensions, one may define an appropriate dimensionless parameter that characterizes the departure of the velocity distribution in granular flow from an approximate Maxwellian equilibrium. This parameter would then be the analog of the Peclet number, and is the ratio of the collision time to energy balance. For small values of \( \dot{\gamma} \) for a molecular gas in the absence of flow.

The word ‘equilibrium’ here refers to the nature of the velocity distribution function. The latter is, of course, exactly Maxwellian for a molecular gas in the absence of flow.

Thus, the rheological properties of the granular flow are expected to be a function function of the ratio \( \dot{\gamma}a^\phi/T^\phi \). In a manner similar to molecular gases, the former may be defined as \( a^\phi \) for a dilute granular flow, where \( a^\phi \) is the mean free path for \( \phi \ll 1 \), and \( T \) is the granular temperature, and an appropriate measure of the intensity of granular velocity fluctuations. Thus, the rheological properties of the granular flow are expected to be a function function of the ratio \( \dot{\gamma}a^\phi/T^\phi \). The temperature here is determined in terms of the external driving force from a macroscopic energy balance. For small values of \( \dot{\gamma}a^\phi/T^\phi \), the random motion of the grains, in a manner analogous to molecules, is responsible for the existence of a granular viscosity. However, even in the limit where the random motion is very intense, and the velocity distribution of the grains is thence close to a Maxwellian, the resulting rheological behavior is not Newtonian; the granular viscosity, unlike the molecular viscosity, remains dependent on the shear rate via \( T \).

3 Phase diagram for disperse multi-phase flows

In this section, we attempt to organize the various known and studied flow regimes of disperse multi-phase systems in an appropriate three-dimensional parameter space (see figure 2). The latter consists of the three most important dimensionless parameters introduced in the earlier sections viz. \( Re \), \( St \) and \( Pe \). The lecture notes will largely focus on the regime of non-Brownian Stokesian suspensions. The typical disperse phase element (solid particle) in such systems is too large to be Brownian \((Pe \rightarrow \infty)\), but is still sufficiently small, and the suspending fluid sufficiently viscous, so both the inertia of the disperse phase element, and that associated with the flow of the suspending fluid on comparable length scales are negligible \((Re = 0, St = 0)\).

Since the vertical axis in figure 2 denotes the value of \((1/Pe)\), the Stokesian suspension regime is, strictly speaking, represented by a single point in parameter space - the origin. In practice, this regime refers to the limiting scenario \( Re, St \ll 1, Pe \gg 1 \), and therefore encompasses a small region around the origin of parameter space. One may approach this limiting region in parameter space in different ways, and different lines of approach correspond to different Stokesian systems; for instance, systems that tend to the origin along the line \((Re = St, Pe \rightarrow \infty)\) correspond to non-Brownian neutrally buoyant Stokesian suspensions, where the densities of the disperse and continuous phases are identical. Systems approaching the origin in the region \( Re < St (Pe \rightarrow \infty)\) correspond to sedimenting Stokesian suspensions, while those approaching the origin with \( Re > St \) correspond to buoyant emulsions, where the disperse phase comprises liquid drops.

If the force were not velocity dependent, the entire left hand side of (19) may be regarded as a convected derivative in phase space.

Note that the granular flow is still out of equilibrium, and will always be, on account of inelastic dissipation. The use of the word ‘equilibrium’ here refers to the nature of the velocity distribution function. The latter is, of course, exactly Maxwellian for a molecular gas in the absence of flow.

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or gas bubbles, and is lighter than the suspending liquid. Such a classification helps put the general regime of Stokesian suspensions in perspective; while the study of suspensions in this regime has been a dominant theme in the field of suspension rheology, as figure 2 indicates, the regime is only one among several other regimes of flow encountered for disperse multi-phase systems.

Again referring to figure 2, we will first describe the limiting flow regimes encountered as one starts from the origin and moves along each of the three axes. With \( Re = St = 0 \) and decreasing \( Pe \), we move over to the regime of Brownian or colloidal suspensions (seen earlier in section 2.2.1). The particle size in these systems ranges from a tens of nanometres to about a micron, and inertia, either that of the particle or fluid, is evidently unimportant at these length scales. The tiny particles undergo Brownian motion, and the flow characteristics of these suspensions, for a given particulate volume fraction, are therefore a function of \( Pe \), being determined by a balance between the deforming action of flow and the restoring action of Brownian forces. Since the ratio \( Pe/Re = Sc \), the Schmidt number, is constant for a given multi-phase system, the increasing magnitude of inertial forces (increasing \( Re \)) usually implies the diminishing importance of thermal (Brownian) forces (increasing \( Pe \)).

With \( Re = Pe^{-1} = 0 \) and increasing \( St \), we first transition to the regime of gas-solid suspensions (\( \rho_p/\rho_f \gg 1 \), \( St > O(1) \)), wherein inertia of the particles (typically ranging in size from about 1 – 50 microns) becomes increasingly important. The total dissipation still has significant contributions from both inelastic collisions, and the viscous dissipation in the interstitial fluid phase. Note that the rather small size of the particles ensures that inertial forces in the fluid remain small; in other words, \( Re \ll 1 \). For still larger particles (\( a \sim \text{mm} \)) we approach the limit of rapid granular flows. These correspond to the limit \( St \gg 1 \), when the effect of fluid viscosity on the motion of the particle phase becomes negligible. Examples of rapid granular flows include gas-solid fluidized beds, chute flows of grains, landslides and avalanches, etc. As seen earlier in section 2.2.2, modelling of such systems exploits their similarity to molecular gases.

With \( St = Pe^{-1} = 0 \) and increasing \( Re \), we have a scenario where particle inertia is negligible, but fluid inertial forces become increasingly important, for instance, in inertial bubbly flows (\( \rho_p/\rho_f \ll 1 \)). In general, there is very little known with regard to the rheology of multi-phase flows with continuous phase inertia. However, progress has been made in a particular flow regime - the flow of bubbly dispersions at large \( Re \) and small Weber numbers (\( We \)); here, \( We \) is a dimensionless parameter that measures the relative importance of inertial stresses (\( \propto \rho_f U^2 \)) and that due to surface tension (\( \sigma/a \), where \( \sigma \) is the coefficient of surface tension). Thus, small \( We \) is the limit of dominant surface tension when the bubbles are close to spherical in shape. In addition, the free-slip condition on the bubble surface (valid in the absence of surface-active agents) implies that the motion of the liquid is well approximated by potential flow. Since pressure fluctuations in an incompressible propagate at an infinite speed, the equations governing potential flow (\( \nabla^2 \phi = 0 \), where \( \phi \) is the velocity potential) are both linear and quasi-steady; thus, they afford the same simplifications as the equations for Stokes flow (see 5). In the above limit, the vorticity in the continuous phase remains small, and one may have a kinetic theory formulation for the bubble phase that is, in a sense, similar to those for granular flows. In fact, the added mass of a bubble plays role analogous to the actual mass of a grain, except that the former remains a function of the instantaneous bubble configuration. Thus, the random fluctuating motion of the bubbles contributes to a bubble phase viscosity.

In the region away from the axes, both \( Re \) and \( St \) are finite in general. Examples of such flows include those of solid-liquid suspensions where the suspending liquid has a low viscosity (e.g: water). For sufficiently high values of \( Re \) and \( St \), one may even have a transition to a turbulent multi-phase flow. In such systems, the presence of vorticity in the continuous phase enormously complicates the nature of the flow. Much remains to be understood, from a fundamental standpoint, regarding the nature of such flows.

The term ‘phase diagram’ has been used rather loosely here. We have essentially distinguished between various dynamical regimes for disperse multi-phase flows in an appropriate parameter space, rather than attempting a classification of equilibrium/non-equilibrium phases for a particular complex fluid. It must also be kept in mind that any classification of complex fluid flows in terms of only a few parameters is bound
to involve some simplifying assumptions, and hence, excludes other interesting systems or flow regimes. For instance, in figure 2, we have implicitly focussed on suspensions of spherical particles which are characterized by a single length scale (the radius \( a \)). All dimensionless parameters may then be defined with respect to this length scale. For suspensions of anisotropic particles, the aspect ratio also plays an important role in determining the bulk rheology of the suspension. This is of particular importance at small \( Re \), where the underlying fluid flow is primarily determined by the largest length scale of the disperse phase element, for instance, the length rather than the diameter of a slender fibre. Thus, the transport properties of a fibre-reinforced composite are more likely to scale with the hydrodynamic volume fraction (for instance, the length rather than the diameter of a slender fibre). This is because, the inertia of the particles in such flows drives them into very close contact, and may itself be dependent on the flow, as is the case for bubbles or drops that deform and become increasingly elongated in fast flows. In fact, this induced deformation causes the bubbles to shed vorticity, just like solid particles, and bubbly flows in this limit assume the same degree of complexity as inertial flows of solid-liquid suspensions referred to above.

For any disperse multi-phase system, one must, in principle, account for all the relevant degrees of freedom of the disperse phase elements in formulating, for instance, a kinetic equation for the evolving probability density, and the subsequent averaging procedure (see sections 2.2.1 and 2.2.2). Figure 3 lists a few examples of disperse phase elements in order of increasing degrees of freedom. The above discussion does permit us to briefly list the relevant degrees of freedom for various flows involving spherical disperse phase elements:

1. Stokesian suspensions \((Re, St \ll 1)\): \( \{x_i\} \),
2. Granular flows of smooth spheres \((St \gg 1)\): \( \{x_i, U_i\} \),
3. Granular flows of rough spheres \((St \gg 1)\): \( \{x_i, U_i, \Omega_i\} \),
4. Bubbly flows \((St \ll 1, Re \gg 1)\): \( \{x_i, U_i\} \),
5. Inertial suspensions \((Re \sim O(1))\): \( \{x_i, U_i, u(x, t)\} \).

Here, \( i = 1, 2, \ldots, N \) is an index denoting the particle number, while \( x \in V_i \), \( V_i \) being the volume of the suspending fluids. Thus, for the case of inertial suspensions, the evolving disperse phase configuration also depends on the the instantaneous velocity field, \( u(x, t) \), in the entire fluid domain. Alternately, projecting the fluid degrees of freedom onto those of the particles would translate to a dependence on the entire time history of the disperse phase configuration instead. A Markovian description would entail only the instantaneous positions and/or velocities of the disperse phase elements, and the non-Markovian nature of the evolution for \( Re, St \sim O(1) \) clearly reflects the general complexity of multi-phase flows with micro-scale inertia.

We have also not accounted for the existence of non-hydrodynamic forces between particles. These include colloidal and van-der Waals forces between sub-micron particles, and electrostatic forces that arise in suspensions of charged particles. These phenomena usually increase in importance with decreasing particle size, and thus, the \( 1/Pe \) axis in figure 2 may be regarded as a general measure of the importance of such non-hydrodynamic effects. In addition, non-continuum and compressibility effects may often play a key role in determining the stability of aerosols. The former are, in fact, also important in flows of gas-solid suspensions; this is because, the inertia of the particles in such flows drives them into very close contact, and the resulting dissipation is a function of the flow in the thin intervening layer of fluid.

Finally, it must be mentioned that we have not accounted for the effect of concentration (the volume fraction \( \phi \) of the disperse phase) in the above parametric description. This is a very important factor, and as will be seen later, the behavior of concentrated suspensions can differ quite dramatically from dilute ones. The Brownian diffusivity is a function of the suspension volume fraction, and tends to the value, \( kT/(6\pi \mu a) \), appropriate for a single particle, only in the limit \( \phi \to 0 \). In general, hydrodynamic interactions reduce the diffusivity to less than this value with increasing \( \phi \), by essentially increasing the effective viscosity; thus, \( \mu \), used in the above expression for the diffusivity, may now be regarded as an appropriate suspension viscosity.
and is therefore a function of $\phi$. As a result, the configurational relaxation time of the Brownian suspension increases with increasing $\phi$, and it becomes easier to drive a concentrated suspension out of equilibrium, eliciting a non-linear rheological response. Indeed, concentrated Brownian suspensions exhibit significant viscoelasticity. In dimensionless terms, $Pe$ for a concentrated Brownian suspension, defined in terms of a volume fraction dependent diffusivity, becomes quite large even at relatively modest shearing rates. In fact, even in the absence of a flow, a suspension of Brownian particles interacting via excluded volume forces alone undergoes a transition to an ordered solid phase at $\phi \approx 0.5$. Such transitions, and the resulting solid-like rheological behavior become important at much lower volume fractions in the presence of long-ranged electrostatic interactions. In this write-up, however, we will primarily focus on the quantitative analysis of suspension rheology in the dilute limit, implicitly assuming the existence of a disordered phase. Occasional remarks will address the qualitative behavior of suspensions at higher concentrations.

Disperse multi-phase systems

Phase diagram I:

- $Re = \frac{\gamma a^2}{v}$
- $St = \frac{\gamma a^2}{v_p}$
- $Pe = \frac{\gamma a^2}{D}$
- $Sc = \frac{v}{D}$
- $Sc_p = \frac{v_p}{D}$
- $We = \frac{\rho_p \gamma a^3}{\sigma}$

Inertial Brownian suspensions, Polymer solutions

Suspension mechanics

Hydrodynamics of Stokesian suspensions

1/Pe

$Re = \frac{We}{p}$

Non-Brownian Sedimenting suspensions

Non-Brownian buoyant suspensions/emulsions ($\rho_p < \rho_f$)

Neutrally buoyant suspensions/emulsions ($\rho_p = \rho_f$)

Bubbly flows

Re >> 1 St << 1 We << 1

(Increasing fluid inertia implies athermal systems)

Stokes flows

(Re=0 St=0 Pe= $\infty$)

Inertial flows with continuous phase vorticity

(Fast flows with low viscosity, two-phase turbulence)

Figure 2: The figure helps organize the various possible flow regimes for a suspension in terms of the dimensionless parameters introduced in the previous section. Note that the dimensionless parameters are defined by taking the velocity scale $U$ to be $O(\dot{\gamma}a)$, where $\dot{\gamma}$ is a representative micro-scale shear rate, and $a$ is the size of the disperse phase element.
Disperse multi-phase systems

Phase diagram II:

Other physical phenomena include:
- Colloidal (DLVO) forces - Brownian dispersions
- Electroviscous effects - Charged particles
- Non-continuum and compressibility effects - Coagulating aerosols
- Non-Newtonian suspending fluids - Filled polymeric materials

4 Stokesian suspensions

Herein, we will primarily focus on the limit $Pe = \infty$ in an attempt to understand the role of hydrodynamics in suspension rheology. We will later combine this insight with Brownian motion to examine the rheology of a Brownian suspension as a function of $Pe$.

4.1 Properties of Stokes flow

Recall that the equations governing the motion of the suspending fluid in a Stokesian suspension are the quasi-steady Stokes equations ([5],[6]) together with the equation of continuity:

\[-\nabla p + \nabla^2 \mathbf{u} = 0, \quad (20)\]
\[\nabla \cdot \mathbf{u} = 0, \quad (21)\]

supplemented by no-slip boundary conditions on the surface of each particle. It may easily be seen by taking the divergence and curl of (20) that the pressure $p$, and the vorticity, $\mathbf{\omega} = \nabla \wedge \mathbf{u}$, respectively, satisfy the Laplace’s equation. Thus, $\nabla^2 p = \nabla^2 \mathbf{\omega} = 0$. A consequence of the latter equation is that peak values of vorticity can only be attained on boundaries. Physically, this happens because, in Stokes flow, the vorticity field in the fluid phase is determined solely by diffusion of vorticity generated at solid or fluid boundaries. This immediately implies that an intense concentration of vorticity can never occur in the interior of the fluid phase in Stokes flow. Indeed, quite unlike high $Re$ flows, recirculating streamline patterns in Stokes flows are not indicative of compact vortical structures.
4.1.1 Linearity and Quasi-steadiness

The linearity of the Stokes equations allows for the superposition of solutions. Thus, if \((u_1, p_1)\) and \((u_2, p_2)\) are solutions of the Stokes equations, so is \((\alpha u_1 + \beta u_2, \alpha p_1 + \beta p_2)\), where \(\alpha\) and \(\beta\) are constants. The superposition principle may often be used to obtain the solution to a flow problem without ever having to solve the equations of motion in detail. An example is illustrated in figure 4, where linear superposition leads to the conclusion flow that a translating and rotating particle in an ambient simple shear will not experience a lift force at zero \(Re\). This is on account of the symmetry inherent in the simpler constituent problems. The final result again runs counter to one’s intuition developed from high \(Re\) flows. At high \(Re\), both a spinning particle in a uniform flow, and a translating particle in shear flow experience lift forces, and this may be explained, in part, by appealing to the existence of transverse pressure gradient as in Bernoulli’s principle. The latter principle is, of course, only valid in the inviscid limit \((Re \rightarrow \infty)\).

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Translating in a quiescent fluid

\((u_1, p_1)\)

Rotating in a quiescent fluid

\((u_2, p_2)\)

Fixed particle in simple shear

\((u_3, p_3)\)

Translating and rotating particle in simple shear

\((\alpha u_1 + \beta u_2 + \gamma u_3, \alpha p_1 + \beta p_2 + \gamma p_3)\)

Figure 4: The figure shows that the linearity of the Stokes equations are incompatible with the presence of a lift force on a translating, rotating particle in simple shear flow.

The quasi-steady nature of the Stokes equations ensures that Stokes flows driven by solid boundaries are not explicitly dependent on time. Physically, this results from assuming the diffusion of vorticity to occur instantaneously to all parts of the possibly unbounded fluid domain. Thus, any such Stokes flow is only a function of the instantaneous motion and position of a particular bounding surface. For a Stokesian suspension, in particular, this would imply that velocity field in the interstitial fluid would only depend on the instantaneous positions and velocities of the suspended particles. This constitutes an enormous simplification even when compared to the unsteady Stokes equations which still neglect the non-linear convective terms. Indeed, if one were to include unsteady viscous effects, then the drag on an accelerating particle would no longer be given just by the Stokes drag corresponding to its instantaneous velocity, but also involves the so-called Basset term that depends on its entire history of acceleration\(^7\). The situation for drops is quite analogous. In this case, one must likewise specify the instantaneous positions and velocities of boundaries if any, as well as the instantaneous shape of the possibly deformed drop. This then determines the instantaneous velocity field in both the external fluid domain, and within the drop\(^8\).

4.1.2 Insensitivity to geometry

The force on a translating particle at zero \(Re\) is remarkably insensitive to its shape. An example is that of a slender fibre; even in the limit of infinite aspect ratio, the drag on a fibre settling vertically is only twice that in the transverse orientation. In either case, the drag differs from that on a sphere, with radius equal to the fibre length, only by the logarithm of the fibre aspect ratio. This insensitivity is quite remarkable in

\(^7\)It is this history dependence that led to a corresponding history dependence in the kinetic equation for the probability density describing the statistics of Brownian motion on time scales of \(O(\tau_p)\) or smaller.

\(^8\)There is an intrinsic relaxation time even for the case of non-Brownian drops on account of surface tension. The appropriate time scale for the surface tension driven motion \(O(\mu a/\sigma)\), where \(\mu\) is higher of the two viscosities. The ratio of this time scale to \(\dot{\gamma}^{-1}\) is the Capillary number \((Ca)\). This dimensionless parameter characterizes the departure from equilibrium of a dilute emulsion; one again expects a non-Newtonian response for sufficiently large \(Ca\), when the drops are no longer spherical.
light of the fact that, at high $Re$, a slender body settling broadside-on may exhibit vortex shedding, its drag being dominated by pressure forces (form drag), while the drag in its longitudinal orientation may still be dominated by skin friction in the boundary layer, and thereby be much much smaller. The resulting drag for the longitudinal and transverse orientations at high $Re$ may differ by $O(Re^2)$.

The insensitivity of the drag to the detailed configuration of the particle may also be used to obtain an estimate for the intrinsic viscosity of a dilute polymer solution. Since the stress due to the polymer chains has an entropic origin, one may write the polymer contribution to the viscosity as $\eta_p = O(G\lambda)$, where $G = n kT$ is the elastic modulus, and $\lambda = R_g^2/D$ is the relevant configurational relaxation time; here, $n$ is the number density of polymer chains, $R_g$, the radius of gyration, is a measure of the size of a polymer coil, and $D$ is its diffusion coefficient. On account of the shape insensitivity, the viscous resistance of such a coil may be taken as that corresponding to a sphere of radius $R$.

This then breaks the reversibility symmetry. In the case of a deformed drop is always so as to minimize its interfacial area regardless of the direction of boundary motion. As shown in figure 5, reversing the motion of the particle interchanges the fore and aft regions, while reversing time does not. Thus, the two situations, that of reversed boundary motion and that of reversed time, can only be equivalent when the fore and aft regions are mirror images of each other, or in other words, the velocity field is fore-aft symmetric. More intuitively, ‘reversing boundary motions and external forces reverses time in Stokes flow’. As will be illustrated using a few examples below, reversibility imposes severe constraints on the nature of Stokes flow solutions\(^3\), and these constraints have significant implications for the rheology of suspensions in the hydrodynamic limit ($Pe = \infty$).

We begin by considering the simplest case, the velocity field around a translating spherical particle at zero $Re$ (see figure 5). Reversibility implies that the velocity field around a translating fore-aft symmetric particle must also be fore-aft symmetric. Let us see why this is so in the context of the aforementioned spherical particle by assuming a fore-aft asymmetric velocity field to begin with (see the left most illustration in figure 5). As shown in figure 5, reversing the motion of the particle interchanges the fore and aft regions, while reversing time does not. Thus, the two situations, that of reversed boundary motion and that of reversed time, can only be equivalent when the fore and aft regions are mirror images of each other, or in other words, the velocity field is fore-aft symmetric.

Next, consider an axisymmetric particle, for instance, a cylindrical fibre, sedimenting at zero $Re$. In this case, one has an additional degree of freedom - the fibre orientation. Referring to figure 6, we first note that, unlike a sphere, the Stokes settling velocity of a fibre coincides with the direction of gravity only for the horizontal and vertical orientations. The smaller resistance encountered when a fibre sediments in the longitudinal (vertical) orientation causes an inclined fibre to ‘slide’ along a direction that is intermediate between that of gravity and its axis of symmetry. Now, if such a fibre were to rotate while sedimenting, then, in accordance with the reversibility principle, reversing the direction of gravity must cause it to rotate in the opposite sense. As figure 6 shows, a change in reference frame leads to an immediate contradiction: that the fibre might as well rotate in either direction. The resolution of this apparent contradiction is that a sedimenting fibre, or more generally any axisymmetric particle, at zero $Re$ must retain its orientation. It

\(^3\)Note that the principle of reversibility does not apply to the motion of drops at zero $Re$. The surface tension driven flow in the case of a deformed drop is always so as to minimize its interfacial area regardless of the direction of boundary motion. This then breaks the reversibility symmetry.
Figure 5: Reversibility implies that the velocity field around a translating sphere must be fore-aft symmetric. Thus, the notion of a wake behind translating particles does not apply at zero \( Re \).

must, however, be mentioned that even a tiny amount of inertia causes such an axisymmetric particle to eventually drift towards the horizontal orientation.

We now examine the motion of a neutrally buoyant (density matched) particle in an imposed flow, specifically, that of a freely rotating particle in a pipe or a channel flow at a location away from the centreline. Such a particle will not migrate laterally at zero \( Re \). In light of the above examples, the relevant argument employing the reversibility principle is easily seen. Say, the particle migrates toward the centre of the pipe. A reversal of the flow should instead lead to a wall-directed migration in what, to within a change in reference frame, is an identical physical scenario. thus, one is led to conclude that the particle may migrate either way! The reason for this absurd inference is, of course, our initial assumption of a non-zero transverse velocity. In fact, it may be shown that, at zero \( Re \), even an anisotropic axisymmetric particle, such as a fibre, will, on average, not undergo a lateral migration in a pipe flow.

The above scenario is no longer true at finite \( Re \). In fact, even for very small \( Re \), particles in a pipe are known to eventually migrate to a radial location that is intermediate between the centreline and the wall \( (r_{eq} = 0.6R, R \) being the pipe radius). This is known as the Segre-Silberberg effect after the scientists who originally discovered the phenomenon in 1961 ([8],[9]). In fact, lateral migration of particles in parallel flows is a generic phenomenon at finite \( Re \), since the presence of lift forces are a manifestation of irreversibility; for
instance, particles in a wall bounded couette flow eventually migrate to the centreline at small but finite \( Re \). It will be seen later that lateral migration may even occur at zero \( Re \) in a concentrated suspension on account of shear-induced diffusion (see section 4.9). In general, such particle migration enormously complicates the interpretation of rheological data - inertial (finite \( Re \)) migration for dilute suspensions and migration driven by shear-induced diffusivities in concentrated suspensions. This is because such systems tend towards a highly inhomogeneous state in the limit of long times (see figure 7), and the measured viscosity (say) on these time scales would no longer correspond to a homogeneous suspension.

![Figure 7: Inertial (finite Re) particle migration leading to inhomogeneity in a pipe flow.](image)

Reversibility leads to the ubiquitousness of closed streamlines in Stokes flows. For instance, one expects the existence of regions of closed streamlines around rotating particles, and in addition, closed orbits for particle pairs in planar ambient flows with sufficient vorticity. An elementary reversibility argument in either case, along the lines of the earlier examples, excludes the presence of streamlines or pathlines that either spiral in or out. It will be seen later in section 4.7 that the presence of closed orbits for pairs of particles has profound implications with regard to the microstructure and rheology of dilute Stokesian suspensions. An exactly analogous situation occurs in the shearing flow of a dilute suspension of non-interacting non-Brownian orientable particles. Orientable particles may be characterized by a unit vector along their orientation. Reversibility then implies that the trajectories of this vector on the unit sphere must be closed orbits; these are known as the Jeffery orbits. In the absence of non-hydrodynamic forces or inertia, the orientation distribution across Jeffery orbits remains indeterminate, and the indeterminacy manifests as a dependence of the rheology on the initial distribution of particle orientations! Evidently, the principle of reversibility is a recurring theme in Stokes flow, and has far-reaching implications with regard to the dynamics and rheology of Stokesian suspensions.

### 4.1.4 Kinematic irreversibility

While the Stokes equations are linear in the velocity variables, the resulting pathlines of infinitesimal fluid elements are given by \( \frac{dx}{dt} = u(x, t) \), \( u(x, t) \) being the Stokes velocity field, and these kinematic equations are non-linear in almost every case. Thus, although the linearity of the Stokes equations lead to dynamic reversibility, the non-linearity of the kinematic equations, and the resulting sensitive dependence on initial conditions, may nevertheless lead to kinematic irreversibility in many instances, in particular, when the configuration of particles becomes sufficiently complicated. In other words, such Stokes flows yield chaotic streamlines or particle pathlines. For instance, in a sheared suspension at sufficiently high concentrations, even the smallest disturbances would lead to the particles departing from their perfectly reversible trajectories. Dynamical systems theory dictates that such a departure will occur at an exponential rate, and very soon, all correlation with initial conditions will be lost. Clearly, in such a situation, reversing the boundary motion won’t result in particles or fluid elements retracing their original trajectories, unless one looks at an increasingly short interval of time. The concept of kinematic irreversibility is, in fact, crucial to the existence

\[ ^{10} \text{On account of degeneracies introduced by the principle of reversibility at zero Re, even a small irreversible force, such as that due to inertia or viscoelasticity may have a significant effect when acting over a long period of time. As a result, one expects even small non-hydrodynamic forces to have important consequences for the steady state rheological properties of a suspension.} \]
of long-time diffusive behavior in Stokesian suspensions (see 4.9).

4.2 Solving Stokes equations

Our discussion in the previous sections with regard to Stokes flow phenomena has been strictly qualitative. Beginning with this section, we will analyze a few Stokes flow problems in quantitative detail \[10\].

In order to solve

\[-\nabla p + \nabla^2 u = 0, \tag{22}\]

we write the velocity field as the sum of a homogeneous and a particular solution:

\[u = u^H + u^P, \tag{23}\]

where

\[\nabla^2 u^H = 0, \tag{24}\]

\[\nabla^2 u^P = \nabla p. \tag{25}\]

A solution to (25) is given by

\[u^P = \frac{p}{2} \frac{x_i}{r}, \tag{26}\]

which may be seen as follows. Using index notation,

\[\nabla^2 u^P_i = \frac{1}{2} \frac{\partial^2}{\partial x_j^2} \left( p x_i \right), \tag{27}\]

\[= \frac{1}{2} \left[ x_i \frac{\partial^2 p}{\partial x_j^2} + 2 \frac{\partial p}{\partial x_j} \frac{\partial x_i}{\partial x_j} \right], \tag{28}\]

where we have used Lebnitz’s rule for the differentiation of a product, and that \(\nabla^2 x = 0\). Since \(\nabla^2 p = 0\), and \(\frac{\partial x}{\partial x_j} = \delta_{ij}\), one obtains

\[\nabla^2 u^P_i = \frac{\partial p}{\partial x_i}, \tag{29}\]

verifying that (26) is indeed a particular solution of (25). Now, both \(u^H\) and \(p\) satisfy the Laplace’s equation, and the general form of the solution to the Laplace’s equation may be written down using the machinery of spherical harmonics.

In order to do this, we first note that the fundamental solution (Green’s function) of the Laplace’s equation in three dimensions is \(1/r\) where \(r = |x| = x_1 x_i\); that is to say, \(\nabla^2 (1/r) = \delta(x)\). Other, independent solutions, in an invariant tensor formulation, may be determined by successive differentiation, followed by multiplication with an appropriate power of \(r\). The solutions that vanish with increasing \(r\) are the decaying harmonics, while those that increase with increasing \(r\) are the growing harmonics\(^{11}\)\(^{11}\). The \(n\)th order decaying harmonic is \(O(r^{-n-1})\), being given by \(\frac{\partial^n}{\partial x_1 \partial x_2 \ldots \partial x_n} \left( \frac{1}{r} \right)\); the corresponding growing harmonic is \(O(r^n)\), and is given by \(r^{2n+1} \frac{\partial^n}{\partial x_1 \partial x_2 \ldots \partial x_n} \left( \frac{1}{r} \right)\). The former are relevant to exterior Stokes problems involving unbounded domains, where one requires the velocity field to vanish at large distances, while the growing harmonics come into play in bounded domains. The first few members, both growing and decaying harmonics, derived in this manner are tabulated below:

\[^{11}\text{Note that the decaying harmonics are not really solutions of the homogeneous equations. However, the forcing function is a generalized function localized at the origin. For the fundamental solution, this is, of course, the delta function. Thus, all decaying harmonics are singular at the origin, while growing harmonics, with the exception of the constant, are singular at infinity. This then is consistent with the well-known Liouville’s theorem in complex analysis, which states that the only non-trivial entire function is a constant.}\]
Thus, the general form of both $\mathbf{u}^H$ and $\mathbf{p}$ is given by a linear superposition of the above harmonics. The order of the harmonics, and the values of the multiplying constants are determined from the equation of continuity and boundary conditions. The infinite series of growing and decaying spherical harmonics, when written out in spherical polar coordinates $(r, \theta, \phi)$, takes the form well-known from solving the Laplace’s equation in the same coordinate system using separation of variables, that is, the general term in this infinite series is of the form $r^n P^m_n(\theta) e^{im\phi}$, where $P^m_n(\theta)$ is the associated Legendre polynomial. However, we prefer to work with spherical harmonics, since the solution so derived is also in an invariant tensor notation. Thus, the spherical harmonics are first used as building blocks to derive the appropriate Stokes solution. Thereafter, the requirement that the resulting solution be linear in parameters appearing in the boundary conditions, and respect the appropriate coordinate invariance, determines the velocity and pressure fields. The requirement of coordinate invariance is readily implemented using spherical harmonics.

4.3 Motion of a single particle

4.3.1 Single rotating sphere at zero $Re$

We examine an isolated sphere rotating with angular velocity $\Omega$ in an otherwise quiescent fluid. Choosing the origin to be the centre of the sphere, the problem, in dimensionless terms, may be stated as follows:

$$\nabla^2 \mathbf{u} - \nabla p = 0, \quad (30)$$
$$\nabla \cdot \mathbf{u} = 0, \quad (31)$$

with the boundary conditions

$$\mathbf{u} = \Omega \wedge \mathbf{r} \quad \text{at} \quad r = 1, \quad (32)$$
$$\mathbf{u}, p \to 0 \quad \text{for} \quad r \to \infty. \quad (33)$$

Now, the disturbance pressure field must vanish as $r \to \infty$, $r$ being the distance from the centre of the sphere, and in addition, be linear in $\Omega$. Since $\Omega$ is a vector, the required spherical harmonic is $x/r^3$. Thus,

$$p = C \frac{\Omega \cdot x}{r^3} = C' \frac{\epsilon_{ijk} \Omega_j x_k}{r^3}, \quad (34)$$

$C$ being a constant of proportionality. However, $\Omega$ is a pseudo-vector, having a handedness associated with it. Such vectors transform differently (compared to true vectors) on a reflection of the coordinate system. Thus, the right hand side of (34) is a pseudo scalar that transforms differently on a reflection when compared to the pressure $p$, a true scalar. Clearly, one can’t have an equality that holds good only in a particular coordinate system, and not in another obtained from the given system by a reflection transformation. Therefore, $C = 0,$
and a rotating particle does not give rise to a pressure field. In turn, this implies \( \mathbf{u}' = 0 \) and \( \mathbf{u} = \mathbf{u}^H \). Using the same arguments as those used above for \( p \), one obtains that

\[
\mathbf{u} = C' \frac{\Omega \wedge \mathbf{x}}{r^3} \tag{35}
\]

In (35), the cross product of two vectors, one of which is a pseudo vector (\( \Omega \)), gives rise to a true vector (\( \mathbf{u} \)), and (35) is therefore a valid invariant relation between two vectors for any value of \( C' \). Since (35) satisfies the equation of continuity, \( C' \) must determined from the surface boundary condition (\( r = 1 \)). Now, \( \mathbf{u} = \Omega \wedge \mathbf{n} \) on the surface of the rotating sphere, where \( \mathbf{n} \) is the outward the unit normal. Using \( \mathbf{x} = \mathbf{n} \) and \( r = 1 \) in (35), one obtains \( C' = 1 \). Thus, the velocity disturbance due to a sphere rotating in an unbounded fluid medium at zero \( Re \) is given by

\[
\mathbf{u} = \Omega \wedge r \frac{1}{r^3}, \tag{36}
\]

which gives rise to circular streamlines in a plane perpendicular to the axis of rotation (see figure 8). Using (36), one may calculate the stress tensor as:

\[
\sigma_{il} = -p \delta_{il} + \left( \frac{\partial u_i}{\partial x_l} + \frac{\partial u_l}{\partial x_i} \right), \tag{37}
\]

\[
= \left( \epsilon_{ijk} \Omega_j \delta_{il} + \epsilon_{ijl} \Omega_j \delta_{ik} \right) - \frac{3(\epsilon_{ijk} x_l x_k \Omega_j + \epsilon_{ijl} x_k \Omega_j)}{r^5}, \tag{38}
\]

\[
= \frac{1}{r^5} \left( \epsilon_{ijl} + \epsilon_{jil} \right) \Omega_j - \frac{3(\epsilon_{ijk} x_l x_k \Omega_j + \epsilon_{ijl} x_k \Omega_j)}{r^5}, \tag{39}
\]

\[
= -\frac{3}{r^5} \left( \epsilon_{ijk} x_l x_k \Omega_j + \epsilon_{ijl} x_k \Omega_j \right), \tag{40}
\]

where we have used the antisymmetry of \( \epsilon_{ijk} \) with respect to an odd number of permutations of its indices. Finally, (40) is used to calculate the torque on the rotating sphere.

\[
\mathcal{L} = \int S_p \mathbf{x} \wedge (\mathbf{\sigma} \cdot \mathbf{n}) dS, \tag{41}
\]

\[
= \int S_p \epsilon_{ijk} x_j \sigma_{kl} n_l dS, \tag{42}
\]

where \( S_p \) is the surface of the rotating sphere. Using \( r = 1 \) and \( x_i = n_i \) on the surface of the sphere \( S_p \), and that \( dS = (1)^2 d\Omega, d\Omega \) being the integral over the solid angle, one has

\[
\mathcal{L}_i = \int S_p \left[-3\epsilon_{ijk} n_j n_a (\epsilon_{klm} n_m n_a \Omega_l + \epsilon_{alm} n_k n_m \Omega_l)\right] d\Omega, \tag{43}
\]

\[
= -3 \int S_p (\epsilon_{ijk} \epsilon_{klm}) n_j n_m (n_a n_a) \Omega_l d\Omega. \tag{44}
\]

The second term in (43) reduces to zero since \( \epsilon_{alm} n_a n_m = 0 \), this being the contraction of an antisymmetric tensor with a symmetric one. Using \( n_a n_a = 1 \), and that \( \epsilon_{ijk} \epsilon_{klm} = (\delta_{il} \delta_{jm} - \delta_{im} \delta_{jl}) \), one obtains

\[
\mathcal{L}_i = -3\Omega_l \int S_p n_j n_m (\delta_{il} \delta_{jm} - \delta_{im} \delta_{jl}) d\Omega, \tag{45}
\]

\[
= -\Omega_i \int d\Omega + 3\Omega_j \int n_j n_l d\Omega, \tag{46}
\]

\[
= -8\pi \Omega_i, \tag{47}
\]

where we have used \( \int n_j n_l d\Omega = \frac{4\pi}{3} \delta_{ij} \). In dimensional terms, the fluid exerts a torque \( \mathcal{L} = -8\pi \mu a^3 \Omega \) on the rotating sphere.
For a sphere translating with velocity $U$ in an unbounded fluid at zero $Re$, the problem, in dimensionless terms, may be stated as follows:

$$\nabla^2 u - \nabla p = 0,$$

(48)

$$\nabla \cdot u = 0,$$

(49)

with the boundary conditions

$$u = U \quad \text{at} \quad r = 1,$$

(50)

$$u, p \to 0 \quad \text{for} \quad r \to \infty.$$  

(51)

A calculation exactly along the lines of the previous subsection gives the following expression for the disturbance velocity field due to a translating particle is given by

$$u_i = \frac{3}{4} U_j \left( \frac{\delta_{ij}}{r} + \frac{x_i x_j}{r^3} \right) + \frac{3}{4} U_j \left( \frac{\delta_{ij}}{3r^3} - \frac{x_i x_j}{r^5} \right),$$

(52)

and leads to a fore-aft symmetric streamline pattern (see figure 9). Similar to the case for the rotating sphere, one may use (52) to determine the stress tensor $\sigma_{ia}$ at $r = 1$, and then find the force on the translating particle by evaluating the surface integral $\int_{S_p} \sigma_{ia} n_a dS$.

However, for a translating particle, there is an easier way to evaluate the force that is more insightful, and involves considerably less algebra. For this purpose, one first shows that the force exerted on any fluid surface $S'$ enclosing the translating sphere equals the force $F$ on the translating sphere. This may be seen by applying the divergence theorem to obtain:

$$\int_{S_p} \sigma \cdot n dS = - \int_{V} \nabla \cdot \sigma + \int_{S'} \sigma \cdot n dS,$$

(53)

$$\Rightarrow \int_{S_p} \sigma \cdot n dS = \int_{S'} \sigma \cdot n dS,$$

(54)

since $\nabla \cdot \sigma = 0$ in Stokes flow. Now, taking $S'$ to be a spherical surface concentric with the sphere, and with $R \to \infty$, one obtains

$$F = \lim_{R \to \infty} \int_{S_R} \left[ -p n + (\nabla u + \nabla u^t) \cdot n \right] dS.$$  

(55)

Since $dS = R^2 d\Omega$, the terms in (52) that are $O(1/R^3)$ give terms in $\nabla u$ that are $O(1/R^4)$, and correspond to vanishingly small contributions in the limit of large $R$. Thus, one need only consider the first term in $\nabla u$.

Physically, this arises because the pressure field generated in a purely azimuthal flow field is on account of centrifugal forces. The latter are inertial in nature, and therefore absent at $Re = 0$. 

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**Figure 9**: Single particle translating in an infinite quiescent fluid at zero $Re$. 

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12Physically, this arises because the pressure field generated in a purely azimuthal flow field is on account of centrifugal forces. The latter are inertial in nature, and therefore absent at $Re = 0$. 

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Figure 10: Single particle immersed in an ambient linear flow at zero Re.

(52), when evaluating (55). It may then be shown that, on $S_R$, one has the following:

$$-p n_i = \frac{3(U_k n_k)n_i}{2R^2},$$

$$\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right)n_j = \frac{3}{2R^2}U_k [n_k \delta_{ij} - 3n_i n_j n_k] n_j,$$

$$= -\frac{3(U_k n_k)n_i}{R^2}.$$  

Using these in (55), obtains $F_i = -6\pi U_i$, or in dimensional terms, the well-known Stokes drag $F = -6\pi \mu a U$. It is only the $O(1/r)$ portion of the velocity field given by (52) that leads to a drag. Moreover, the above analysis also shows that a third of the total drag is due to pressure forces, while the remaining two thirds is due to viscous forces.

In potential flow ($Re = \infty$), a translating sphere acts as a source-sink doublet, acting to push out the fluid in front of it, and pull in the fluid behind it, leading to a velocity disturbance that decays as $O(1/r^3)$. On the other hand, at zero $Re$, the dominant effect is that of the sphere dragging the surrounding fluid along with it as it translates, leading to a velocity disturbance that is $O(1/r)$, and therefore, that decays much more slowly. Both the momentum and the kinetic energy of the fluid in this case are infinite. That the velocity field due to the translating sphere must decay as $O(1/r)$ may be seen as follows: At zero $Re$, it is easy to show from the Stoke equations, that there is no net force acting on any volume of fluid that does not enclose a solid surface. Now, taking this volume of fluid to be a large spherical annulus surrounding the sphere, we observe that for the net force on this volume of fluid to be exactly zero, the force on the outer surface of the annulus must balance the force exerted by the sphere on the inner surface. Since, the area of the outer surface increases as $O(r^2)$, the viscous stresses must decay as $O(1/r^2)$, so that the net force on the outer surface is $O(1/r^2)O(1/r^2)$, and therefore a constant that balances the force exerted by the sphere. Now, the viscous stresses are proportional to the velocity gradient, which immediately implies that the velocity field itself must be $O(1/r)$.

### 4.3.3 Single sphere in an ambient linear flow at zero Re

In a manner similar to the above problems, it may shown that the velocity disturbance due to a neutrally buoyant (and, thence, force-free) sphere immersed in a linear flow, $\mathbf{u}^\infty = \mathbf{\Gamma} \cdot \mathbf{x}$, is given by

$$u_i = \Gamma_{ij}x_j - \left[1 + \frac{a^2}{10} \frac{\partial^2}{\partial x_i^2}\right] \left(\frac{E_{ij}x_j}{r^3} - \frac{3(E_{jk}x_j x_k)x_i}{r^3}\right).$$

In (59), $\mathbf{\Gamma}$ is the transpose of the velocity gradient tensor. Note that the velocity disturbance due to the particle only depends on the symmetric part of $\mathbf{\Gamma}$, viz. the rate of strain tensor $\mathbf{E}$. Physically, the particle rotates at an angular velocity that is commensurate with the vorticity in the ambient linear flow; thus, the antisymmetric portion of $\mathbf{\Gamma}$, which is proportional to the ambient vorticity, doesn’t induce a velocity disturbance. On the other hand, the rate of strain in the ambient flow tends to deform the particle as shown in the figure. The solid particle, of course, doesn’t deform, but instead generates a velocity disturbance.
field that acts to oppose the ambient straining (see figure 10). Note that, at large distances, the velocity disturbance takes the limiting form

$$\lim_{r \to \infty} u_i \approx \Gamma_{ij} x_j + \left( \frac{3x_i (x_j x_k E_{jk})}{r^3} - \frac{E_{ij} x_j}{r^3} \right),$$

(60)

and is therefore $O(1/r^2)$. This is because, at large distances, the effect of the particle, in resisting the ambient straining motion, is that of a symmetric force dipole, that is, a pair of equal and opposite forces. Thus, the $O(1/r)$ disturbance velocity fields due to each of the forces cancel out, leading to the $O(1/r^2)$ decay for large $r$. The symmetric force dipole in the above problem gives rise to a non-zero symmetric first moment of the force distribution, $S_{ij}$. It will be seen later in section 4.5 that the latter is an important quantity with regard to the rheology of a dilute suspension. It is given by the expression

$$S_{ij} = \frac{1}{2} \int_{S_p} \left[ x_i (\sigma_{jk} n_k) + x_j (\sigma_{ik} n_k) - \frac{2}{3} \delta_{ij} (\sigma_{lm} x_l n_m) \right] dS.$$

Using (59), it may be shown that

$$S_{ij} = \frac{20 \pi}{3} \mu a^3 E_{ij}.$$

(61)

in dimensional form.

### 4.3.4 Unified matrix representation of translation, rotation and stress problems for a single particle

It was seen in the earlier section that the nature of the velocity disturbance due to a spherical particle depends on the nature of its motion, and that of the ambient flow. Thus, a particle translating in a quiescent fluid exerts a force, producing a velocity disturbance field that decays as $1/r$ at large distances. An example is a heavy particle sedimenting in an ambient fluid medium; in this case, the net drag force (Stokes drag) exerted by the fluid must balance the weight of the particle, giving us the familiar expression for the terminal settling velocity of a particle at zero $Re$. On the other hand, a particle rotating in a quiescent fluid exerts a torque, producing an $O(1/r^2)$ velocity disturbance field. Clearly, a density matched particle will not translate relative to the suspending fluid medium; thus, it will not exert a force. On the other hand, if the ambient fluid is not quiescent, or in a state of uniform translation, then a linear flow is a good approximation for the fluid flow on the scale of the (small) density matched particle. The previous section demonstrated that the particle in this situation would act as a force-dipole, and thereby generate an $O(1/r^2)$ velocity disturbance. It will be seen that this induced force-dipole contributes to the stress in a dilute suspension.

The force, torque and stress problems for a spherical particle in Stokes flow above may be conveniently represented in a matrix formulation [13]:

$$\begin{bmatrix} F \\ L \\ S \end{bmatrix} = - \begin{bmatrix} 6\pi \mu a I & 0 & 0 \\ 0 & 8\pi \mu a^3 I & 0 \\ 0 & 0 & \frac{20 \pi}{3} \mu a^3 \tilde{\Pi} \end{bmatrix} \begin{bmatrix} U \\ \Omega \\ -E \end{bmatrix},$$

(62)

where the vector on the left may be regarded as the generalized force vector, and that on the right as the generalized velocity vector. The matrix relating the two vectors is known as the resistance matrix, and as may be seen in (62), is diagonal for a spherical particle; note that the matrix element relating the two second-order tensors, viz. the stresslet and the rate of strain, has been written in its most general form, a fourth order tensor $\tilde{\Pi} \equiv \delta_{ij} \delta_{kl}$. In the general case, where one has more than one particle, or even a single non-spherical particle, the matrix is not necessarily diagonal. The off-diagonal components act, for instance, to couple translational and rotational degrees of freedom; as will be seen below, this is the case even for a pair of particles. This translation-rotation coupling has already been encountered in the context of the bacterium $E. Coli$ (see section 4.1.2), where the rotation of the helical flagellar bundle also induces a translational motion. Evidently, the resistance matrix for a helix, or an object with a handedness in general, will have off-diagonal terms. The general form of (62) is consistent with the linearity of Stokes equations, and is, in fact, valid for any particulate Stokes flow. The resistance matrix characterizes the linear response
of the system to an external forcing, and contains the entire hydrodynamic information related to the specific problem; thus, in the general case, it is a function of all the configuration degrees of freedom of the system - for instance, the positions of all particles in a suspension.

Equation (62) suggests, in particular, that one may determine the forces and torques from known values of the translational and angular velocities of (say) all the particles in a Stokesian suspension. This is known as the Resistance problem. It is often the case, however, that the forces and torques on the particles are known apriori (for instance, in sedimentation, the force on each particle is that due to gravity), and it is required to determine the resulting motion of the particles, and hence, the evolution of the suspension microstructure. This is the Mobility problem. For a single spherical particle, the mobility formulation is trivially related to the resistance formulation, the elements of the diagonal mobility matrix being reciprocals of the corresponding resistance matrix elements. Thus,

$$
\begin{bmatrix}
U \\
\Omega \\
S
\end{bmatrix} = - \begin{bmatrix}
\frac{1}{6\pi \mu a} & 0 & 0 \\
0 & \frac{1}{8\pi \mu a} & 0 \\
0 & 0 & \frac{1}{2\pi \mu a^3}
\end{bmatrix} \cdot \begin{bmatrix}
F \\
L \\
-E
\end{bmatrix}.
$$

(63)

However, the aforementioned element to element correspondence between the resistance and mobility matrices is no longer true for a system of many particles, and this gives rise to profound differences in the physics governing the resistance and mobility scenarios.

### 4.4 Boundary Integral formulation and the multipole expansion

We have examined three canonical Stokes flow examples where a particle acts to exert a force, a torque and a force dipole, respectively. In the general case, a solid particle immersed in an ambient flow acts as a surface distribution of forces. Thus, the velocity field in presence of the particle is given by

$$
u(x) = \nu^\infty(x) + \int_{S_p} G(x, x_0) \cdot f(x_0) dS(x_0),
$$

(64)

where

$$
G(x, x_0) = \frac{1}{8\pi} \left[ \begin{array}{c}
I + \frac{(x - x_0)(x - x_0)}{r^3}
\end{array} \right],
$$

(65)

with \( r = |x - x_0| \), and as will be seen below, is the velocity disturbance due to a point force acting at the point \( x_0 \) on the surface of the particle. In (64), \( f(x_0) \) represents the force density induced on the surface \( S_p \) of the particle, and \( \nu^\infty(x) \) is the ambient flow. Thus, if one knows the motion of the particle, say, its translational and angular velocities, then using \( \nu(x) = U + \Omega \times x \) for \( x \in S_p \), (64) gives us an integral equation (of the first kind) for the unknown force distribution.

For large distances (\( r = |x| \gg 1 \)), equation (64) may be expanded in the form of a moment hierarchy, and the resulting expansion takes on an intuitive asymptotic form known as the multipole expansion. For a spherical particle, this may be seen by Taylor expanding the kernel of the integral in (64) about the particle centre \( x_c \). Thus,

$$
\lim_{|x| \to \infty} \nu(x) = \nu^\infty(x) + \int_{S_p} [G(x, x_c) + (x_0 - x_c) \cdot \nabla G(x, x_c)] \cdot f(x_0) dS(x_0) + \ldots,
$$

(66)

$$
= \nu^\infty(x) + G(x, x_c) \cdot \int f(x_0) dS(x_0) + \nabla G(x, x_c) : \int (x_0 - x_c) f(x_0) dS(x_0) + \ldots
$$

(67)

Further, separating the second term in (67) into symmetric and antisymmetric parts, and using index notation, one finally obtains

$$
\lim_{|x| \to \infty} u_i(x) = u_i^\infty(x) + G_{ij}(x, x_c) F_j + \frac{1}{2} \left( \frac{\partial}{\partial x_k} G_{ij} + \frac{\partial}{\partial x_j} G_{ik} \right) S_{jk} + \frac{1}{4} \epsilon_{kjl} \left( \frac{\partial}{\partial x_k} G_{ij} - \frac{\partial}{\partial x_j} G_{ik} \right) L_l
$$

(68)


\textsuperscript{13}The expression for the velocity field induced by a drop is more complicated, and in the general case, includes a surface distribution of dipoles too.
with a dipole, respectively, and are termed as the Stokeslet. In a similar manner, the velocity fields (73) and (74) correspond to a point torque and a symmetric point rotlet, respectively, and can therefore only be balanced by a singular viscous stress. This implies that a force-free (density matched) particle generates a smaller force dipole. Again, this is consistent with a problem analyzed in an earlier section, that of a spherical particle immersed in an ambient linear flow. Therefore, at distances much larger than its size, any particle, of the force dipole, the former giving rise to a stress and the latter being equivalent to a torque, and so on.

\[ F_i = \int_{S_p} f_i(x_0) dS(x_0), \]  
\[ S_{jk} = \int_{S_p} \left[ \hat{x}_{ik} f_j(x_0) + \hat{x}_{ij} f_k(x_0) - \frac{2}{3} \delta_{jk} \{ \hat{x}_{0l} f_l(x_0) \} \right] dS(x_0), \]  
\[ \mathcal{L}_i = \int_{S_p} \epsilon_{ijk} \hat{x}_{0j} f_k(x_0) dS(x_0). \]

Here, \( \hat{x}_{0} = x_0 - x_c \); \( F_i \) and \( \mathcal{L}_i \) are the total force and torque on the particle, the latter being evaluated about the point \( x_c \). \( S_{jk} \) is the symmetric and traceless first moment of the surface force distribution; this latter quantity, seen earlier in section 4.3.3, is known as the stresslet. The corresponding velocity fields are determined by the Green’s function \( G(x, x_c) \), and its symmetric and antisymmetric derivatives. Using (65), one obtains

\[ G_{ij}(x) = \frac{1}{8\pi} \left( \frac{\delta_{ij}}{r} + \frac{x_i x_j}{r^3} \right), \]
\[ \left( \frac{\partial}{\partial x_k} G_{ij} + \frac{\partial}{\partial x_j} G_{ik} \right) = \frac{1}{4\pi} \left( \frac{x_i \delta_{jk} - 3 x_i x_j x_k}{r^3} \right), \]
\[ \epsilon_{kilm} \left( \frac{\partial}{\partial x_k} G_{ij} - \frac{\partial}{\partial x_j} G_{ik} \right) = \frac{1}{4\pi} \epsilon_{jml} x_m. \]

Equations (72) and (74) are consistent with the behavior already encountered earlier in the context of a spherical particle. The disturbance velocity fields given by (72) and (74) are \( O(1/r) \) and \( O(1/r^2) \) and approximate the far-field behavior of the velocity field due to a rotating and translating spherical particle, respectively. The velocity field given by (73) is also \( O(1/r^2) \), and corresponds to the far-field behavior of a force dipole. Again, this is consistent with a problem analyzed in an earlier section, that of a spherical particle immersed in an ambient linear flow. Therefore, at distances much larger than its size, any particle, at leading order, generates an \( O(1/r) \) velocity disturbance corresponding to the total force acting on it, and at the next order, generates \( O(1/r^2) \) velocity disturbances due to the symmetric and antisymmetric parts of the force dipole, the former giving rise to a stress and the latter being equivalent to a torque, and so on. This implies that a force-free (density matched) particle generates a smaller \( O(1/r^2) \) velocity disturbance field at leading order, when compared to a particle acted on by a net force.

The velocity fields (72), (73) and (74) are, in fact, singular solutions of the Stokes equations, the singularity being at the origin, \( r = 0 \) (see figure 11). The velocity field (72) corresponds to that produced by a point force or a Stokeslet. For a force \( F \), this velocity field is given by

\[ u_i^F(x) = \frac{F_i}{8\pi} \left( \frac{\delta_{ij}}{r} + \frac{x_i x_j}{r^3} \right), \]

and satisfies the equations

\[ -\nabla^2 u_i^F + \frac{\partial p^F}{\partial x_i} = F_i \delta(x). \]

It may be obtained from the velocity field to a translating sphere, (52), by taking the limit \( a \to 0 \), \( U \to \infty \) with \( aU \) being finite. In this limit, the \( O(1/r^3) \) terms in (52) become asymptotically small, and (52) reduces to (75) with \( F_j = 6\pi U_j \). The singularity arises in this case because a force of finite magnitude is exerted on a vanishingly small fluid region (a point), and can therefore only be balanced by a singular viscous stress. In a similar manner, the velocity fields (73) and (74) correspond to a point torque and a symmetric point dipole, respectively, and are termed as the stresslet and the rotlet. The former, for instance, can again be related to the disturbance velocity field generated by a sphere in an ambient linear flow in the limiting case where \( a \to 0 \), \( E = |E_{ij}| \to \infty \) with \( a^3 E \) being finite.
Figure 11: The figure depicts the flow patterns associated with the Stokeslet, the stresslet and the rotlet - the fundamental singularities of the Stokes equations.

Figure 12: The figure shows the simple shear flow of a dilute suspension of neutrally buoyant spherical particles, and the response of a single such particle to the ambient shear.

4.5 Rheology of a dilute Stokesian suspension of spherical particles at $O(\phi)$

The response of a single neutrally spherical particle to an imposed linear flow field (section 4.3.3) allows one to determine the rheology of a dilute Stokesian suspension of spherical particles to $O(\phi)$, where $\phi$ is the volume fraction of the particulate phase. At this order of approximation, the suspension is regarded as so dilute that interactions between particles may be neglected. It will then be found that the rheology is Newtonian, and is entirely characterized by an effective viscosity that differs by $O(\phi)$ from the viscosity of the suspending fluid.

However, before we move on to calculate the shear viscosity of a dilute suspension, there is a need to clarify the sense in which these rheological properties are defined in the context of a suspension. Recall that the shear viscosity, once determined from the microstructural dynamics, serves as an input in the equations of motion characterizing the flow of suspension on macroscopic scales. In particular, the shear viscosity would characterize the local relationship between stress and the rate of strain fields on the macro-scale, that is, the relation between stress and strain at each point in the flowing suspension. Now, what appears as a ‘point’ on the macro-scale certainly cannot be a point on the micro-structural scale. For a sufficiently dilute suspension, the macrostructural scale is the scale of a single particle, and clearly the notion of a suspension viscosity on this scale is absurd. In fact, in the micro-structural problem, the viscosity is that of the suspending fluid if the point under consideration lies in the fluid domain, and infinite if it lies in the solid. Thus, the ‘point’ on the macro-scale must actually correspond to a microscopic volume that still accommodates a large number of particles. The local relationship on the macro-scale is then the relationship between properties that are averaged over this microscopic volume. Implicit in this picture is the assumption of scale separation, that is, the existence of an intermediate length scale that is much smaller than any macroscopic length scale, but is still large compared to the inter-particle spacing in a Stokesian suspension [14].

Denoting the aforementioned microscopic averaging volume as $V$, we define the locally averaged stress and
strain as:

\[
\langle \sigma \rangle = \frac{1}{V} \int_V \sigma \, dV,
\]

\[
\langle E \rangle = \frac{1}{V} \int_V E \, dV.
\]

Equation (77) may be rewritten as

\[
\langle \sigma \rangle = \frac{1}{V} \int_{V_{\text{fluid}}} (-p I + 2E) \, dV + \frac{1}{V} \int_{V_{\text{particle}}} \sigma \, dV,
\]

where \(V_{\text{fluid}}\) and \(V_{\text{particle}}\) denote the volumes occupied by the fluid and particulate phase, respectively, and we have used the Newtonian relation for the stress in the fluid phase. With the intent of expressing \(\langle \sigma \rangle\) in terms of \(\langle E \rangle\), we rewrite (79) as

\[
\langle \sigma \rangle = -\langle p \rangle I + 2\langle E \rangle + \langle \sigma_p \rangle
\]

where

\[
\langle \sigma_p \rangle = \frac{1}{V} \int_{V_{\text{particle}}} \left[ \sigma - 2E - \frac{1}{3}(\sigma : I)I \right] \, dV,
\]

is the particle contribution to the suspension stress. Note that we have made (81) traceless. The isotropic term, \(\frac{1}{3}(\sigma : I)I\), subtracted in this regard can be accommodated in the suspension pressure \(\langle p \rangle\). Now, the rate of strain within a particle is zero. In addition, neglecting particle inertia, we have \(\nabla \cdot \sigma = 0\) inside the particle. Using this, one may write

\[
\int_{V_{\text{particle}}} \sigma \, dV = \int_{V_{\text{particle}}} \nabla \cdot (\sigma x) \, dV,
\]

\[
= \sum \int_{S_\alpha} (\sigma \cdot n) x \, dS_\alpha
\]

on applying the divergence theorem, where \(S_\alpha\) denotes the surface of particle \(\alpha\) and the summation runs over all the particles in the canonical microscopic volume \(V\). Now, using (83) in (81), and separating out the symmetric (and traceless) and antisymmetric parts, one has

\[
\langle \sigma_p \rangle = \frac{1}{2V} \sum \int_{S_\alpha} [(\sigma \cdot n) x + x(\sigma \cdot n) - \frac{2}{3}(\sigma \cdot n \cdot x)I] \, dS_\alpha
\]

\[
+ \frac{1}{2V} \sum \int_{S_\alpha} [(\sigma \cdot n) x - x(\sigma \cdot n)] \, dS_\alpha.
\]

In the absence of any external torques, the stress must be symmetric; thus, the antisymmetric term in (84) must be identically zero. Now, making the diluteness approximation, the summation in (84) is seen to consist of independent identical contributions from each particle, and the expression for the particle stress simplifies to

\[
\langle \sigma_p \rangle = \frac{N}{2V} \int_{S_p} [(\sigma \cdot n) x + x(\sigma \cdot n) - \frac{2}{3}(\sigma \cdot n \cdot x)I] \, dS.
\]

In dimensionless terms, the volume fraction \(\phi\) of the particulate phase is related to the number density as \(\phi = (N/V)(4/3)\pi\). In addition, we also recognize the integral in (85) to be the stresslet; this was found to be \(\frac{20\pi}{3}E\) in an earlier section (see section 4.3.3). Using these, one finds

\[
\langle \sigma_p \rangle = \frac{3\phi 20\pi}{4\pi} \frac{3}{3} E,
\]

\[
= 5\phi E
\]

Writing \(\langle \sigma_p \rangle = 2\mu_p \langle E \rangle\) (this is possible because \(E\) and \(\langle E \rangle\) differ only by \(O(\phi)\), and this only leads to an error in \(\langle \sigma_p \rangle\) that is \(O(\phi^2)\)), we obtain the particle contribution to the viscosity as \(\mu_p = \frac{5}{2} \phi\), a result first obtained by Einstein in 1905. Therefore, the viscosity of a dilute suspension \(\mu_s\), in dimensional terms, is given by

\[
\mu_s = \mu + \mu_p
\]

\[
= \mu (1 + \frac{5}{2} \phi).
\]
Thus, to $O(\phi)$, the Stokesian suspension behaves as a Newtonian fluid. Since each constituent particle rotates at a rate commensurate with the imposed vorticity, the rheological response of a dilute non-interacting suspension is only dependent on the imposed rate of strain $E$, as one would expect. The particles act to increase the viscosity by a factor proportional to the volume fraction. Each particle responds to the ambient shearing flow by acting to retard the imposed straining motion. This resistance translates to a higher effective viscosity. It must also be mentioned that we have not made the assumption of a simple shearing flow in the above analysis. Indeed, the above rheological response of a dilute suspension at $O(\phi)$ is valid for any linear flow, that is, any imposed flow of the form $u^\infty = \Gamma \cdot x$ (see Appendix for a description of planar linear flows).

An alternative way of looking at the result (89) is to recognize that the particles introduce velocity disturbances in the fluid phase that increase the effective dissipation. This is, in fact, the approach that Einstein adopted in his calculation of the suspension viscosity. The averaged viscous dissipation is given by

$$D = 2\mu \langle e : e \rangle,$$

$$= 2\mu [\langle e \rangle : \langle e \rangle + \langle e' : e' \rangle],$$

where $e'$ is the fluctuation strain rate due to the presence of the particles; thus, $\langle e' \rangle = 0$. The second term may now be evaluated as

$$\langle e' : e' \rangle = n \int \langle e' \rangle(x|x_1)dx,$$

where $n$ is the number density, and $\langle . \rangle(x|x_1)$ denotes an average conditioned on the presence of a single particle at $x_1$. Making an error of $O(\phi^2)$ (in neglecting pair-particle interactions), one may write

$$\langle e' : e' \rangle = n \int \langle e' \rangle(r) dV,$$

where $e'$ now corresponds to the disturbance strain rate in the single particle problem. The integration is over the volumes of both the particle and the surrounding fluid, and therefore,

$$\langle e' : e' \rangle = n \left[ \int_{v_p} \langle e' \rangle(r) dV + \int_{v_f} \langle e' \rangle(r) dV \right].$$

Since the particles are rigid, the rate of strain inside the particle is zero, and we have $e' = -\langle e \rangle$ inside the particle. For the fluid phase, the fluctuation rate of strain may be obtained from (59). On using the expressions for the fluctuation strain appropriate to the fluid and particle phases, and carrying out the volume integrations, one obtains

$$\langle e' : e' \rangle = \phi \langle e \rangle : \langle e \rangle + \frac{3}{2} \phi \langle e \rangle : \langle e \rangle,$$

where the first and the second term denote the contribution of the volume integrals over the fluid and particle phase, respectively. Addition of the two, of course, leads to the Einstein coefficient.

4.6 Motion of particle pairs

4.6.1 Hydrodynamic interactions

A pair of particles interact with each other via the disturbance velocity field that each of them generates in the suspending fluid. The velocity field due to one particle affects the other, and vice versa. These hydrodynamic interactions, as they are called, are long-ranged, because, as already seen, the disturbance velocity fields at zero $Re$ typically exhibit a slow algebraic decay with increasing distance. The effect of

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14The dependence of the suspension stress on the rate of strain tensor rather than the full velocity gradient tensor may be deduced from the principle of material frame indifference, which states that the constitutive relation must be invariant in form to rigid rotations of the reference frame. This principle, however, breaks down in the presence of micro-scale inertia.
Hydrodynamic interactions become increasingly important with decreasing pair separation, and take the form of lubrication interactions when the separation of the particle pair becomes asymptotically small compared to the size of either particle. Lubrication retards both the approach and separation of a pair of closely spaced particles. This may be understood from figure 14, where, in the left figure, it is shown that in order for two particles to approach each other, the fluid in the intervening gap must be pushed out. Now, a large pressure gradient in order to establish a local Poiseuille flow in the radially outward direction against the resisting viscous stresses. Since the pressure outside the gap is approximately equal to that in a quiescent fluid, this results in the build up of a very large pressure inside the thin gap, which then acts to resist the approach of the two spheres. It may be shown that the pressure inside the gap scales inversely as the minimum separation between the sphere surfaces, and in turn, this leads to the prediction that two perfectly smooth spherical surfaces will touch each other only in the limit of infinite time. It is equally difficult to separate two spheres that nearly touch each other. In this case, the direction of the Poiseuille flow is reversed, since fluid rushes in from outside to fill in the gap left between the receding spheres. This therefore leads to the build up of a large negative pressure in the gap, relative to the ambient, that again acts to retard the separation of the particle pair (see right illustration in figure 14). Thus, lubrication interactions often lead to the preponderance of closed spaced particle pairs in a Stokesian suspension, which in turn affects its bulk rheological properties. Lubrication interactions also affect the rate of coagulation in aerosols and hydrosols, and therefore play a key role in determining their stability. However, lubrication may breakdown for the case of rough particles, or due to the onset of non-continuum fluid behavior for the case when the suspending fluid is a gas. Lubrication may also fail for sufficiently massive particles (as in granular flows) which, on account of their inertia, are driven very close together; in this case, the breakdown of lubrication inevitably leads to solid-body collisions.
Outgoing Poiseuille flow generates large positive pressures

Particles contact only at infinite time

Inward Poiseuille flow generates large negative pressures

Particles contact only at infinite time

Figure 14: The figure depicts lubrication interactions between a pair of approaching and receding particles.

4.6.2 Matrix representation for the pair-particle problem

In a manner similar to the single spherical particle, the Stokes flow problem involving a pair of particles may also be written in a compact resistance matrix formulation as follows ([13],[15]):

\[
\begin{pmatrix}
F_1 \\
F_2 \\
L_1 \\
L_2
\end{pmatrix} = - \begin{pmatrix}
R_{FU}^{11} & R_{FU}^{12} & R_{FU}^{1\Omega} & R_{FU}^{22} & R_{FU}^{2\Omega} \\
R_{FU}^{21} & R_{FU}^{22} & R_{FU}^{2\Omega} & R_{FU}^{12} & R_{FU}^{1\Omega} \\
R_{LU}^{11} & R_{LU}^{12} & R_{LU}^{1\Omega} & R_{LU}^{22} & R_{LU}^{2\Omega} \\
R_{LU}^{21} & R_{LU}^{22} & R_{LU}^{2\Omega} & R_{LU}^{12} & R_{LU}^{1\Omega}
\end{pmatrix}\begin{pmatrix}
U_1 \\
U_2 \\
\Omega_1 \\
\Omega_2
\end{pmatrix},
\]

(96)

where the subscripts associated with each element of the resistance matrix indicate the nature of the dynamical coupling, while the superscripts indicate the particles involved. For instance, \( R_{FU}^{12} \) is the force induced on particle 1 that is stationary \((U_1 = \Omega_1 = 0)\) when particle 2 translates with unit velocity \((U_2 = 1)\) without rotating \((\Omega_2 = 0)\). In a similar manner, \( R_{FL}^{21} \) denotes the force on a stationary particle 2 due to particle 1 that does not translate \((U_1 = 0)\), but rotates with a unit angular velocity \((\Omega_1 = 1)\). In presence of an ambient linear flow, \((101)\) is still valid, except that \( U_i \) is now replaced by \((U_i - U_i^\infty) \) \((i = 1, 2)\), and \( \Omega_i \) is replaced by \((\Omega_i - \omega^\infty) \). Here, \( U_i^\infty \) is the velocity of the imposed linear flow at the location of the \( i \)th spherical particle, and \( \omega^\infty \) is the constant ambient vorticity.

With the neglect of hydrodynamic interactions, the coupling between translational and rotational degrees of freedom, characterized above by the elements of \( R_{FL}^{21} \) and \( R_{LU}^{11} \), must vanish, and the diagonal elements, viz. the \( R_{FU}^{11} \)'s and \( R_{LU}^{11} \)'s will be proportional to the identity tensor, as is the case for a single spherical particle (for instance, in the dimensional form, \( R_{LU}^{11} = R_{LU}^{22} = 6\pi\mu a^2 I \), \( R_{LU}^{1\Omega} = R_{LU}^{2\Omega} = 8\pi\mu a^2 I \)). Now, in the pair-particle problem, the non-dimensionalized resistance matrix elements will only be a function of the pair separation vector \( r = x_2 - x_1 \). Therefore, for very large pair separations, the resistance matrix must, at leading order, be diagonal, indicating the vanishingly small hydrodynamic coupling between the two particles.

The physics governing the translation-rotation coupling in the pair-particle problem is illustrated in figure 15 in the limit of large pair separations. For instance, considering the left illustration therein, we observe that particle 2, rotating with angular velocity \( \Omega_2 \), must induce a velocity disturbance, \( u_{12} \sim O(1/r^2) \), at the location of particle 1 which is fixed. This leads to a force proportional to the velocity disturbance, and therefore also of \( O(1/r^2) \), on particle 1. Thus, one expects that \( R_{FU}^{12} \sim O(1/r^2) \) for \( r \gg 1 \). On the other hand, in the right illustration, we observe that particle 2 translating with velocity \( U_2 \) produces a velocity disturbance of \( O(1/r) \) in the otherwise quiescent fluid. The resulting vorticity at the location of particle 1 is \( O(1/r^2) \), being proportional to the local velocity gradient. In turn, this leads to a torque of \( O(1/r^2) \) on the fixed particle 1; thus, in the limit \( r \gg 1 \), one also has \( R_{LU}^{1\Omega} \sim O(1/r^2) \). Similar estimates may be obtained.
for other resistance elements by considering a particle-pair problem with appropriate restrictions. Below, we use analogous arguments to estimate the strengths of hydrodynamic interactions in the resistance and mobility scenarios in dilute systems.

We now compare the nature of hydrodynamic interactions in the resistance and mobility problems. In order to illustrate the differing physics, it is sufficient to examine the diagonal elements of the relevant hydrodynamic matrix viz. $R_{LL}^{11}$ for the resistance problem, and $M_{LU}^{11}$ for the mobility problem. The former is determined by considering a system of particles wherein particle 1 moves with a fixed velocity $U_1 = 1$, while the other particles are held fixed. $R_{LL}^{11}$ then determines the force on particle 1. At leading order, for a dilute system of particles, this force is, of course, just the Stokes drag, and therefore, at leading order, $R_{LL}^{11} \sim (6\pi\mu a)$. The leading order correction to the Stokes drag term may be estimated as follows. A translating particle 1 generates an $O(1/r)$ velocity disturbance that induces a force of $O(1/r)$ on a second fixed particle, particle 2 (say). This induced force, in turn, generates an $O(1/r^2)$ velocity disturbance that exerts an $O(1/r^2)$ force on particle 1. Thus, the correction to the Stokes drag is $O(1/r^2)$; in other words, $R_{LL}^{11} \sim 6\pi\mu a [1 + O(1/r^2)]$ (see the left illustration in figure 17).

On the other hand, considering the mobility problem, $M_{LU}^{11}$ determines the velocity of particle 1 due to a unit force on particle 1 when all other particles are force free. Clearly, particle 1’s velocity, at leading order, is $1/(6\pi\mu a)$, using the Stokes relation. The leading order correction, as a function of the inter-particle separation, may now be estimated. The force on particle 1 produces an $O(1/r)$ velocity disturbance at the location of particle 2. However, unlike the resistance scenario, particle 2 is force-free. Thus the $O(1/r)$ velocity disturbance cannot elicit a net force, but instead acts to induce a force dipole in particle 2. The strength of this force dipole is proportional to the local velocity gradient, and is therefore $O(1/r^2)$. This dipole in turn generates an $O(1/r^2).1/r^2 \approx O(1/r^4)$ velocity field that then changes the velocity of particle 1 by $O(1/r^4)$. Thus, $M_{LU}^{11} \sim (6\pi\mu a)^{-1}[1 + O(1/r^4)]$ (see the right illustration in figure 17). The much faster decay with $r$ of the leading order interaction term when compared with the resistance scenario ($O(1/r^2)$ compared to $O(1/r^4)$), clearly suggests the weaker nature of hydrodynamic interactions in the mobility scenario. Sedimenting Stokesian suspensions as well as suspensions of neutrally buoyant particles are examples of systems characterized by mobility-type hydrodynamic interactions between particles. In the former case, the force on each particle is fixed, being that due to gravity. Hydrodynamic interactions in this system act to retard the settling velocity of a heavy suspension, leading to the well known phenomenon of hindered settling; figure 16 shows the variation of the sedimenting velocity as a function of volume fraction. For a sheared neutrally buoyant suspension, all particles are force-free, and hydrodynamic interactions act to enhance the suspension shear viscosity; the $O(\phi^2)$ correction to the shear viscosity, due to pair-hydrodynamic interactions, is analyzed in the next section.

Returning to hydrodynamic interactions in the resistance problem, we recall that that a given particle (the
‘test’ particle) moving with velocity $U$ produces a velocity disturbance of $O(Ua/r)$ for $r \gg a$. In turn, this induces a force of $O(6\pi\eta U(a/r))$ on each neighboring particle, and in a direction opposing the force on the test particle. Assuming the system of particles to have a uniform number density ($n$), if one adds up the induced forces on all particles surrounding the ‘test’ particle in a spherical volume of radius $R$, the total induced force in this spherical volume is $O(n6\pi\eta U(a/R^2)R^3)$, and therefore increases as $R^2$. For $R \sim 1/(an)^{2} \sim a\phi^{-\frac{1}{2}}$, the induced force field becomes comparable in magnitude to the force on the test particle 1. In other words, the particles (with fixed velocities) surrounding the test particle act as sinks for the momentum associated with the generated disturbance velocity field, and thereby act to screen the force on the test particle on a length scale of $O(a\phi^{-\frac{1}{2}})$; note that, in the limit $\phi \ll 1$, $a\phi^{-\frac{1}{2}} \gg 1$. This length scale is known as the screening length, and the estimate $R_{11} \propto 1 + O(1/r^2)$ made above is, in fact, only valid for distances less than this screening length, since it was based on solutions of the Stokes equations. For distances greater than screening length, the system behaves as a porous medium rather than as a Stokesian fluid, and the velocity disturbance due to a given particle decays faster, being $O(1/r^3)$ for $r \gg a\phi^{-\frac{1}{2}}$. On length scales larger than the screening length, particles interact via screened resistance interactions. Hydrodynamic interactions of this nature occur in gas-solid suspensions. As discussed earlier, in the context of Figure 2 particle inertia is dominant in these systems, while the fluid inertial forces are small. Thus, the fluid still satisfies Stokes equations, and responds instantaneously to the motion of the particles. On the other hand, since the inertia of a particle is so large that the velocity disturbance due to a neighboring particle will not lead to a change in its velocity, but instead acts to induce a force [16]. These induced forces then lead to the aforementioned screening phenomenon (see figure 17). Screened interactions are an important ingredient in the calculation of the viscous dissipation in a gas-solid suspension as a function of the volume fraction. In turn, the viscous dissipation, together with the pressure of the particulate phase, plays a key role in determining the stability of a homogeneous gas-solid fluidized bed.

4.7 Effect of hydrodynamic interactions on the rheology of a dilute suspension

Having gained an appreciation of the importance of hydrodynamic interactions in Stokesian suspensions, we are now in a position to comment on their role in determining the rheology of a Stokesian suspension. Recall that the force-free torque-free particles in a dilute Stokesian suspension enhance the viscosity at $O(\phi^2)$ in the absence of interactions. The next contribution to the rheology of a dilute Stokesian suspension comes from hydrodynamic interactions.

In the limit $\phi \ll 1$, the probability of finding a pair of particles within a region of the order of their own size (that is, in a region where they can interact hydrodynamically) is $O(\phi^2)$, while the probability of a three-particle interaction is asymptotically smaller, being $O(\phi^3)$. Thus, one expects the next contribution to the stress in a dilute suspension to be $O(\phi^2)$, and to be determined by pair-hydrodynamic interactions. The motion of a pair of particles in an imposed linear flow at zero $Re$ is well characterized, as implied by the analysis in earlier sections. In the matrix formulation, the motion of a pair of force-free torque-free particles

![Figure 16: The plot shows variation of the hindered settling velocity with volume fraction.](image-url)
Constant force (Mobility) interactions

\[ U_2 = 0 \]

Constant velocity (Resistance) interactions

- Resistance Interactions are strong enough to lead to screening effects

Characteristic length scale - \( a^{-\frac{1}{2}} \)

\[ F_s \sim O(1/r) \rightarrow R_{FU}^{21} \sim O(1/r) \]

\[ R_{FU}^{11} \sim 6 \pi \eta a + O(1/r^2) \]

\[ U_2 = 0 \]

\[ F_s \sim O(1/r^2) \]

\[ F_2 \sim 0 \]

\[ M_{UF}^{11} \sim (6 \pi \eta a)^{-1} + O(1/r^4) \]

\[ O(1/r) \]

\[ O(1/r^2) \]

\[ O(1/r^3) \]

\[ O(1/r^4) \]

\[ M_{UF}^{11} \]

\[ O(1/r^3) \]

\[ O(1/r^4) \]

\[ \eta \]

\[ \pi \]

\[ a \]

Figure 17: The figure depicts the differing nature of hydrodynamic interactions in the resistance and mobility scenarios.
may be written as:

\[
\begin{bmatrix}
0 \\
0 \\
\mathbf{S}
\end{bmatrix} = - \begin{bmatrix}
R_{FU} & R_{F\Omega} & R_{F\mathbf{E}} \\
R_{LU} & R_{L\Omega} & R_{L\mathbf{E}} \\
R_{SU} & R_{S\Omega} & R_{S\mathbf{E}}
\end{bmatrix} \cdot \begin{bmatrix}
\mathbf{U} - \Gamma \cdot \mathbf{x} \\
\mathbf{U} - \Omega \cdot \mathbf{x} \\
- \mathbf{E}
\end{bmatrix},
\]

where \( \mathbf{u}^\infty = \Gamma \cdot \mathbf{x} \) is the imposed linear flow, and we have used

\[
R_{FU} = \begin{bmatrix}
R_{FU}^{11} & R_{FU}^{12} \\
R_{FU}^{21} & R_{FU}^{22}
\end{bmatrix}
\]

and so on, in the interest of brevity. The expressions for the resistance tensors that appear in (97) are well known \[13\].

However, one still needs to know the frequency of occurrence of various pair-particle configurations in order to be able to average over these configurations (the microstructural average), and determine the relevant bulk rheological characteristic. In this context, we define the pair distribution function \( g(\mathbf{r}) \), where \( \phi g(\mathbf{r}) \) is of the probability of finding a second particle at \( \mathbf{r} \) given a test particle at the origin. Clearly, in the absence of any hydrodynamic correlation between a pair of particles, one expects that \( g(\mathbf{r}) \) will equal unity for any \( r \) greater than \( 2a \). The problem of determining the \( O(\phi^2) \) rheology of a dilute suspension then reduces to determining \( g(\mathbf{r}) \), that then completely characterizes the microstructure at the pair-particle level.

It may be shown that the pair-distribution is governed by a convection equation \[17\], as must be the case in the hydrodynamic limit (\( Pe \rightarrow \infty \)):

\[
\frac{\partial g}{\partial t} + \mathbf{V} \cdot (\nabla g) = 0,
\]

where \( \mathbf{r} = \mathbf{x}_2 - \mathbf{x}_1 \) is the pair separation vector, and \( \mathbf{V} = \mathbf{U}_2 - \mathbf{U}_1 \) is the relative velocity of the particle-pair including hydrodynamic interactions. Thus, in a reference frame moving with particle 1, the pair distribution function \( g \) is convected along the trajectory of particle 2. Rewriting (99) as

\[
\frac{\partial g}{\partial t} + \mathbf{V} \cdot \nabla r g = -g (\nabla \mathbf{r} \cdot \mathbf{V}),
\]

we observe that the change in \( g \), as it is convected along the particle pathline, arises owing to the compressibility of the particle pathlines; that is, unlike fluid streamlines, for the particle pathlines, we have \( \nabla \mathbf{r} \cdot \mathbf{V} \neq 0 \). This then gives rise to a non-uniformity in the pair-probability, and certain pair configurations are preferred over others. Since the value of \( g(\mathbf{r}) \) is, in principle, determined by integrating \( \nabla \mathbf{r} \cdot \mathbf{V} \) along a particle pathline, one needs to start from a suitable initial condition. This initial condition is evident for open pathlines or pathlines that originate from infinity (see the left illustration in figure 18); the initial condition is that of no correlation in the region far upstream where the hydrodynamic interactions are vanishingly small, that is, \( g(\mathbf{r}) \rightarrow 1 \) for \( r \rightarrow \infty \). On the other hand, the presence of closed particle pathlines poses a fundamental problem with regard to the choice of initial condition (the right illustration in 18). Thus, in flows where hydrodynamic interactions lead to closed particle pathlines, the problem of calculating the statistics of the particle-pair configurations, and hence the \( O(\phi^2) \) rheology of a dilute Stokesian suspension is rendered indeterminate.

It turns out that for linear flows with open pair-particle pathlines, \( g(\mathbf{r}) = g(\mathbf{r}) \), that is, the pair distribution function is independent of angular coordinates, or in other words, the microstructure at the pair-level is isotropic \[15\]. This may be shown to lead to a Newtonian viscosity given by \( \mu_s = \mu (1 + \frac{1}{2} \phi + 7.6 \phi^2) \). However, it must be emphasized that the rheology of a Stokesian suspension at \( O(\phi^2) \) is not really Newtonian, since the existence of a viscosity is contingent on the flow comprising entirely of open pathlines at the micro-scale. This peculiar characteristic of the rheological problem at \( O(\phi^2) \) arises due to the absence of an intrinsic relaxation process at the micro-structural level at infinite \( Pe \).

The aforementioned indeterminacy, related to convective effects in presence of closed trajectories, is also encountered in the context of heat or mass transfer from a single particle; this was discussed briefly in \[15\].

The isotropy in itself is quite remarkable considering the strong angular dependence of the relative velocity of the particle-pair.
Figure 18: The figure shows an example of an open and a closed pathline for a pair of particles in an imposed linear flow.

In the single particle problem, as a consequence of the closed streamlines around the rotating particle, increasing the magnitude of $Pe$ has no effect on $Nu$, and the latter asymptotes to an $O(1)$ value for large $Pe$ (for spheres, this value is 4.5 in simple shear flow). Thus, at zero $Re$, the rate of heat transfer, even at infinite $Pe$, is diffusion-limited, being determined by diffusion of heat or mass across the closed streamsurfaces. The indeterminacy in the pair-particle problem may, in principle, be removed in a similar manner. In a manner analogous to the single particle problem, one could therefore incorporate thermal effects, thereby providing a relaxation mechanism. Thus, the effects of a small Brownian diffusion across closed pathlines will eventually lead to a well defined $g(r)$ that is independent of $Pe$, and the resulting suspension viscosity will evidently have an $O(\phi^2)$ coefficient that differs from 7.6 above. Thus, although the formal order of a Brownian diffusivity is $O(1/Pe)$ in the limit $Pe \to \infty$, such a diffusion, acting over an infinite period of time will have an $O(1)$ effect. On the other hand, one could also have the presence of a small amount of fluid inertia, or inertia of the particles themselves, acting to open up the pathlines, and thereby removing the indeterminacy. The resulting rheology in these cases is likely to be non-Newtonian. In any case, it should be clear that the rheology of a suspension of non-Brownian spheres tends to be rather sensitive to the presence of non-hydrodynamic forces.

We may now attempt to give a brief description of the rheology of a Brownian suspension as a function of $Pe$ in simple shear flow. At small $Pe$, Brownian motion is dominant, and the microstructure is nearly isotropic. However, the small departure of the microstructure from isotropy, in presence of an imposed shear, leads to a viscous stress. Note that the Einstein coefficient is unaffected by Brownian motion. At $O(\phi)$, the effect of Brownian motion is only to give rise to an isotropic osmotic pressure proportional to $nkT$, $n$ being the particle number density. Thus, the aforementioned small microstructural anisotropy is that in $g(r)$, and therefore affects the $O(\phi^2)$ contribution to the suspension viscosity on account of pair-hydrodynamic interactions. As $Pe$ increases, the microstructure, on account of Brownian motion, is unable to ‘keep pace’ with the imposed rate of deformation, and this leads to a shear thinning. The increasing microstructural anisotropy also leads to normal stress differences. The extent of shear thinning is a function of $\phi$, since it

---

\[^{16}\text{This is an example where the order of the two limits } t \to \infty \text{ and } Pe \to \infty \text{ is not interchangeable. Taking } Pe \to \infty \text{ first leads to the indeterminate rheology. However, taking the limit of infinite time instead will resolve the rheological indeterminacy at the pair-particle level; although, it must be mentioned that this scenario is a rather tenuous one, and it is quite likely that three-particle interactions would have the same effect on a shorter time scale in a non-dilute suspension.}\]

\[^{17}\text{The contribution resulting from the anisotropic microstructure is viscous in nature, because it results from the product of the large } O(kT/a) \text{ stress associated with Brownian motion, with the small } O(Pe) \text{ anisotropy.}\]
Figure 19: The figure shows the fluid streamlines and the pair-particle pathlines in a sequence of linear flows ranging from irrotational plane extension ($\lambda = 1$) to an elliptic linear flow ($\lambda < 0$). The topology of pair-particle pathlines plays a key role in the bulk rheology of Stokesian suspension.
is determined by the lag of the microstructure with respect to the imposed deformation; in turn, this is a function of the relaxation time $\alpha^2/D(\phi)$, and therefore a function of $\phi$.

For simple shear flow, as already seen, the high $Pe$ limit is not straightforward. A possible scenario, again discussed earlier, is where a vanishingly small Brownian diffusion, acting over an asymptotically long, $O(Pe)$ period of time, serves to determine $g(r)$. However, this scenario is a little unrealistic, since it requires $Pe \ll 1/\phi$, and is thereby restricted to very dilute suspensions. The symmetry properties of the resulting $g(r)$ would, in fact, lead to a viscosity but no normal stress differences. A second scenario appeals to the existence of short-range repulsive forces [18] that then dominate the effects of near-field lubrication interactions. In this case, one finds a thin boundary layer on the compressive quadrants, where the balance of flow and repulsive forces, together with the smoothing effects of Brownian motion, lead to very high values of $g(r)$. There is no such enhancement of the pair-distribution in the extensional quadrants of simple shear flow. The obvious anisotropy in the microstructure then leads to a non-Newtonian rheology (with normal stress differences) that is rather insensitive to $Pe$ in the limit $Pe \rightarrow \infty$. This is a more realistic scenario in non-dilute suspensions, since the establishment of the microstructural anisotropy requires diffusion only across an asymptotically thin boundary layer, and thus may operate even at higher concentrations.

### 4.8 Motion of three or more particles

The motion of three or more particles requires numerical methods. However, the underlying formulation in terms of resistance and mobility matrices remains the same. Thus, for a system of $N$ particles, one may write

$$
\begin{bmatrix}
F_1 \\
F_2 \\
\vdots \\
F_N \\
L_1 \\
L_2 \\
\vdots \\
L_N \\
S_1 \\
S_2 \\
\vdots \\
S_N
\end{bmatrix} = - \begin{bmatrix}
R_{FU}^{11} & \ldots & R_{FU}^{1N} & R_{F\Omega}^{11} & \ldots & R_{F\Omega}^{1N} & R_{FE}^{11} & \ldots & R_{FE}^{1N} \\
\vdots & \ddots & \vdots & \vdots & \ddots & \vdots & \vdots & \ddots & \vdots \\
R_{SU}^{11} & \ldots & R_{SU}^{1N} & R_{S\Omega}^{11} & \ldots & R_{S\Omega}^{1N} & R_{SE}^{11} & \ldots & R_{SE}^{1N}
\end{bmatrix} \begin{bmatrix}
U_1 - \langle U \rangle \\
U_2 - \langle U \rangle \\
\vdots \\
U_N - \langle U \rangle \\
\Omega_1 - \langle \omega \rangle \\
\Omega_2 - \langle \omega \rangle \\
\vdots \\
\Omega_N - \langle \omega \rangle \\
- \langle E \rangle \\
\vdots \\
- \langle E \rangle \\
\end{bmatrix},
$$

or, in a more compact form, as

$$
\begin{bmatrix}
F \\
S
\end{bmatrix} = - \begin{bmatrix}
R_{FU} & R_{FE} \\
R_{SU} & R_{SE}
\end{bmatrix} \begin{bmatrix}
U - \langle U \rangle \\
- \langle E \rangle
\end{bmatrix},
$$

where the translational and rotational degrees of freedom have now been combined, and

$$
\mathcal{R} = \begin{bmatrix}
R_{FU} & R_{FE} \\
R_{SU} & R_{SE}
\end{bmatrix}
$$

is termed the grand resistance matrix. Stokesian dynamics [15] is a simulation technique that exploits the properties of low Reynolds number hydrodynamics, and is extensively employed in simulating Stokesian suspensions. In this method, the far-field hydrodynamic interactions, in their mobility formulation, are treated in a pairwise manner using multipole expansions truncated at the dipole order (see section 4.4). On the other hand, the near-field lubrication interactions are incorporated in their exact analytical form, thereby allowing one to simulate a system of hydrodynamically interacting particles subject to an external flow. The method is computationally intensive, however, and the computational effort scales as $O(N^3)$, $N$ being the number of particles. A recent, more efficient version of this method, ‘Accelerated Stokesian dynamics’, has been developed [19], and the computational effort herein scales as $O(N \ln N)$, allowing one to simulate rather...
large systems of a few hundred particles. The Lattice Boltzmann method offers an alternate scheme to simulate the motion of particles in a viscous fluid, and is usually more efficient for suspensions at non-zero Reynolds numbers \([20]\).

### 4.9 Shear-induced diffusion

Shear-induced diffusion refers to the phenomenon where a non-Brownian particle at zero \(Re\), on account of interactions with its neighbors in a sheared Stokesian suspension, starts to diffuse on time scales long compared to that characterizing a single interaction \([22]\). Using elementary dimensional arguments, it is seen that the shear-induced diffusivity in a non-Brownian system must be \(O(\dot{\gamma}a^2f(\phi))\), where \(\dot{\gamma}\) is a characteristic shear rate, and \(f(\phi)\) denotes an appropriate dependence on the volume fraction. Physically, the diffusion of a particle results from random displacements of \(O(a)\) suffered due to interactions that occur on a time scale of \(O(\dot{\gamma}^{-1}f(\phi)^{-1})\), where \(f(\phi)\) is now a measure the probability of the interaction event. In simple shear flow, one expects that such a diffusive motion in the transverse direction (that is, gradient or vorticity directions) must result in a dilute suspension on account of pair-hydrodynamic interactions, the resulting diffusivity then being \(O(\dot{\gamma}a\phi)\), since the probability of finding a second particle is \(O(\phi)\). However, reversibility dictates that pair-particle pathlines be fore-aft symmetric, and therefore, a given particle does not suffer any net lateral displacement on account of successive uncorrelated pair-interactions. Thus, one is led to conclude that diffusivity in a dilute suspension, in the absence of non-hydrodynamic forces, can only arise due to interactions of three or more particles, and must therefore be \(O(\dot{\gamma}a^2\phi^2)\) in the limit \(\phi \ll 1\).

However, this is not so, since the principle of dynamic reversibility is inconsistent with the notion of diffusion in a Stokesian suspension. This is seen from the following argument. Let us suppose that we have a suspension with a random microstructure, and that we start to shear it in the forward direction (say). As a result, let us that assume all pairs of particles diffuse relative to each other in this forward motion, that is, the mean square separation of each particle pair increases linearly with time. On reversing the shearing motion, one must reverse time in accordance with reversibility. This then implies that all particles must now ‘anti-diffuse’ in what is essentially the same physical scenario. The only way of resolving this apparent contradiction is to realize that the mean square separation between two particles will, on average, not grow in a diffusive manner, implying also that a single particle will, in the absence of non-hydrodynamic forces, not diffuse at zero \(Re\). Thus, in principle, dynamic reversibility is incompatible with shear-induced diffusion.

The phrase ‘in principle’ used in the last sentence of the earlier paragraph is important. This is because the motion of a sheared suspension comprising a large number of particles is almost certainly chaotic owing to the non-linearity of the kinematic equations of motion. The equations characterizing pair-interactions are integrable, but particle trajectories arising from interactions of three or more particles are likely to be sensitive to initial conditions. Thus, the tiniest of disturbances - this may be truncation error in simulations, or ambient noise/vibration in experiments - is enough for the actual paths of particles to depart from their theoretical reversible trajectories at an exponential rate, and thence, lead to diffusion for long times in a bounded system. Thus, while it is true that dynamic reversibility does not allow for diffusive motion at zero \(Re\), the chaotic dynamics underlying the kinematic equations imply that one would need an infinite degree of accuracy in a simulation, for instance, in order not to observe diffusive motion. On account of kinematic irreversibility associated with the equations governing the motion of three or more particles at zero \(Re\), the diffusion coefficients in a dilute Stokesian suspension are indeed \(O(\dot{\gamma}a^2\phi^2)\).

In practice, non-hydrodynamic forces invariably play a role in pair-hydrodynamic interactions, especially during the close approach of particle-pairs. Such forces include, for instance, those arising on account of surface roughness, or short-range repulsive forces. Each of these factors then leads to pair-interactions that are fore-aft asymmetric. The resulting lateral motion on account of a sequence of such uncorrelated interactions therefore leads to a shear-induced diffusivity that is \(O(\dot{\gamma}a^2\phi)\) (see figure 20). Clearly, the shear-induced or hydrodynamic diffusivity varies both with volume fraction and the applied shear rate, and this has important consequences when either of the two fields is inhomogeneous. For instance, in the pipe flow of a sufficiently concentrated suspension, the higher shear rates close to the wall cause particles to diffuse...
Figure 20: The figure shows the tagged particle undergoing a random walk due to uncorrelated transverse displacements (indicated by double headed arrows) occurring due to asymmetric pair-interactions.

away from this region towards the centre until the radially inward diffusive flux arising from this gradient in the shear rate balances the radially outward flux arising from the resulting gradient in volume fraction ($\phi$ is higher at the centre and lower near the walls), leading to a steady state. At steady state, most of the particles have, in fact, migrated to the centre. The presence of a concentrated highly viscous suspension at the centre, and almost pure suspending liquid close to the walls causes the original Poiseuille flow profile to take on a more plug-like form [21]. Note that such migration occurs in a concentrated suspension occurs at zero $Re$, and its origin is distinct from the Segre-Silberberg effect discussed earlier; the latter is on account of lift forces that arise at finite $Re$.

Shear-induced diffusion might complicate the rheological characterization, of concentration suspensions in particular, via traditional viscometric measurements using the couette or parallel plate geometry. In both devices, particle migration is expected to occur due to a gradient in a shear rate, again leading to inhomogeneity. For the couette device, the particles migrate from the sheared gap to the unsheared bottom, while in the parallel plate, the particles will migrate towards the centre, since the shear rate is highest at the edge of the plates. Thus, during rheological characterization of a concentrated suspension, it is important to determine the time scale characterizing shear-induced diffusion in the system, so one may complete the measurement procedure before the suspension starts to become inhomogeneous.

5 Appendix: Planar linear flows

Planar linear flows are characterized by the axes of the rate of strain tensor being orthogonal to the vorticity direction. They may be represented as a one parameter family with the following expressions for the rate of strain and vorticity tensors:

$$E = \begin{bmatrix} \frac{(1+\lambda)}{2} & 0 & 0 \\ 0 & -\frac{(1+\lambda)}{2} & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad \Omega = \begin{bmatrix} 0 & \frac{(1-\lambda)}{2} & 0 \\ \frac{(1-\lambda)}{2} & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \quad (104)$$

in a cartesian coordinate system with the $x$ and $y$ axes aligned along the principal axes of the rate of strain tensor. Here, $\lambda$ is a parameter that measures the relative magnitudes of extension and vorticity. In the above, $E = \frac{1}{2}(\nabla v + \nabla v^\dagger)$ and $\Omega = \frac{1}{2}(\nabla v^\dagger - \nabla v)$, $\nabla v$ being the velocity gradient tensor. Linear flows where extension dominates ($\lambda > 0$) have open streamlines and are known as hyperbolic linear flows (the streamlines in these cases being hyperbolae), while those with $\lambda < 0$ have closed streamlines and are known as elliptic linear flows (the streamlines in these cases being ellipses). As $\lambda$ ranges from 1 to $-1$, the linear flow changes from an irrotational planar extension, with streamlines that are rectangular hyperbolae, to solid-body rotation with circular streamlines (see figure 21). Simple shear flow ($\lambda = 0$) represents an exact balance between extension and vorticity, leading to rectilinear streamlines, and is a rheologically important flow. The direction of vorticity ($z$), of course, remains perpendicular to the plane of flow ($xy$) in all cases.
Figure 21: The figure shows the variation in the streamline pattern of a planar linear flow with the parameter $\lambda$; the latter indicates the relative magnitudes of extension and vorticity in the linear flow.
References


