Generalized Stokes laws for active colloids and their applications

Rajesh Singh and R Adhikari
1 The Institute of Mathematical Sciences–HBNI, CIT Campus, Chennai 600113, India
2 DAMTP, Centre for Mathematical Sciences, University of Cambridge, Wilberforce Road, Cambridge CB3 0WA, United Kingdom

E-mail: rsingh@imsc.res.in, rjoy@imsc.res.in and ra413@cam.ac.uk

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Abstract

The force per unit area on the surface of a colloidal particle is a fundamental dynamical quantity in the mechanics and statistical mechanics of colloidal suspensions. Here we compute it in the limit of slow viscous flow for a suspension of N spherical active colloids in which activity is represented by surface slip. Our result is best expressed as a set of linear relations, the ‘generalized Stokes laws’, between the coefficients of a tensorial spherical harmonic expansion of the force per unit area and the surface slip. The generalized friction tensors in these laws are many-body functions of the colloidal configuration and can be obtained to any desired accuracy by solving a system of linear equations. Quantities derived from the force per unit area—forces, torques and stresslets on the colloids and flow, pressure and entropy production in the fluid—have succinct expressions in terms of the generalized Stokes laws. Most notably, the active forces and torques have a dissipative, long-ranged, many-body character that can cause phase separation, crystallization, synchronization and a variety of other effects observed in active suspensions. We use the results above to derive the Langevin and Smoluchowski equations for Brownian active suspensions, to compute active contributions to the suspension stress and fluid pressure, and to relate the synchrony in a lattice of harmonically trapped active colloids to entropy production. Our results provide the basis for a microscopic theory of active Brownian suspensions that consistently accounts for momentum conservation in the bulk fluid and at fluid–solid boundaries.

1. Introduction

G G Stokes, in 1851, derived the force per unit area acting on the surface of a sphere moving slowly in an incompressible viscous fluid and thus obtained his celebrated laws for the drag force and torque on a moving sphere. Einstein used these laws in his phenomenological theory of Brownian motion and obtained a relation between the diffusion coefficient of a spherical colloid and the friction constant in Stokes law, the so-called Stokes-Einstein relation, the first example of a fluctuation–dissipation relation [1–3]. Smoluchowski, in 1911, presented an iterative method to calculate the force per unit area on a moving sphere in the presence of another and thereby initiated the study of hydrodynamic interactions between colloidal particles [4]. His method was refined by many authors culminating in the ‘induced force’ method of Mazur and coworkers [5, 6]. The theory of Brownian motion with hydrodynamic interactions developed in parallel with major contributions from Kirkwood [7, 8], Zwanzig [9, 10], and Batchelor [11, 12], amongst others [13, 14]. Through the course of these studies it became apparent that the force per unit area on the surface of the colloid is the key dynamical quantity necessary for developing both the mechanics and the statistical mechanics of suspensions. Despite the classical nature of the subject, the study of colloidal suspensions remains a research area of sustained fecundity [15].

Bearing testimony to this is the recent spate of experimental work on colloidal particles that produce flow in the ambient fluid in the absence of externally applied forces or torques [16–20]. The stress in the fluid produced by this flow reacts back on the colloids and may cause them to translate or rotate. The hydrodynamic interactions between such colloids are without analogue in their classical counterparts. An enormously rich
variety of phenomena have been observed in suspensions of such ‘active colloids’ and a systematic study of this new class of colloidal particles is now underway. It has also been realized that phoretic phenomena in gradients of externally applied fields [21], the swimming of micro-organisms [22, 23], and the self-propulsion of drops [24, 25], though distinct in the microscopic mechanisms that produce fluid flow, share many points of similarity with synthetic active colloids when viewed at the suspension scale. Their study has been revitalized [26–28] and subsumed into the field of active colloids.

The principal feature common to both synthetic active colloids and classical phoretic motion in external fields is the appearance of surface forces, of a range much shorter than the colloidal dimensions, that drive fluid flow in a thin region at the colloidal-fluid boundary. The typical width of this boundary layer is several nanometers, which is approximately a thousand times smaller than the size of the colloidal particle. This wide separation of scales makes it possible to partition the problem of determining the global fluid flow into an interior problem inside the boundary layer, where the flow is driven by the surface forces, and an exterior problem outside the boundary layer, where the exterior fluid is presented with a boundary condition determined by the flow in the boundary layer. The non-universal aspects of the problem are contained, therefore, entirely within the boundary layer leaving the exterior fluid flow to assume universal forms determined by the boundary conditions. Smoluchowski was the first to pursue this approach in his study of electrophoresis [29]. It was used by Derjaguin to study diffusiophoresis [30] and other kinds of phoretic motion [31] and subsequent applications have confirmed the generality and the power of this approach [21].

The swimming of ciliated microorganisms presents another instance of the separation of scales, as the length of the cilium on the surface of such organisms is typically a hundred times smaller than the dimensions of their body. The flow in the ciliary layer can be obtained from the prescribed kinematics of each cilium and the exterior flow then obtained by matching boundary conditions at the interface. This analysis has been developed extensively by Blake [32], following Lighthill’s abstract analysis of the squirming motion of a sphere [33]. These authors only considered axisymmetric squirming motion and it is only recently that the most general non-axisymmetric motion has been studied systematically [34–39].

The self-propulsion of drops is distinct from the previous examples due to the permeability of the interface that separates the internal and external fluids. In these systems, motion is caused by expulsion and suction of fluid at different parts of the interface while approximately maintaining the volume of the drop. In contrast to the previous examples, where the slip flow is tangential to the boundary, in this case, the slip flow has a normal component and thus belongs to the class of osmophoretic phenomena [21, 40]. The method presented here applies such motion, provided that the interfacial forces are sufficiently strong to maintain the spherical drop shape and the rate of dissolution is sufficiently slow that the volume remains approximately constant on the time scales of drop motion.

In all of the above examples, the matching condition at the edge of the boundary layer is the continuity of the fluid velocity, which now contains, in addition to the rigid body motion of the colloid, the contribution from the active flow \( \mathbf{v}^a \). This ‘slip’ velocity is a general, possibly time-dependent, vector field on the colloid surface, subject only to the constraint of mass conservation \( \int \mathbf{v}^a \cdot \mathbf{n} \, dS = 0 \). The fluid mechanical problem is solved when the exterior flow is obtained in terms of the slip. The force per unit area on the colloidal surface follows directly from the Cauchy stress in the fluid and the motion of the colloid is obtained, when colloidal inertia is negligible, by setting the net force and net torque to zero.

The correct expression for the force per unit area was first obtained by Blake [32] for the special case of axisymmetric slip on an isolated sphere distant from all boundaries. This expression was used to compute the work done on the fluid by the slip. However, extensions of this analysis to (i) the most general form of slip in (ii) a suspension of many spheres and (iii) including the effect of proximate boundaries remain to be completed. The availability of such an expression would immediately allow the mechanics and statistical mechanics of active colloidal suspensions to reach the level of development of their passive counterparts. We address ourselves to this task here.

Below, we obtain the force per unit area in a suspension of \( N \) spherical active colloids with the most general form of slip and including the effect of proximate boundaries such as plane walls or plane interfaces. Our results are best expressed as an infinite set of linear relations between the irreducible coefficients of a tensorial harmonic expansion of the force per unit area and the slip. As these contain the Stokes laws for the force and torque as special cases, we refer to them as ‘generalized Stokes laws’.

The generalization has several aspects. First, expressions are obtained for every irreducible mode of the force per unit area, and not just the first two modes representing the force and the torque. Second, the many-body nature of the force per unit area in a suspension of \( N \) colloids is explicitly considered, whereas Stokes laws in their original form apply only to a single particle. Third, the effect of proximate boundaries, such as plane walls, are taken into account to obtain corrections of the kind first derived by Faxén [41] to the original form of Stokes laws. The generalized Stokes laws contain new coefficients, analogous to the translational and rotational friction tensors, that are many-body functions of the colloidal configuration. We show how these generalized friction
tensors can be calculated, to any desired degree of accuracy, from the solution of a linear system. This material is presented in sections 2 and 3.

The generalized Stokes laws immediately provide the quantities of interest to suspension mechanics: the force, the torque, and the stresslet on the colloids. Through their use, succinct expressions are obtained for the fluid flow, the fluid pressure, and the entropy production. We apply these results in the following sections to demonstrate their utility. In section 4 we use results for the force and torque to obtain the over-damped Langevin equations for hydrodynamically interacting Brownian active colloids. The corresponding Smoluchowski equation is also derived and the activity-induced breakdown of the fluctuation-dissipation relation is demonstrated. A contracted description for the one-body distribution function, the first member of a BBGKY-like hierarchy, is derived and the active contribution is highlighted. In section 5, we use the result for the stresslet to obtain an exact expression for the Landau–Lifshitz–Batchelor suspension stress as a function of colloidal configuration. We confirm that the active contribution to the suspension stress vanishes to linear order in volume fraction in an isotropic suspension. In contrast, the active contributions for a non-isotropic suspension of contractile (or extensile) active colloids leads to an increase (or decrease) of the suspension stress. In section 6 we turn to mechanical quantities in the fluid, focusing on the fluid pressure in a suspension of active colloids confined to the interior of a spherical volume. We find that the dynamics of the colloids and the distribution of the fluid pressure are different for suspensions of extensile and contractile colloids. The difference in the collective dynamics of the colloids is understood qualitatively from the fluid flow created by the active colloids. In section 7, we use the Langevin equations to study the collective motion of active colloids in harmonic traps. The traps are centered on a lattice and we consider only one active colloid per trap. Through detailed numerical simulations, we find that a one-dimensional lattice of traps supports time-independent configurations but a two-dimensional lattice of traps only supports time-dependent configurations. In particular, we show that the colloids exhibit a synchronized dynamics about the axis of symmetry in a square lattice of traps and we relate synchrony to the entropy production in a triangular lattice. We conclude in section 8 by discussing other methods for studying active suspensions and by suggesting further applications of the generalized Stokes laws.

The results presented here are based on several key ideas from our previous work on the suspensions of active colloids. The tensorial harmonic expansion of the slip was introduced in \cite{35} and the infinite-dimensional linear system that results from a Galerkin discretization of the boundary integral equation in this expansion basis was obtained in \cite{42}. The linear system was only partially solved to obtain the rigid body motion directly in terms of propulsion tensors. In particular, it was not recognized that a complete solution of the linear system for the coefficients of the force per unit area provide all quantities necessary for developing a complete theory of the mechanics and statistical mechanics of active suspensions. It is this aspect of the problem that is explored here.

The methods of this paper have been applied previously to rationalize the stability of, and transitions between, spontaneously aggregated states of active colloids near boundaries in terms of the hydrodynamically mediated active forces and torques \cite{43,44}. Such a dynamical point of view, in which forces and torques are primary and velocities and angular velocities are derived, yields a generic theory for fluid-induced phase separation (FIPS) which is complementary to the kinematical theory of motility-induced phase separation (MIPS) \cite{45,46,47,48,49,50}. We foresee many other instances where FIPS should provide an accurate representation of the physical forces that drive active aggregation.

2. Generalized Stokes laws

We now present our main result relating the force per unit area \( f \) to the active slip \( \psi^A \) for \( i = 1, \ldots, N \) active colloidal spheres of radius \( b \) suspended in an incompressible viscous fluid of viscosity \( \eta \). The \( i \)th sphere centered at \( \mathbf{R}_i \) has radius vector \( \mathbf{r}_i \) and orientation vector \( \mathbf{p}_i \) as shown in figure 1. The velocity and angular velocity of the sphere are \( \mathbf{V}_i \) and \( \Omega_i \) respectively. In the microhydrodynamic regime, Newton’s equations reduce to instantaneous balance of forces and torques

\[
F^{Hi} + F^p_i + \mathbf{H}_i = 0, \quad T^{Hi} + T^p_i + \mathbf{H}_i = 0. \tag{1}
\]

Here \( F^{Hi} = \int f \, dS \) are hydrodynamic forces, \( F^p_i \) are body forces and \( \mathbf{F}_i \) are Brownian forces, while \( T^{Hi} = \int \mathbf{P}_i \times f \, dS \) and \( \mathbf{T}_i \) are corresponding torques. The velocity \( \mathbf{u}(\mathbf{R}_i + \mathbf{r}_i) \) of the fluid at a point \( \mathbf{R}_i + \mathbf{r}_i \) on the colloid boundary is a sum of its rigid body motion and the active slip \( \psi^A \)

\[
\mathbf{u}(\mathbf{R}_i + \mathbf{r}_i) = \mathbf{V}_i + \Omega_i \times \mathbf{r}_i + \psi^A(\mathbf{r}_i). \tag{2}
\]

The wide applicability of slip to model motion driven by interfacial forces has been mentioned above. To these, we add the recent observation that \( \psi^A \) need not be identified with the material surface of the colloid but can, instead, be thought of as the velocity at the surface of an effective sphere enclosing a non-spherical active body
such as the biflagellate green algae *C. Reinhardtii* [35]. A sphere with slip, therefore, provides a good representation for a surprisingly large variety of active flows and should be preferred as the simplest dynamical model of a finite-sized active body.

Both the slip and the force per unit area are vector fields on the sphere, which makes harmonic expansions natural. We choose the tensorial spherical harmonics $Y_l^m$ as the expansion basis for reasons discussed below. These harmonics are dimensionless, symmetric, irreducible Cartesian tensors of rank $l$ that form a complete, orthogonal basis on the sphere [51–53]. The expansion of the slip and the force per unit area on the surface of the $i$th colloid is

$$v_i^{(s)}(R_i + \rho_i) = \sum_{l=1}^{\infty} \frac{1}{(l-1)!(2l-3)!} V_i^{(s)} \cdot Y_l^{(l-1)}(\hat{\rho}_i), \quad f(R_i + \rho_i) = \sum_{l=1}^{\infty} \frac{2l-1}{4\pi b^2} F_i^{(s)} \cdot Y_l^{(l-1)}(\hat{\rho}_i).$$

The expansion coefficients $V_i^{(s)}$ and $F_i^{(s)}$, of dimensions of force and velocity respectively, are tensors of rank $l$, symmetric and irreducible in their last $l-1$ indices [42]. Elementary group theory assures [54] that they can be expressed as the sum of three irreducible tensors of rank $l$, $l-1$ and $l-2$. The three irreducible parts are

$$V_i^{(s)} = P_i^{(s)}, \quad V_i^{(s)} = P_i^{(s)}, \quad F_i^{(s)} = P_i^{(s)} ,$$

where the index $\sigma = s, a$ and $t$, labels the symmetric irreducible, antisymmetric and trace parts of the reducible tensors. The operator $P_i^{(s)} = \Delta_i^{(l)}$ extracts the symmetric irreducible part, $P_i^{(a)} = \Delta_i^{(l-1)} e$ the antisymmetric part and $P_i^{(t)} = \delta$ the trace of the operand. Here $\Delta_i^{(l)}$ is tensor of rank $2l$, projecting any $l$th order tensor to its symmetric irreducible form [53], $e$ is the Levi-Civita tensor and $\delta$ is the Kronecker delta. As the $V_i^{(s)}$ are irreducible tensors, it is natural to parametrize them in terms of the tensorial spherical harmonics. Their uniaxial parametrizations are

$$V_i^{(s)} = V_0^{(s)} Y_i^{(l)}(p_i), \quad V_i^{(a)} = V_0^{(a)} Y_i^{(l-1)}(p_i), \quad V_i^{(t)} = V_0^{(t)} Y_i^{(l-2)}(p_i).$$

It follows that the coefficients are either even (apolar) or odd (polar) under inversion symmetry $p_i \rightarrow -p_i$. The leading terms of the expansion, categorized according to their symmetry under inversion and mirror reflection, are
Table 1. Active slip velocity $v_A$ in terms of spherical polar coordinates ($\rho$, $\theta$, $\phi$), for leading coefficients of polar, apolar and chiral symmetry (equation (6)). We have used orientation $p$ of the colloids for uniaxial parametrization of the coefficients of the slip expansion (equation (5)). Without any loss of generality, we choose $p$ to be along $z$-axis, such that $p = \cos \theta \hat{p} - \sin \theta \hat{\theta}$.

\begin{tabular}{|c|c|c|c|}
\hline
$n$ & $v_{ij}$ & $v_{ii}$ & $v_{i\phi}$ \\
\hline
2s & $V_{ij}^{(2s)} \left( \frac{1}{r^3} - \sin^2 \theta \right)$ & $-\frac{1}{2} V_{ij}^{(2s)} \sin 2\theta$ & 0 \\
3s & $V_{ij}^{(3s)} \cos \theta \left( \cos^2 \theta - \frac{1}{3} \right)$ & $-V_{ij}^{(3s)} \sin \theta \left( \cos^2 \theta - \frac{1}{2} \right)$ & 0 \\
3a & 0 & 0 & $\frac{1}{3} V_{ij}^{(3a)} \sin 2\theta$ \\
3t & $\frac{1}{3} V_{ij}^{(3t)} \cos \theta$ & $\frac{1}{3} V_{ij}^{(3t)} \sin \theta$ & 0 \\
4a & 0 & 0 & $\frac{1}{10} V_{ij}^{(4a)} \sin \theta \left( \cos^2 \theta - \frac{1}{2} \right)$ \\
\hline
\end{tabular}

\[ v_A^{(\rho)}(\rho) = -V_i^{(1,\rho)} + \frac{1}{2} V_i^{(3,\rho)}(\hat{\rho}) + \frac{3}{2} \frac{1}{a^2} \rho_i, \quad \Omega_i^{(\rho)} = -\frac{1}{2} V_i^{(2a)}(\hat{\rho}) \\
\]

4$\pi a^2 V_i^{(\phi)} = -\int v_A^{(\phi)}(\rho) dS, \quad 4\pi a^2 \Omega_i^{(\phi)} = -\frac{3}{2a^2} \int \rho_i \times v_A^{(\phi)}(\rho) dS, \quad (6a) \]

(6b)

Here $V_i^{(1,\rho)}$ is the active translational velocity, while $\Omega_i^{(\rho)}$ is the active angular velocity for a sphere in unbounded medium $[34, 35, 55]$. The tangential flows generated by these leading terms are shown in figure 1. Their complete form, including radial terms, is provided in table 1 of the appendix.

The tensorial harmonics have several advantages over the more common vector spherical harmonics or surface polynomials $[56, 57]$. First, both basis functions and expansion coefficients transform as Cartesian tensors which allows physical quantities like the force, the torque and the stresslet to be represented covariantly. Second, the linear independence of the basis functions ensures that linear systems for the coefficients are always of full rank. Third, the basis functions are well-suited for Taylor expansions $[42]$ and their addition theorems are considerably simpler than the usual vector spherical harmonics. This property allows for fast methods of summing long-ranged harmonics $[58]$ that are less complex than the classical methods $[59]$. Previous studies of spherical passive suspensions have amply demonstrated some of these advantages $[5, 6, 35, 42, 60]$. The problem, now, is to relate the unknown values of the traction coefficients $F_i^{(\sigma)}$ to the known values of the slip coefficients $V_i^{(\sigma)}$.

In the regime of slow viscous flow, the fluid velocity $u$ in equation (2) obeys the Stokes equation. The linearity of the governing equation and of the boundary condition implies, then, a linear relation between the velocity and traction coefficients

\[ F_i^{(\sigma)} = -\gamma_i^{(\sigma, 1, \alpha)} : V_j - \gamma_i^{(\sigma, 2a)} : \Omega_j, \quad \Omega_i^{(\sigma)} = \sum_{\ell = 1}^{\infty} \gamma_i^{(\sigma, 2\ell)} : V_j^{(2\ell)}, \quad (7) \]

where repeated particle indices are summed over. We call the above infinite set the generalized Stokes laws. Their best-known special cases are Stokes laws for translation $F_i = -6\pi \eta b V_i$ and rotation $T_i = -8\pi \eta b^3 \Omega_i$. The first two terms are passive frictional contributions due to rigid body motion while the remaining terms are active contributions due to slip. The $\gamma_i^{(\sigma, 2\ell)}$ are generalized friction tensors and their symmetry properties, to which we shall return below, allow them to be interpreted as generalized Onsager coefficients. Their rank varies from $l + 1$ to $l + 1' = 4$ depending on the values of $\sigma$ and $\sigma'$. The friction tensors depend on the positions of all spheres, reflecting the many-body character of the hydrodynamic interaction but, due to isotropy, are independent of the sphere orientations. The traction coefficients, however, depend on the orientations of the spheres carried in the coefficients of the slip. Given the velocity at the boundary of each sphere, the friction tensors completely determine the force per unit area on every sphere. In the next section, we derive exact expressions for the generalized friction tensors in terms of Green’s functions of Stokes flow.

3. Boundary integral solution

The most direct way of computing the generalized friction tensors is through the boundary integral representation of Stokes flow $[42, 56, 61–66]$

\[ u_i(r) = -\int G_{\sigma\beta}(r, r_j) f_j(r_j) dS_j + \int K_{\sigma\beta}(r, r_j) \hat{p}_j u_j(r_j) dS_j, \quad (8a) \]

\[ \hat{p}_j u_j(r_j) dS_j, \quad (8b) \]

5
\[ p(r) = -\int P_0(r, r_j) f_j(r_j) \, dS_j + \int Q_{\omega;3}(r, r_j) \hat{\partial}_\omega u\omega(r_j) \, dS_j, \] (8b)

where \( p \) is the fluid pressure. In the above, repeated Cartesian and particle indices are summed over and points on the boundary of the sphere are given by \( r_i = R + \rho_i \). The kernels in the boundary integral representations are the Green’s function \( G \), the pressure vector \( P \), its gradient \( Q = 2\eta \nabla P \), and the stress tensor \( K \). They satisfy

\[ \nabla_i G_{\omega;3}(r, r') = 0, \quad -\nabla_i P_j(r, r') + \eta \nabla^2 G_{\omega;3}(r, r') = -\delta(r-r')\delta_{i\omega}, \] (9a)
\[ K_{\omega;3}(r, r') = -\delta_{i\omega} P_j(r, r') + \eta (\nabla_i G_{\omega;3}(r, r') + \nabla_j G_{\omega;3}(r, r')). \] (9b)

The flow and pressure satisfying the Stokes equation, \( \nabla \cdot u = 0 \) and \( \nabla \cdot \sigma = 0 \) with \( \sigma = -p I + \eta (\nabla u + (\nabla u)^T) \) the Cauchy stress in a fluid of viscosity \( \eta \), is the sum of the ‘single-layer’ integral of the local force per unit area \( f = \hat{\rho} \cdot \sigma \) and the ‘double-layer’ integral of the boundary velocity \( \mathbf{V} + \mathbf{\Omega} \times \mathbf{r} + \mathbf{r}'(\mathbf{r}) \). From now on, we shall refer to \( f \) as the traction. Enforcing the boundary conditions in the integral representation produces a Fredholm integral equation of the first kind for the unknown force per unit area. The problem is thereby reduced from the solution of a partial differential equation in a three-dimensional volume to the solution of an integral equation over two-dimensional surfaces.

Several methods of solution have been developed for Fredholm integral equations for Stokes flows [6, 42, 56, 63–65, 67]. These differ in their choice of formulation, discretization, and strategy of minimizing the residual. Here, we use a direct formulation, in which the single-layer density is the physical force per unit area, in contrast to other formulations where such an interpretation is unavailable [6, 56]. The choice of discretization in terms of the tensorial spherical harmonics is natural for spheres and such global basis functions yield the greatest accuracy for the least number of unknowns [65]. Finally, Galerkin’s method of minimizing the residual is chosen as it yields a self-adjoint linear system for the coefficients, an advantageous property for numerical solutions. This choice of formulation, discretization and residual minimization is of greatest utility for spheres, as the matrix elements of the linear system can be evaluated analytically [42] and the numerical quadrature typically associated with the Galerkin method can be avoided entirely. We note that for particles of non-spherical shape, these advantages are no longer available.

The linear system we obtain using this direct formulation with the global tensorial harmonic basis functions and Galerkin’s method of minimizing the residual is [42]

\[ -G^{(s)}_{ij}(r, r_j) \cdot F_j^{(s)} + K^{(s)}_{ij}(r, r_j) \cdot V_j^{(s)} = \begin{cases} \frac{1}{2}(V_i - V_j^s) & \text{for } \sigma = 1, \\ \frac{1}{2}(b\Omega_i - b\Omega_j^s) & \text{for } \sigma = 2, \\ \frac{1}{2}V_i^{(s)} & \text{otherwise,} \end{cases} \] (10)

where the matrix elements \( G^{(s)}_{ij}(r, r_j) \) and \( K^{(s)}_{ij}(r, r_j) \)

\[ G^{(s)}_{ij}(r, r_j) = \frac{(2l_1 - 1)(2l_2 - 1)}{4\pi b^2} \int Y^{(l_1-1)}(\hat{r}) G(R_i + \rho_i, R_j + \rho_j) Y^{(l_2-1)}(\hat{r}) \, dS_i dS_j, \]

\[ K^{(s)}_{ij}(r, r_j) = \frac{2l_1 - 1}{4\pi b^2(l_1 - 1)(2l_1 - 3)} \int Y^{(l_1-1)}(\hat{r}) K(R_i + \rho_i, R_j + \rho_j) \cdot \hat{\rho}_j Y^{(l_2-1)}(\hat{r}) \, dS_i dS_j, \]

are given in terms of the Green’s function \( G(R_i, R_j) \) and its derivatives, see appendix A. This linear system differs in important ways from superficially similar linear systems derived for passive colloids [6]. First, the right-hand sides of the equations corresponding to the rigid body motions contain self-propulsion and self-rotation contributions; these are absent in passive colloids. Second, the double-layer integral adds a non-trivial contribution to the linear system; this is trivial in passive colloids as rigid body motions are in the eigenspace of the double-layer. Finally, the equations for the non-rigid-body modes are inhomogeneous, both due to double layer and slip contributions; these equations are homogeneous for passive systems as both double-layer and slip contributions are zero. Linear systems for passive colloids, derived earlier by Mazur [3] and by Ladd [6], are recovered from the above when all \( V_j^{(s)} \) are set to zero.

The generalized Stokes laws are obtained as solutions of this linear system. The formal solution of the discrete linear system yields the following expression for the friction tensors

\[ \gamma^{(s,r,r')} = P^{(s,r')} \cdot \left[ G^{-1} \cdot \left( \frac{1}{2} I - K \right)^{(s,r')} \right] \cdot P^{(s,r')} . \] (11)

In the above, we have introduced the block matrices \( G \) and \( K \) whose \((l, l')\) element in the \(ij\) block are \( G_{ij}^{(s)}(R_i, R_j) \) and \( K_{ij}^{(s)}(R_i, R_j) \) respectively. The many-body character of the friction tensors is apparent from this expression, as the inverse of \( G \) cannot be expressed in terms of sums of the pairwise interactions encoded in its matrix elements. Since the matrix elements only involve particle positions and not orientations, the friction tensors are, likewise, independent of particle orientation.
Table 2: The Green’s functions of Stokes equation for a system of two colloids at $R_i$ and $R_j$ respectively [127–129]. $\mathcal{M} = I - 2\mathbf{K}$, where $\mathbf{K} = \mathbf{G} \cdot \mathbf{R}$ is the image of the $j$th colloid at a distance $h$ from the interface/wall at $z = 0$.

| Green’s function | Unbounded fluid: $G_{ij}^{(R_i - R_j)} = \frac{1}{4\pi h} \nabla_i \delta_{ij} - \nabla_i \delta_{ij} | r_i |^{-2} $.
| --- | --- |
| Plane interface (fluid-gas): $G_{ij}^{(R_i, R_j)} = G_{ij}^{(r_i)} + (\delta_{ii} \delta_{jj} - \delta_{ij} \delta_{ij}) G_{ij}^{(r_i)} $. |
| Plane interface (fluid-fluid): $G_{ij}^{(R_i, R_j)} = G_{ij}^{(r_i)} + M_{ij} G_{ij}^{(r_j)} - 2h \nabla_i \nabla_j G_{ij}^{(r_i)} M_{ij} + h^2 \nabla_i \nabla_j G_{ij}^{(r_j)} M_{ij} $. |
| Plane no-slip wall: $G_{ij}^{(R_i, R_j)} = G_{ij}^{(r_i)} - G_{ij}^{(r_j)} - 2h \nabla_i G_{ij}^{(r_i)} M_{ij} + h^2 \nabla_i \nabla_j G_{ij}^{(r_j)} M_{ij} $. |

The method of solution for the linear system, or equivalently, of inverting $G$ must be chosen according to need. For analytical solutions, Jacobi’s iterative method is straightforward and its widespread use in Stokes flows can be traced back to Smoluchowski’s method of reflections [68]. Other tractable analytical methods rely on series expansion of $G^{-1}$ in powers of $G$ [69]. We give explicit analytical expressions for the friction tensors obtained from the Jacobi method in appendix A. For numerical solutions, iterative solvers with faster rates of convergence must be used. Typically, these search for the inverse in the Krylov subspace of $G$ and have the advantage of requiring, instead of $G$, only its action on a vector of the appropriate size. The need to store a large dense matrix is thereby avoided. For the self-adjoint linear system above, the stable, efficient and accurate conjugate gradient method [70] may be used. This requires $O(M^2)$ computational effort for $M$ unknowns when matrix–vector products are computed directly. The use of fast summation methods can reduce the cost to $O(M \log M)$ [71, 72] or even $O(M)$ [39, 73, 74]. With these preliminary remarks, we postpone the study of numerical solutions for the friction tensors to a future work.

Expressions for the friction tensors in terms of Green’s functions of the Stokes equation have been derived in appendix A. Examples of the Green’s functions of Stokes flow for typical flow geometries used in the experiments of active colloids is provided in table 2. As shown in appendix A, the leading order contribution to the friction tensor is

$$
\gamma_{ij}^{(R_i, R_j)} \sim \nabla_i \nabla_j G(R_i, R_j) .
$$

(12)

In an unbounded fluid, this decays as $|R_i - R_j|^{-(l + l') + 1}$ and is thus long-ranged for $l + l' \leq 4$. Thus, all slip modes up to $l = 3$ produce long-ranged forces and up to $l = 2$ produce long-ranged torques. Notably, the active forces and torques have contributions even without self-propulsion, $\mathbf{V}_i^A = 0$, or self-rotation, $\Omega_i^A = 0$, indicating that colloids can be ‘active’ without necessarily being self-propelling or self-rotating. The friction tensors depend on the positions of all the particles and are thus many-body functions of the instantaneous colloidal configurations. A remarkable feature of active forces and torques is that they depend not only on the relative position of the particles, as in a passive suspension of spheres, but also through the slip moments, on their orientations. Thus active colloids, even if they are geometrically isotropic, are hydrodynamically anisotropic. An intuitive understanding of the orientation–dependent forces and torques can be obtained by studying figure 6 of the appendix, where a change in orientation of the active colloid alters the flow field it ‘carries’ and consequently the stresses that are transmitted by this flow field. Finally, though these forces and torques appear in Newton’s equations, they are, emphatically, not body forces and torques: they are the sum of the dissipative surface forces that act at the fluid-solid boundary.

The boundary integral representation of equation (8) can be used to obtain explicit expressions for the fluid velocity and the fluid pressure at any point in the bulk of the fluid. By expanding the traction and velocity in the boundary integral expression we obtain expression for fluid flow and pressure in terms of the tensorial coefficients as

$$
u(r) = \sum_{l=1}^{\infty} \left( -G_j^{(l)}(r, R_j) \cdot F_j^{(l)} + K_j^{(l)}(r, R_j) \cdot V_j^{(l)} \right),
$$

(13a)

$$
\rho(r) = \sum_{l=1}^{\infty} \left( -P_j^{(l)}(r, R_j) \cdot F_j^{(l)} + Q_j^{(l)}(r, R_j) \cdot V_j^{(l)} \right),
$$

(13b)

where the boundary integrals for fluid flow and pressure are

$$
G_j^{(l)}(r, R_j) = \frac{2l - 1}{4\pi b^2} \int G(r, R_j + \rho_j) Y^{(l-1)}(\hat{\rho}_j) dS_j,
$$

(14a)

$$
K_j^{(l)}(r, R_j) = \frac{1}{(l - 1)(2l - 3))} \int K(r, R_j + \rho_j) \cdot \hat{\rho}_j Y^{(l-1)}(\hat{\rho}_j) dS_j,
$$

(14b)
\[ P_{ij}^{(l)}(r, R_j) = \frac{2l - 1}{4\pi b^2} \int \mathbf{P}(r, R_j + \rho_j) Y^{(l-1)}(\hat{\rho}_j) \, dS_j, \]  
\[ Q_{ij}^{(l)}(r, R_j) = \frac{1}{(l-1)!(2l - 3)!!} \int \mathbf{Q}(r, R_j + \rho_j) \cdot \hat{\rho}_j Y^{(l-1)}(\hat{\rho}_j) \, dS_j. \]  

These boundary integrals are obtained explicitly in terms of the Green’s function of Stokes flow and fluid pressure and their derivatives. These results are derived in appendix C. Using the generalized Stokes laws of equation (7) and balance conditions of equation (1), the unknown traction coefficients can be written in terms of the known slip coefficients and the body forces and torques. The expression for fluid velocity and pressure can be then expressed entirely in terms of known quantities as

\[ u(r) = G_{ij}^{(l)}(r, R_j) \cdot \mathbf{F}_j^p + G_{ij}^{(2a)}(r, R_j) \cdot \mathbf{T}_j^p + \sum_{l=2}^{\infty} \Pi_{ij}^{(lr)} \cdot \mathbf{V}_{ij}^{(lr)}, \]

\[ p(r) = P_{ij}^{(l)}(r, R_j) \cdot \mathbf{F}_j^p + \sum_{l=2}^{\infty} \Lambda_{ij}^{(l)} \cdot \mathbf{V}_{ij}^{(l)}. \]

Here \( G_{ij}^{(l)} \) and \( G_{ij}^{(2a)} \) are matrices which, respectively, relate the fluid velocity with body forces and torques while \( \Pi_{ij}^{(lr)} \) give the linear relation between coefficients of slip velocity to fluid velocity. The contribution of body forces to pressure is given by \( P_{ij}^{(l)} \), while \( \Lambda_{ij}^{(l)} \) gives the contributions from the symmetric irreducible modes of the active slip. It is instructive to note that only symmetric traceless parts \( \mathbf{V}_{ij}^{(l)} \) of the slip modes contribute to the fluid pressure, as pressure is harmonic, unlike the fluid velocity which is biharmonic. \( \Pi_{ij}^{(lr)} \) and \( \Lambda_{ij}^{(l)} \) are tensors of rank \( l \) and \( l + 1 \), respectively, and in an unbounded fluid, decay as \( r^{-l} \) and \( r^{-l+1} \) with distance \( r \).

The power dissipation in the volume of the fluid [75] can be reduced to integrals on the colloidal boundaries using the divergence theorem

\[ \mathcal{E} = \int \sigma : (\nabla u) \, dV = -\sum_i N_1 f_i (R_i + \rho_i) \cdot u(R_i + \rho_i) \, dS_i, \]

and using equation (3) along with the orthogonality of the basis, can be expressed in terms of the expansion coefficients as

\[ \mathcal{E} = -\sum_i \left( \mathbf{F}_i^H \cdot \mathbf{V}_i + \mathbf{T}_i^H \cdot \Omega_i + \sum_{l=1}^{\infty} \mathbf{F}_{ij}^{(l)} \cdot \mathbf{V}_{ij}^{(l)} \right). \]

The generalized Stokes laws can now be used to eliminate the unknown traction coefficients for the known slip coefficients to obtain

\[ \mathcal{E} = -\mathbf{F}_i^H \cdot \mathbf{V}_i - \mathbf{T}_i^H \cdot \Omega_i + \mathbf{V}_{i1}^{(l)} \cdot \gamma_{ij}^{(l,1a)} \cdot \mathbf{V}_j + \mathbf{V}_{i2}^{(l)} \cdot \gamma_{ij}^{(l,2a)} \cdot \Omega_j + \mathbf{V}_{ij}^{(l)} \cdot \gamma_{ij}^{(l,F,s)} \cdot \mathbf{V}_{ij}^{(l,F,s)}. \]

The positivity of power dissipation requires the friction tensors to be positive-definite. Proofs of the latter are available in [76, 77].

The differences with passive colloids are noteworthy: the colloids can produce flow in the absence of external forces and torques and both this flow and the pressure depend on the position and the orientation of the colloids. The latter fact is illustrated in figure 6. The contributions from the slip makes flow in active suspensions intrinsically more rich and leads to novel phenomena with no analogue in passive suspensions.

To summarize, the main result of this section is equation (11), which gives exact expressions for the generalized friction tensors in terms of the matrix elements of the discretized boundary integral equation. An approximate solution to this, in leading powers of distance between the colloids, is provided in appendix A. We now turn to applications of the above results.

4. Langevin and Smoluchowski descriptions

The net hydrodynamic force \( \mathbf{F}_i^H \) and torque \( \mathbf{T}_i^H \) acting on the \( i \)th colloid are related to its first two irreducible coefficients of the traction \( \mathbf{f} \) and from the generalized Stokes laws these are

\[ \mathbf{F}_i^H = -\gamma_{ij}^{TT} \cdot \mathbf{V}_j - \gamma_{ij}^{TR} \cdot \Omega_j - \sum_{l=1}^{\infty} \gamma_{ij}^{(l,T,lr)} \cdot \mathbf{V}_{ij}^{(lr)}, \]

\[ \mathbf{T}_i^H = -\gamma_{ij}^{RT} \cdot \mathbf{V}_j - \gamma_{ij}^{RR} \cdot \Omega_j - \sum_{l=1}^{\infty} \gamma_{ij}^{(l,R,lr)} \cdot \mathbf{V}_{ij}^{(lr)}. \]

Here, the \( \gamma_{ij}^{\alpha\beta} \) with \( \alpha, \beta = T, R \) are relabellings of the four friction tensors \( \gamma_{ij}^{(l,F,s')} \) with \( l, l' s' = 1s, 2a \), where the correspondences are \( T \leftrightarrow 1s \) and \( R \leftrightarrow 2a \). The \( \gamma_{ij}^{(T,lr)} \) and \( \gamma_{ij}^{(R,lr)} \) are relabellings of the friction tensors \( \gamma_{ij}^{(1s,lr)} \) and \( \gamma_{ij}^{(2a,lr)} \) respectively. The first two terms in the force and torque expressions above are the usual rigid
body drag [5, 6] while the remaining terms are the contributions from the slip. The effect of activity is then clear: it adds long-ranged, many-body correlated, orientation-dependent dissipative forces and torques to the familiar Stokes drags.

The hydrodynamic forces and torques obtained above can be used to construct the Langevin equations describing the motion of active colloids in a thermally fluctuating fluid [9, 77–82]. This is obtained from the balance of hydrodynamic, body and Brownian forces and torques as provided in equation (1). The Brownian forces and torques, \( \mathbf{f}_i \) and \( \mathbf{\Gamma}_i \), are zero-mean, Gaussian white noises and the fluctuation-dissipation relation fixes their variances to be

\[
\langle \mathbf{f}_i(t) \mathbf{f}_j(t') \rangle = 2k_B T \gamma^{TT}_{ij} \delta(t - t'), \quad \langle \mathbf{f}_i(t) \mathbf{\Gamma}_j(t') \rangle = 2k_B T \gamma^{TR}_{ij} \delta(t - t'), \quad \langle \mathbf{\Gamma}_i(t) \mathbf{\Gamma}_j(t') \rangle = 2k_B T \gamma^{RR}_{ij} \delta(t - t').
\]

(20a, 20b)

where \( k_B \) is the Boltzmann constant and \( T \) is the temperature. We do not consider any fluctuations corresponding to activity, since its inherently non-equilibrium nature precludes any possible balance between fluctuation and dissipation. Using explicit forms, the Langevin equations for active colloids with hydrodynamic interactions are

\[
-\gamma^{TT}_{ij} \cdot \mathbf{V}_j - \gamma^{TR}_{ij} \cdot \mathbf{\Omega}_j + \mathbf{F}^T_i + \mathbf{f}_i = \sum_{l=1}^{\infty} \gamma^{(T,ls)}_{ij} \cdot \mathbf{V}^{(ls)}_j = 0,
\]

\[
-\gamma^{RT}_{ij} \cdot \mathbf{V}_j + \gamma^{RR}_{ij} \cdot \mathbf{\Omega}_j + \mathbf{T}^P_i + \mathbf{\Gamma}_i = \sum_{l=1}^{\infty} \gamma^{(R,ls)}_{ij} \cdot \mathbf{V}^{(ls)}_j = 0.
\]

(21a, 21b)

The above equations contain forces due to Stokes drags, body forces, thermal fluctuations, and activity. Motion is driven by the last three terms and their relative importance can be captured by two ratios. We choose the first of these to be the ratio of active and body forces and the second to be the ratio of thermal and active forces. Similar considerations apply for the torque balance. These motivate the introduction of the following dimensionless numbers

\[
A_T = \frac{|\gamma^{(T,ls)}_{ij} \cdot \mathbf{V}^{(ls)}_j|}{|\mathbf{F}^T_i|}, \quad A_R = \frac{|\gamma^{(R,ls)}_{ij} \cdot \mathbf{V}^{(ls)}_j|}{|\mathbf{T}^P_i|}, \quad B_T = \frac{|\mathbf{f}_i|}{|\gamma^{(T,ls)}_{ij} \cdot \mathbf{V}^{(ls)}_j|}, \quad B_R = \frac{|\mathbf{\Gamma}_i|}{|\gamma^{(R,ls)}_{ij} \cdot \mathbf{V}^{(ls)}_j|}.
\]

(22)

Here \( A_T \) and \( A_R \) are ‘activity’ numbers quantifying the relative importance of active and body terms [83–85] while \( B_T \) and \( B_R \) are ‘Brown’ numbers quantifying the relative importance of thermal and active terms We estimate these numbers for two typical active colloidal systems below.

Explicit Langevin equations for the velocity and angular velocity are obtained by inverting equation (21). Since the \( \gamma^{os}_{ij} \) and the \( \gamma^{(os,ls)}_{ij} \) are identical for \( ls = 1s, 2a \), it is convenient to group the velocity with the self-propulsion and the angular velocity with the self-rotation so that the summation is from \( ls = 2s \) onward. With this regrouping, the result is

\[
\mathbf{V} = \mathbf{\mu}^{TT}_{ij} \cdot \mathbf{T}^T_j + \mathbf{\mu}^{TR}_{ij} \cdot \mathbf{\Omega}^T_j + \sqrt{2k_B T \mathbf{\mu}^{TT}_{ij} \cdot \mathbf{\eta}^T_j} + \sqrt{2k_B T \mathbf{\mu}^{TR}_{ij} \cdot \mathbf{\zeta}^T_j} + \sum_{ls=2s}^{\infty} \pi^{(T,ls)}_{ij} \cdot \mathbf{V}^{(ls)}_j + \mathbf{V}^A_i,
\]

\[
\mathbf{\Omega} = \mathbf{\mu}^{RT}_{ij} \cdot \mathbf{F}^P_j + \mathbf{\mu}^{RR}_{ij} \cdot \mathbf{\Omega}^P_j + \sqrt{2k_B T \mathbf{\mu}^{RT}_{ij} \cdot \mathbf{\eta}^P_j} + \sqrt{2k_B T \mathbf{\mu}^{RR}_{ij} \cdot \mathbf{\zeta}^P_j} + \sum_{ls=2a}^{\infty} \pi^{(R,ls)}_{ij} \cdot \mathbf{V}^{(ls)}_j + \mathbf{\Omega}^A_i.
\]

(23a, 23b)

Here \( \mathbf{\eta}^P, \mathbf{\zeta}^P \) are Gaussian white noises with zero-mean and variances \( 1, 1/b \) respectively. The mobility matrices \( \mathbf{\mu}^{os}_{ij} \) are inverses of the friction matrices \( \gamma^{os}_{ij} [5, 6, 67, 86–92] \). The propulsion tensors \( \pi^{(os,ls)}_{ij} \), first introduced in [42], relate the rigid body motion to modes of the active velocity. They are related to the slip friction tensors introduced here by

\[
-\pi^{(T,ls)}_{ij} = \mathbf{\mu}^{TT}_{ik} \cdot \gamma^{(T,ls)}_{kj} + \mathbf{\mu}^{TR}_{ik} \cdot \gamma^{(R,ls)}_{kj},
\]

\[
-\pi^{(R,ls)}_{ij} = \mathbf{\mu}^{RT}_{ik} \cdot \gamma^{(T,ls)}_{kj} + \mathbf{\mu}^{RR}_{ik} \cdot \gamma^{(R,ls)}_{kj}.
\]

(24a, 24b)

The translational propulsion tensors \( \pi^{(T,ls)}_{ij} \) are dimensionless while the rotational propulsion tensors \( \pi^{(R,ls)}_{ij} \) have dimensions of inverse length. The above form of the propulsion tensors is particularly useful when mobilities are evaluated by combining the far-field and near-field lubrication contributions. Through this approximation, the need to resolve the rapidly varying flow at particle contact can be avoided, and accurate results can be obtained by keeping only the long-ranged contributions when solving the linear system.

Stochastic trajectories can be obtained by integrating the kinematic equations

\[
\mathbf{R}_i = \mathbf{V}_i, \quad \mathbf{p}_i = \mathbf{\Omega}_i \times \mathbf{p}_i.
\]

(25)
using the standard Brownian dynamics integrators, for example that due to Ermak and Mc Cammon [93]. We note that the explicit form of the Langevin equation was first obtained in [85] using heuristic arguments.

The Smoluchowski equation for the distribution function \( \Psi(\mathbf{R}_1, \ldots, \mathbf{R}_N; \mathbf{p}_1, \ldots, \mathbf{p}_N) \equiv \Psi(\mathbf{R}^N; \mathbf{p}^N) \) of positions and orientations follows immediately from the Langevin equations. We write it in the form of a conservation law in configuration space

\[
\frac{\partial \Psi}{\partial t} = - \sum_{ij} \mathcal{L}_{ij} \Psi = - \sum_{ij} (\nabla_{R_i} \cdot \mathbf{V}_{R_i} + \mathbf{p}_i \times \nabla_{p_i} \cdot \mathbf{V}_{p_i}) \Psi,
\]

where the ‘velocities’ \( \mathbf{V}_{R_i} \) and \( \mathbf{V}_{p_i} \) are

\[
\mathbf{V}_{R_i} = \mu_{ij}^{TT}(\mathbf{F}^p_j - k_bT \mathbf{V}_{R_j}) + \mu_{ij}^{TR}(\mathbf{T}^p_j - k_bT \mathbf{p}_j \times \mathbf{V}_{R_j}) + \sum_{l=2s}^{\infty} \pi_{ij}^{(T,ls)} \cdot \mathbf{V}_{R_l}^{(ls)} + \mathbf{V}_{R_i}^{A},
\]

\[
\mathbf{V}_{p_i} = \mu_{ij}^{TT}(\mathbf{F}^p_j - k_bT \mathbf{V}_{R_j}) + \mu_{ij}^{RR}(\mathbf{T}^p_j - k_bT \mathbf{p}_j \times \mathbf{V}_{R_j}) + \sum_{l=2s}^{\infty} \pi_{ij}^{(R,ls)} \cdot \mathbf{V}_{R_l}^{(ls)} + \Omega_i^A.
\]

Here \( \mathbf{F}^p_j = - \mathbf{V}_{R_j} U, \mathbf{T}^p_j = - \mathbf{p}_j \times \mathbf{V}_{R_j} U, \) and \( U \) is a potential that contains both positional and orientational interactions. In the absence of activity, the equation obeys the fluctuation-dissipation relation and the Gibbs distribution \( \Psi \sim \exp\left(-U/k_bT\right) \) is the stationary solution. This solution has a vanishing current. In contrast, in the presence of activity, the Gibbs distribution is no longer the stationary distribution and in general, only the divergence of the current is zero. Circulation currents are a generic possibility in such circumstances and have, indeed, been observed in models of active colloidal systems [42, 45, 94, 95].

The Smoluchowski equation above is a partial differential equation in 6N variables and is generally intractable as it stands. However, by standard reduction methods, equations for the one and two particle distribution functions may be obtained from it, providing a principled way of recovering coarse-grained macroscopic equations from the microscopic dynamics [96–100]. To this end, we define the \( n \)-body density \( c^{(n)} \) as,

\[
c^{(n)}(\mathbf{R}^n, \mathbf{p}^n, t) = \frac{N!}{(N-n)!} \int \Psi(\mathbf{R}^N; \mathbf{p}^N) \prod_{i=1}^{N} d\mathbf{R}_i d\mathbf{p}_i.
\]

The dynamics of one-body density is then obtained by using the Smoluchowski equation

\[
\frac{\partial c^{(1)}}{\partial t} = -N \sum_{ij} \int \mathcal{L}_{ij} \Psi(\mathbf{R}^N; \mathbf{p}^N) \prod_{i=2}^{N} d\mathbf{R}_i d\mathbf{p}_i = -N \sum_{ij} (\nabla_{R_i} \cdot \int \mathbf{V}_{R_j} \Psi + \mathbf{p}_i \times \nabla_{p_i} \cdot \int \mathbf{V}_{R_j} \Psi) \prod_{i=2}^{N} d\mathbf{R}_i d\mathbf{p}_i.
\]

We now use the fact that one-body mobilities are diagonal and scalar \( \mu_{11}^{RR} = \mu_k^{k} \mathbf{I}, \mu_{11}^{TT} = \mu_k^{k} \mathbf{I} \) in an unbounded fluid flow. The one-body density, then satisfies,

\[
\frac{\partial c^{(1)}}{\partial t} = \nabla_{R_i} \cdot (\mu_k^{k} \nabla_{R_i} U - \mathbf{V}_i^{A}) c^{(1)} + k_b T \nabla_{R_i}^2 c^{(1)} + \mu_k^{k} \mathbf{p}_i \\
\times \nabla_{p_i} \cdot (c^{(1)} \mathbf{p}_i \times \nabla_{p_i} U - \Omega_i^{(1)} + k_b T \mathbf{p}_i \times \nabla_{p_i} c^{(1)})
\]

\[-N(N-1) \left( \nabla_{R_i} \cdot \int \mathbf{V}_{R_i} \Psi d\mathbf{R}_i d\mathbf{p}_2 + \mathbf{p}_i \times \nabla_{R_i} \cdot \int \mathbf{V}_{R_i} \Psi d\mathbf{R}_i d\mathbf{p}_2 \right) \prod_{i=3}^{N} d\mathbf{R}_i d\mathbf{p}_i.
\]

We now use the definition of \( c^{(2)} \) and the operator \( \mathcal{L}_{12} \) to obtain

\[
\frac{\partial c^{(2)}}{\partial t} = -\mathcal{L}_{12} c^{(2)} = -\int \mathcal{L}_{12} c^{(2)} d\mathbf{R}_2 d\mathbf{p}_2.
\]
small. For the same experiment, the angular speed of the colloids is \( \omega_r \sim 50 \text{ s}^{-1} \), which implies typical active torque is of order \( T_{\text{d}} \sim 10^{-16} \text{ Nm} \). The Brownian torques are of order \( \mathcal{O}(k_B T) \sim 10^{-21} \text{ Nm} \), which implies that the rotational Brown number \( B_R \) is of the order of \( 10^{-5} \). The radius of the green algae in [105] is \( \sim 3 \mu m \) and it swimming speed is \( 134 \mu m/s \). The Brown number for this experiment is then \( B_T \sim 10^{-3} \). In another set of experiment on bacteria [106, 107] and Janus colloids [18, 20], the size \( b \sim 1 \mu m \), and the speed \( v_r \sim 10 \mu m/s \). This leads to \( T_{\text{d}} \sim 10^{-13} \text{ Nm} \), which implies that the Brown number is \( B_T \sim 10^{-2} \). Thus Brown numbers \( B_T, B_R \to 0 \) for commonly studied active colloids.

5. Suspension stress

Landau and Lifshitz showed that the stress \( \Sigma^H \) in a suspension of force-free particles, averaged over scales large compared to the particle size, is given by \( \Sigma^H = 2\eta E + \Sigma^\ell \), where \( E \) is the macroscopic strain rate in a suspension of volume \( V \) and

\[
\Sigma^p = \frac{1}{V} \sum_i \left[ \int \left[ f^H(R_i + \rho_i) \rho_i - \eta \left\{ u(R_i + \rho_i) \dot{\rho}_i + \dot{\rho}_i u(R_i + \rho_i) \right\} \right] dS_i, \tag{29}
\]

is the contribution to the stress from the particles [75]. From the expansion of equation (3), it follows that

\[
\Sigma^p = \frac{1}{V} \sum_i \left[ b_{ij} \mathbf{e}_i^{(2)} - \frac{8\pi\eta b^2}{3} \mathbf{V}_i^{(2)} \right], \tag{30}
\]

The symmetric part of the particle contribution to the bulk stress was denoted by Batchelor as the stresslet [108]. To obtain the rheological response of the suspension, the above quantity has to be calculated in the presence of an externally imposed flow \( \mathbf{u}^\infty \). The irreducible tensorial harmonics coefficients of external flow

\[
\mathbf{V}_i^{(l)} = \frac{2l - 1}{4\pi b^2} \int \mathbf{u}^\infty(R_i + \rho_i) \mathbf{Y}^{(l-1)}(\dot{\rho}_i) dS_i, \tag{31}
\]

gives corresponding coefficients of the traction \( \mathbf{F}_i^{(l)} \) from the solution of linear system for external flow

\[
\mathbf{V}_i^{(l)} = \mathbf{G}^{(l)}_{ij} \cdot \mathbf{F}_j^{(l)}, \tag{32a}
\]

\[
\mathbf{F}_i^{(l)} = \mathbf{\Sigma}^{(l)}_{ij} = \mathbf{\Sigma}^{(l)}_{ij} + \mathbf{\Omega}^{(l)}_{ij} + \mathbf{\Psi}^{(l)}_{ij}, \tag{32b}
\]

Here \( \mathbf{\Sigma}^{(l)}_{ij} = \mathbf{P}^{(l)} \cdot \left[ \mathbf{G}^{(l)} \right] \cdot \mathbf{P}^{(l)} \) are generalized friction tensors encoding the response to external flow and its explicit expression is obtained by repeating the steps in section 2. The particle contribution to the suspension stress, then, is the sum of external and active contributions:

\[
V \Sigma^p = \sum_{l=1}^{\infty} \left[ \frac{2}{2l+1} \mathbf{e} \cdot \mathbf{\Gamma}_{ij}^{(2l)} + \frac{1}{2l+1} \mathbf{\Gamma}_{ij}^{(2l+2)} \right] \cdot \mathbf{V}_i^{(l)} = \sum_{l=1}^{\infty} \left[ \frac{2}{2l+1} \mathbf{e} \cdot \mathbf{\Gamma}_{ij}^{(2l)} + \frac{1}{2l+1} \mathbf{\Gamma}_{ij}^{(2l+2)} \right] \cdot \mathbf{\mathbf{V}_i^{(l)}}. \tag{33}
\]

All particle indices are summed over in the above. The friction tensors in an unbounded fluid, obtained in appendix A, can be used to estimate the active contribution to the suspension stress. The above expression must be statistically averaged over the position and orientation of the colloids to obtain the average stress in the suspension using distribution function \( \Psi(R^N, \mathbf{p}^N) \) defined in equation (26).

We now consider a force-free, torque-free suspension and assume that the leading contributions from the external flow produces a pure strain on the surface of the colloids. Thus, the first symmetric traceless moment of the external flow \( \mathbf{v}^\infty \) is most-dominant and is parametrize as \( V^{(2)} = bE \). Expression for \( \Sigma^p \) then reduces to

\[
\Sigma^p = \sum_i \left[ \frac{20\pi\eta b^4}{3} - \frac{28\pi\eta b^2}{3} \right] \mathbf{V}_i^{(0)} + O(\phi^2), \tag{34}
\]

where the first two terms are the leading order one-body contribution due to external flow and activity respectively. They are the \( O(\phi) \) contribution to the suspension stress, where \( \phi \) is the suspension volume fraction. At \( O(\phi) \), the average stress depends on the average of the irreducible dipole \( \langle \mathbf{V}_i^{(2)} \rangle_{\mathbf{p}} \) over the orientational distribution function. The orientational distribution function of spheres remains unchanged in a shear flow. In an isotropic suspension, therefore, the average \( \langle \mathbf{V}_i^{(2)} \rangle_{\mathbf{p}} \) vanishes and there is no contribution to the suspension stress at \( O(\phi) \) due to activity, as was first pointed out by Pedley and Ishikawa [109]. However, if the distribution is not isotropic, then the average of \( \langle \mathbf{V}_i^{(2)} \rangle_{\mathbf{p}} \) is proportional to \( \mathbf{V}_0^{(2)} \) and non-zero. In particular, if the first symmetric moment of the orientational distribution function is non-zero, then, the stress may increase or decrease depending on \( \mathbf{V}_0^{(2)} \) begin negative (for contractile colloids) or positive (for extensile colloids).
The exact relation between the suspension stress and the generalized friction tensor obtained above can be used to derive the $O(\phi^2)$ and higher corrections to the suspension stress. Such a calculation requires a careful regularization of conditionally convergent integrals [116] and will be presented in a future work.

6. Active pressure in external potential

From the forces, torques and stresslets on the colloids discussed in the previous sections, we now turn our attention to the mechanical pressure in the fluid. The active contribution to the pressure in the fluid is given by the second term in equation (15b). We consider a suspension of active colloids confined by an external spherical potential such that they are always inside a sphere of radius $R$. The confining surface is not a physical boundary and there is free motion of the fluid across it. We use the results of the sections 3 and 4 to calculate the fluid pressure on the confining surface due to the motion of the active colloids in the interior volume. This geometry is motivated by the confinement of bacteria inside a fluid drop with a porous interface.

We retain slip modes $\sigma = \pm 2a$ which correspond to the symmetric irreducible dipole and the chiral octupole. The first generates a long-ranged flow while the second produces self-rotation [35]. For simplicity, we choose these modes to be uniaxial, parametrized in terms of the orientation $\hat{p}$ of the colloid, as given in equation (5). The microscopic dynamics and the pressure distributions are sensitive to the sign of $V^3_{\text{fl}}$ as we shall see below. The fluid flow due to the chiral term, which decays as $r_{ij}^{-4}$, induces a net rotation of the system. The most dominant contribution to the fluid flow comes from the long-ranged dipolar flow, which decays as $r_{ij}^{-2}$. The fluid pressure decays as one power higher than the fluid flow. The dipole, thus, crucially determines the dynamic of active colloids in the spherical confinement and fluid pressure on the confining surface.

The confining potential $U^1(R_i) = k^2 \exp \left(\frac{1}{R_i - R} - \frac{1}{R_{\text{max}} - R} \right)$ for $R_i > R$ and $U^1(R_i) = 0$ otherwise. Here $R_{\text{max}}$ is chosen to be few particle radius more than $R$ in simulations and $k^2$ is the strength of the potential. The colloids also have an additional steric interaction, which is modeled by a short ranged repulsive potential, and depends on the separation $r_{ij} = R_i - R_j$ between the colloids. This potential is modeled using the WCA potential for separation $r_{ij} < r_{\text{min}}$, $U(r_{ij}) = \epsilon \left(\frac{\tan \theta_{ij}}{\tan \theta_{\text{c}}} \right)^{12} - 2\epsilon \left(\frac{\tan \theta_{ij}}{\tan \theta_{\text{c}}} \right)^{6} + \epsilon$, and zero otherwise [111]. Here $\epsilon$ is the strength of the potential. The specification of the slip and body forces complete the description of our model. We start the simulations with a completely random distribution of hard spheres positioned symmetrically about the origin [112]. The orientations of all the colloids are pointing along the $\hat{z}$-axis. The tendency of the hydrodynamic torques acting on the colloids to rotate their orientation is nullified by external torques $\mathbf{T}^3_i = T_0 (\hat{p}_i \times \hat{z})$ arising from bottom-heaviness. Thus the orientation of all the colloids remain along $\hat{z}$–axis for all times. We then study the collective dynamics and measure the pressure on the ‘confining’ surface.

The sign of the strength of the symmetric irreducible dipole, $V^3_{\text{fl}}$, is positive (negative) for an extensile (contractile) active colloid. We plot the fluid flow produced by a contractile and extensile colloid in first two panels in figure 2. The orientation of the colloids is assumed to be along the $\hat{z}$ direction. The source colloid is colored in green while white arrows on the tracer colloids show the direction of the force acting on them. The direction of the forces on these colloids gives a heuristic understanding of the dynamics in the spherical confinement as we explain below. The last two panels of figure 2 contain the fluid pressure due to contractile and extensile active colloid. It can be seen that the pressure along the equator is higher for for contractile colloids while pressure at the poles is higher for extensile colloids. Collective dynamics of contractile and extensile active colloids under spherical confinement follow from the flow field of the individual colloids.
A contractile colloid pushes the particles away in the plane perpendicular to the dipole axis, which leads to an instability in an initially isotropic suspension of colloids. The colloids final reach to a steady state which they organize into a continuously rearranging ‘oblate’ structure. The dipoles tend to push each other as far as possible but the spherical confinement coupled with short-ranged repulsion make them undergo rolls, which accounts for the continuous rearrangement of the structure. In the first two rows of figure 3, we show instantaneous configurations of contractile colloids and the state of the active pressure on the bounding sphere. The fluid pressure is then maximum on the equator of the confining sphere.

The last two rows of figure 3 show the corresponding configurations and fluid pressure on a confining sphere for extensile dipoles. The initial isotropic distribution of the colloids finally finds a steady state in a ‘prolate’ distribution of of extensile dipoles. The colloids continuously rearrange this structure as the dipolar flow tries to push them apart while the confining sphere holds them back. Moreover, the pressure is higher at the poles in this case.

To summarize, we have shown that the dynamics and fluid pressure measured in a suspension of extensile and contractile active colloids are completely different. Movie 1 and 2 of4 show the dynamics of contractile and extensile colloids, respectively, in a spherical confinement. The reported dynamics continues to hold for a random initial distribution with arbitrary separation between the colloids, and even at a very small initial volume fraction. This is because of the long-range (1/r^2) attractive forces, between the colloids, driving their aggregation. Here, we have assumed that the orientations of colloids are fixed by their bottom-heaviness. A more detailed study of the given system, where this condition is relaxed, will be pursued in a future work.

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4 See supplemental material at stacks.iop.org/jpcos/2/025025/mmedia for movies.
7. Dynamics in an optical lattice

The simplest system in which an interplay of non-uniform external fields, activity and Brownian motion can be studied is an active colloid confined in a three-dimensional harmonic potential. As this system is both experimentally realizable in optical trapping experiments [18, 113] and analytically tractable [42, 45, 94] it serves as the 'Ising model' of active colloidal physics. In this section, we use the Langevin equations to study the motion of active colloids confined in a square array of harmonic confining potentials. The principal question we focus on is the collective dynamics of the colloids and, in particular, their synchronization. The system we study can be realized experimentally in holographic tweezers. From estimates of the dimensionless groups presented in the previous section, it is clear that Brownian motion can be ignored in the first approximation. Accordingly, we neglect noise and study the mechanics of the system, postponing the study of its statistical mechanics to a future work.

We consider self-propelling, polar, achiral active colloids, with non-zero values of $V_i^A$, $V_i^{2(3)}$ and $V_i^{3(3)}$. With this choice, an isolated colloid translates with velocity $V_i^A = \nu_i \vec{p}_i$ while producing dipolar and quadrupolar flows of strengths proportional to $V_i^{2(3)}$ and $V_i^{3(3)}$ respectively [35, 42]. The centers of the $N$ traps are at $R_i^0$, arranged linearly or in a $\sqrt{N} \times \sqrt{N}$ square lattice. Each trap contains a single active colloid which feels a body force from that trap alone. The moment of force about the trap center is zero. Therefore, in a trap of stiffness $k$ centered at $R_i^0$

$$\mathbf{F}_i^p = -k(R_i - R_i^0), \quad \mathbf{T}_i^p = 0.$$  \hspace{1cm} (35)

First, we consider the dynamics ignoring hydrodynamic interactions. Then, force and torque balance give

$$-6\pi \eta b \cdot (\mathbf{V}_i - \nu_i \mathbf{p}_i) - k(R_i - R_i^0) = 0, \quad -8\pi \eta b^3 \Omega_i = 0.$$  \hspace{1cm} (36)

In the absence of hydrodynamic and Brownian torques, there is no angular velocity and the colloid translates in a direction $\mathbf{p}_i$ chosen by the initial condition. It is brought to rest at a radius $R^* = 6\pi \eta b \nu_i / k = \mathcal{A} \eta b$, when the propulsive and trap forces are balanced [42, 94]. Here $\mathcal{A}_i$ is the activity number defined in equation (22), which quantifies the ratio of active propulsive force to the passive confining force. The stationary state, without hydrodynamic interactions, is one in which all colloids are confined at a distance $R^*$ from the center of its trap and oriented radially outward in a direction that is, in general, different for each colloid.

This state is destabilized with hydrodynamic interactions [42, 94, 114] due to the torque induced by the flow of the $\nu \sigma = 1$s and $2$s modes [42]. The leading contributions to the hydrodynamic torque is

$$\mathbf{T}_i^H = -\gamma_{ij}^{RT} \cdot (\mathbf{V}_i - \nu_i \mathbf{p}_i) - \gamma_{ij}^{(R,2s)} \cdot \mathbf{V}_j^{(2s)},$$  \hspace{1cm} (37)

which, upon using the explicit forms of the generalized friction tensors from appendix $A$, is

$$\mathbf{T}_i^H = 8\pi \eta b^3 \left[ \frac{\tilde{r}_i^2}{r_i^3} \frac{k(R_i - R_i^0)}{6\pi \eta} + \frac{14}{r_i^3} \frac{(\mathbf{p}_i \cdot \tilde{r}_i)}{r_i^3} (\mathbf{p}_i \times \tilde{r}_i) V_0^{(2s)} b^2 \right] + O(r_i^{-3}).$$  \hspace{1cm} (38)

Here $r_i = R_i - R_i^0$ and the force balance equation has been used to eliminate $\mathbf{V}_i - \nu_i \mathbf{p}_i$ in favor of the trapping force. The hydrodynamic torque vanishes when the colloids are collinear and their orientations are along the line joining their centers. Thus stable states of rest are possible even in the presence of hydrodynamic interactions for specially chosen initial conditions [42]. In general, though, the interplay of self-propulsion, confinement, and hydrodynamic interactions produce steady states with continuous motion.

With this understanding, we now present numerical results for dynamics in a lattice of traps. In a linear lattice of traps, we find stable stationary states, reached irrespective of initial conditions, in which all colloids are oriented along the line joining the trap centers and at a confinement radius that is slightly altered from $R^*$ due to hydrodynamic interactions. We then study dynamics in a $3 \times 3$ square lattice of traps. The dynamics is shown in figure 4 and Movie 3 of see footnote 3. The initial condition is chosen to be a stable state in the absence of hydrodynamic interactions. We find that the particles at the center do not rotate by symmetry while particles on the left of this symmetry axis, rotate clockwise and particle on their right rotate counter-clockwise. This can be understood by estimating the hydrodynamic torques on each colloid. The dynamics can also understood intuitively from the flow field of figure 4. The colloids at an equal distance from the symmetric plane have synchronized dynamics. This leads to a long-range correlation between the colloids. In summary, (a) there is a rotational instability in the system if the traps centers are not collinear, and (b) dynamics is synchronized about an axis of symmetry in non-collinear traps.

In figure 5, we estimate the power dissipation for the synchronized and periodic motion of three active colloids in a triangular lattice of harmonic traps. The dynamics in a triangular lattice of harmonic traps is similar to that of the square lattice of traps. The colloid in the central trap has no rotational dynamics due to symmetry, while the one on the left rotates clockwise and the colloid on the right rotates counter-clockwise. The power dissipated in the system is minimum when the colloids are widely separated, while the dissipated power is
maximum when they are closer to each other. Thus, the first configuration of figure 5, corresponds to the minimum of power dissipation, while the second is the maximum, as indicated by the markers on the power dissipation curve, and the third configuration is an intermediate value.

The motion of many hydrodynamically interacting active colloids in a harmonic potential was first studied by Nash et al [94] using lattice Boltzmann simulations and then by singularity [114] and boundary integral [42] methods. There, the interplay between self-propulsion, confinement and hydrodynamically-induced reorientation yields orbits for a pair of confined particles [42]. These individual orbits coalesce to produce sustained convection in a confined suspension [42] producing the so-called ‘self-assembled pump’ [94]. In contrast, here we study the motion of many active colloids in a lattice of harmonic traps to uncover strikingly different dynamics.

8. Discussion and summary

By exploiting the linearity of slow viscous flow, as manifest in its boundary integral representation, we have derived linear relations between the coefficients of the force per unit area and the active slip in a suspension of active colloidal spheres. These linear relations we call the ‘generalized Stokes laws’ and the tensorial coefficients relating the force per unit area to the slip we call the generalized friction tensors. We have derived explicit expressions for these tensors in terms of the Green’s function of Stokes flow. The boundary integral representation provides the flow at any point in the bulk fluid, given the force per unit area and the active slip on
the fluid-colloid boundary. This leads to numerical methods that are more efficient than those that need to resolve the bulk fluid flow. [83, 94, 115].

From the generalized Stokes laws, we directly obtain the forces, torques, fluid flow, fluid pressure, power dissipation and suspension stress. Since forces and torques are the fundamental dynamical quantities in Newtonian or Langevin descriptions of particle dynamics, our contribution forms the basis for a microscopic theory of active suspension mechanics and statistical mechanics that conserves momentum in both the bulk fluid and at fluid-solid boundaries. The formalism is applied to experimentally realizable situations to derive testable predictions. It should be noted that in this work, the simulations are done in the mobility formulation and the leading terms of the forces and torques are calculated using the quick and transparent Jacobi iterative scheme. When greater accuracy is desired, conjugate gradients or other Krylov subspace methods may be used [70].

The Langevin and Smoluchowski description of active colloids are obtained in terms of mobility matrices and the propulsion tensors. The far-field limit of the mobility matrices are obtained in terms of the Green’s function of the Stokes flow while a lubrication approximation may be used when the colloids are close to each other [91]. The propulsion tensors are obtained in terms of the lubrication-corrected mobilities and the friction tensors. Thus we account for both the far-field hydrodynamic interactions, to any order of desired accuracy, and the near-field lubrication interactions. The Galerkin discretization of the boundary integral equation provides most accurate results for smooth boundaries, like spheres, for least number of unknowns and preserves the self-adjointness of the problem [63, 65]. Dynamic simulation of hundreds of thousand of active colloids on a multi-core computational architectures is possible.

In this work, we assume a spherical particle with active slip on its surface, which is then expanded in a Galerkin basis to obtain the force per unit area. Thus, any generic mechanism generating the active velocity can be modeled in our approach. Typically, the slip mechanism for synthetic active colloids is phoretic and then, we do need to solve separately for a concentration field [116]. Here, we have assumed that the non-hydrodynamic parts of the problem have been solved separately. This assumption requires the decoupling of advection and diffusion and, therefore, is restricted to low Péclet numbers.

It is useful to compare the results presented here with existing results for hydrodynamic interactions of many spheres. Excluding the contribution from active slip and truncating the Galerkin expansion at \( f = 2 \), results in the method of computing far-field hydrodynamic interactions in the so-called FTS Stokesian dynamics method of Brady and coworkers [90, 91]. This latter method ignores the entire \( f = 3 \) contribution which decays as \( r^{-3} \) for unbounded flow and is long-ranged. This low-order truncation has been extended up to 7th order by Ichiki [57]. The method uses different bases for the expansion of the force per unit area and the surface velocity and several elaborate transformations are necessary to obtain a full-rank linear system. In contrast, our choice of identical basis for both force per unit area and active slip automatically yields a full-rank linear system. In our basis, harmonics indexed by \( l \) produce bulk flows that decay as \( r^{-1} \), a simplicity that is absent in the bases used by Cichocki [92] and Ichiki [57]. For active colloids, earlier work closest in spirit to ours is that of Ishikawa et al [117–119] where axisymmetric slip velocities, truncated to the first two non-trivial modes, are considered. The far-field and near-field hydrodynamic interactions are obtained, respectively, in superposition and lubrication approximations. In contrast, we include the most general form of the slip and use an irreducible basis function for Galerkin discretization which gives a systematic way of evaluating hydrodynamic interactions to any desired order of accuracy. Active colloids can be studied by other models like the force-coupling method, though only in dilute limit, as the distinction between the interior and exterior of a colloid is notional in the method [115]. We refer the readers to recent reviews on low Reynolds number flows for a more comprehensive list of work in the field [120–123].

The present paper focuses on the deterministic parts of the problem, though the stochastic equations have been presented. In a future work, we will explore the stochastic aspects more fully, using the Langevin and Smoluchowski description derived in this paper. The formalism can also be extended to study fluctuations in chains of active particles [83–85, 124]. Applications of our method to collective phenomena in magnetotactic colloids and to active rheology will be presented in forthcoming work as will be the extension to ellipsoidal particles.

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Appendix A. Iterative solution for generalized friction tensors

In this section, we derive iterative solutions of the generalized friction tensors using the properties of the tensorial spherical harmonics \( Y^{(l)}(\hat{\rho}) \). The first four \( Y^{(l)} \)s are

\[
Y^{(0)} = 1, \quad Y^{(1)}_\alpha = \hat{\rho}_\alpha, \quad Y^{(2)}_\alpha = \left( \hat{\rho}_\alpha \hat{\rho}_3 - \frac{\delta_{\alpha 3}}{5} \right), \quad Y^{(3)}_{\alpha\beta\gamma} = \left( \hat{\rho}_\alpha \hat{\rho}_\beta \hat{\rho}_\gamma - \frac{1}{5} \{ \hat{\rho}_\alpha, \hat{\rho}_\beta, \hat{\rho}_\gamma \} + \hat{\rho}_\alpha \delta_{\beta\gamma} + \hat{\rho}_\beta \delta_{\alpha\gamma} + \hat{\rho}_\gamma \delta_{\alpha\beta} \right). \tag{A1}
\]

Tensorial spherical harmonics are orthogonal basis function on the surface of a sphere

\[
\frac{1}{4\pi b^2} \int Y^{(l)}(\hat{\rho}) Y^{(l')} (\hat{\rho}) dS = \delta_{ll'} \frac{4l + 1}{2l + 1} \Delta^{(l)}, \tag{A2}
\]

The expansion of the velocity in this basis has been given in the main text. The orthogonality of the basis functions can be used to obtain the expansion coefficients in terms of surface integrals of traction and velocity as \[6, 35]\]

\[
F^{(l)}_i = \frac{1}{(l - 1)!(2l - 3)!!} \int f(R_i + \rho_j) Y^{(l-1)}(\hat{\rho}) dS_j, \quad V^{(l)}_i = \frac{2l - 1}{4\pi b^2} \int v^A(R_i + \rho_j) Y^{(l-1)}(\hat{\rho}) dS_j. \tag{A3}
\]

The iterative solutions for generalized friction tensors are obtained in terms of the matrix elements of the linear system equation \( (10) \). The key idea to obtain the exact solution of the matrix elements is to Taylor expand the Green’s function about the center of the sphere, express the \( s \) function of the traction and velocity \( c \)ients as we explain now. We use the projection operator \( \mathcal{P} \), which, written in terms of the irreducible tensorial \( c \)ients of the traction and velocity, projects the traction and slip \( c \)ients as

\[
G^{(j)}_{ji}(R_i, R_j) = \begin{cases} 
\frac{(2l - 1)(2l' - 1)}{4\pi b^2} \int Y^{(l-1)}(\hat{\rho}) G(R_i + \rho_j, R_j) Y^{(l'-1)}(\hat{\rho}) dS_i dS_j; & j = i, \\
\frac{l + l' - 2}{4\pi b^2} \mathcal{P}^{(j)}_{ji}; & j \neq i,
\end{cases} \tag{A4}
\]

\[
K^{(j)}_{ji}(R_i, R_j) = \begin{cases} 
\frac{(2l - 1)}{4\pi b^2} \int Y^{(l-1)}(\hat{\rho}) K(R_i + \rho_j, R_j, \rho_j) \cdot \hat{\rho}_j Y^{(l'-1)}(\hat{\rho}) dS_i dS_j; & j = i, \\
\frac{l + l' - 2}{4\pi b^2} \mathcal{P}^{(j)}_{ji}; & j \neq i, 
\end{cases} \tag{A5}
\]

where \( \mathcal{P}^{(j)}_{ji} \) is an operator which encodes the finite size of the sphere \[35, 42\].

We now write the linear system of equations, equation \( (10) \), in terms of the irreducible modes of velocity and traction \( c \)oefficients as we explain now. We use the projection operator \( \mathcal{P}^{(j)} \), defined in equation \( (4) \), which projects the \( \sigma \) component of the traction and velocity \( c \)oefficients, to evaluate the generalized friction tensors \( \gamma^{(j)}_{ij} \). The linear system of equations is then obtained in terms of these irreducible traction and velocity \( c \)oefficients as

\[
\frac{1}{2} V^{(j)}_{ij} = -G^{(j)}_{ji}(R_i, R_j) \cdot \mathcal{P}^{(j)}_{ij} + K^{(j)}_{ji}(R_i, R_j) \cdot V^{(j)}_{ij}. \tag{A6}
\]

Here the first two modes of \( V^{(j)} \) include rigid body motion such that \( V^{(1)} = V - \Omega \cdot \mathcal{D} \) and \( V^{(2)} = b (\Omega - \Omega \cdot \mathcal{D}) \). Having obtained the matrix elements and the linear system in the irreducible form, we, now, provide a constructive solution for the generalized friction tensor, given the matrix elements of the single-layer and double-layer integrals. The solution employs the classical Jacobi iteration, which is the mathematical basis of the physically motivated method of reflections first used by Smoluchowski \[68\]. Other methods with better convergence properties are an avenue for further study. The generalized friction tensor is the solution to the linear system in equation \( (10) \), which, written in terms of the irreducible tensorial \( c \)oefficients and arranged into standard form, is

\[
\gamma^{(j)}_{ij} = \left[ G^{(j)}_{ji}(R_i, R_j) \right] \cdot \left( \frac{1}{2} I^{(j)}_{ij} + K^{(j)}_{ji} \right) = \left[ G^{(j)}_{ji}(R_i, R_j) \right] \cdot \left( B^{(j)}_{ij} \right). \tag{A7}
\]
The right hand side of the linear system consists of the known slip coefficients. Jacobi’s solution of the friction tensor at the nth iteration, for this problem, is then

$$\langle \gamma_{ij}^{(n)}(\sigma, \varphi') \rangle = \frac{1}{G_{k}^{(n)}} \left[ B_{ij}^{(n)}(\sigma, \varphi') - \sum_{k} G_{ij}^{(n)}(\sigma, \varphi') \cdot \langle \gamma_{ij}^{(n-1)}(\sigma, \varphi') \rangle \right]. \quad (A8)$$

The second term above is proportional to the solution obtained at the \((n - 1)\)-th iteration. Here the prime over the summation indicates that the diagonal term, corresponding to \((i = j = k)\) and \((\sigma = \varphi')\) is not included in the summation as per the definition of the Jacobi iteration [126]. The iteration must begin with an initial guess for the solution. As the linear system is diagonally dominant, the one-body solution is always a good starting guess. Thus, explicit expressions for the generalized friction tensors are obtained in terms of a Green’s function of the Stokes flow. Thus the solution is applicable to arbitrary geometries of Stokes flow. A list of Green’s functions of Stokes equation is given in table 2.

### Appendix B. Generalized friction tensors in an unbounded domain

The one-body solution of the linear system can be calculated exactly in an unbounded domain. The generalized friction matrix is fully diagonal and the Jacobi iteration trivially converges. The solution of the linear system is then

$$F_{i}^{(0)} = -\frac{1}{G_{ii}^{(0)}} \gamma_{ii}^{(0)} \mathbf{v}_{i}^{(0)} = -\frac{1}{G_{ii}^{(0)}} \gamma_{ii}^{(0)} \mathbf{v}_{i}^{(0)}, \quad (B1)$$

The first three terms of this exact solution for one-body system and zeroth order guess for a many-body system are

$$F_{i}^{(1)} = -6\pi \eta b (V_{i} - \mathbf{v}_{i}^{(1)}), \quad T_{i}^{(1)} = -8\pi \eta b^{3} (\Omega_{i} - \mathbf{v}_{i}^{(1)}), \quad F_{i}^{(2)} = -\frac{20\pi \eta b}{3} \mathbf{v}_{i}^{(2)} \quad (B2)$$

Implying that, under force-free, torque-free conditions colloids translate and rotate independently with linear and angular velocities determined by equation (6b). This exact result for one-body motion was obtained previously by several authors from a direct solution of the Stokes equation [21, 32, 33], through the use of the reciprocal identity [55] and from the boundary integral approach [35, 42].

Hydrodynamic interactions appear at first iteration, represented by the off-diagonal \((i \neq j)\) terms in the generalized friction tensor. The first order approximation to friction tensors in term of a Green’s function \(G\) of Stokes flow is

$$(\gamma_{ij}^{(1)}_{TT}) = -\gamma_{ij}^{T} \mathbf{G}^{T}_{ij} \mathbf{G}, \quad (\gamma_{ij}^{(1)}_{FR}) = -\frac{1}{2} \gamma_{ij}^{F} \mathbf{G} \times \mathbf{G},$$

$$(\gamma_{ij}^{(1)}_{TR}) = -\frac{1}{2} \gamma_{ij}^{T} \mathbf{G} \times \mathbf{G}, \quad (\gamma_{ij}^{(1)}_{FF}) = -\frac{1}{4} \gamma_{ij}^{F} \mathbf{G} \times \mathbf{G},$$

$$(\gamma_{ij}^{(1)}_{R2}) = -\frac{28\pi \eta b^{2}}{2} \gamma_{ij}^{T} \mathbf{G} \times \mathbf{G}, \quad (\gamma_{ij}^{(1)}_{R3}) = -\frac{28\pi \eta b^{2}}{6} \gamma_{ij}^{T} \mathbf{G} \times \mathbf{G},$$

$$(\gamma_{ij}^{(1)}_{R3}^{(2)}) = -\frac{13\pi \eta b^{3}}{9} \gamma_{ij}^{T} \mathbf{G} \times \mathbf{G}, \quad (\gamma_{ij}^{(1)}_{R3}^{(3)}) = -\frac{13\pi \eta b^{3}}{18} \gamma_{ij}^{T} \mathbf{G} \times \mathbf{G},$$

$$(\gamma_{ij}^{(1)}_{R3}^{(4)}) = \frac{4\pi \eta b^{3}}{5} \gamma_{ij}^{T} \mathbf{G} \times \mathbf{G}, \quad (\gamma_{ij}^{(1)}_{R3}^{(5)}) = 0,$$

$$(\gamma_{ij}^{(1)}_{R3}^{(6)}) = \frac{121\pi \eta b^{4}}{10} \gamma_{ij}^{T} \mathbf{G} \times \mathbf{G},$$

$$(\gamma_{ij}^{(1)}_{R3}^{(7)}) = \frac{121\pi \eta b^{4}}{20} \gamma_{ij}^{T} \mathbf{G} \times \mathbf{G}.$$ 

Here \(\gamma^{T} = 6\pi \eta b, \gamma^{R} = 8\pi \eta b^{3}. \) The friction tensor corresponding to \(l\sigma = 2t\) are obtained in terms of the pressure Green’s function as

$$\gamma_{ij}^{(2t,TT)} = P(R_{i}, R_{j}), \quad \gamma_{ij}^{(2t,FR)} = \frac{1}{2} \frac{(t - 1)}{(2t - 1)} \nabla_{ij}^{(1)} P(R_{i}, R_{j}), \quad \gamma_{ij}^{(2t,RR)} = 0, \quad \gamma_{ij}^{(2t,TT)} = 0. \quad (B3)$$

At this level, the hydrodynamic interactions are pairwise-additive and add to the one-body solutions for the force and torque obtained at the zeroth iteration. The resulting force and torque on the active colloids are geometrically isotropic and hydrodynamically anisotropic due to the active slip. An intuitive understanding of the same can be gained from figure 6. The first panel shows the fluid flow around a passive particle which is determined solely by the sum of the body forces. Changing the particle orientation does not change the fluid and, therefore, produces no change to the forces, shown by the white arrows, on the two test particles. The second panel plots the flow around an active particle where the slip contains the modes \(l\sigma = 2s\) and \(l\sigma = 3t. \) In the third panel, the active particle is rotated clockwise by \(\pi/2,\) without any change in its position. The forces are now
different, even though there has been no changes in relative positions. Similar considerations apply for the torque as the reader can easily verify.

The above analysis is then used to to obtain explicit forms of the mobility matrices and the propulsion tensors. Their leading order forms, for \( \mathbf{R}^{\alpha} \), are

\[
\begin{align*}
\mathbf{\mu}^{TT}_{ij} & = \mathbf{F}^{T}_{j} \mathbf{J}^{T}_{i} \mathbf{G}, \\
\mathbf{\mu}^{TR}_{ij} & = \frac{1}{2} \mathbf{\nabla}_{R_{i}} \times \mathbf{G}, \\
\mathbf{\mu}^{RT}_{ij} & = \frac{1}{4} \mathbf{\nabla}_{R_{i}} \times (\mathbf{\nabla}_{R_{i}} \times \mathbf{G}), \\
\mathbf{\pi}^{(2,2)}_{ij} & = \frac{28 \pi \eta b^{2}}{3} \mathbf{F}^{T}_{i} \mathbf{F}^{T}_{j} \mathbf{\nabla}_{R_{i}} \mathbf{G}, \\
\mathbf{\pi}^{(2,3)}_{ij} & = \frac{13 \pi \eta b^{3}}{9} \mathbf{\nabla}_{R_{i}} (\mathbf{\nabla}_{R_{i}} \times \mathbf{G}), \\
\mathbf{\pi}^{(3,3)}_{ij} & = \frac{4 \pi \eta b^{3}}{5} \mathbf{\nabla}_{R_{i}} \mathbf{G}, \\
\mathbf{\pi}^{(3,4)}_{ij} & = \frac{12 \pi \eta b^{4}}{10} \mathbf{\nabla}_{R_{i}} \mathbf{\nabla}_{R_{i}} (\mathbf{\nabla}_{R_{i}} \times \mathbf{G}), \\
\mathbf{\pi}^{(4,4)}_{ij} & = \frac{12 \pi \eta b^{4}}{20} \mathbf{\nabla}_{R_{i}} \times (\mathbf{\nabla}_{R_{i}} \mathbf{\nabla}_{R_{i}} (\mathbf{\nabla}_{R_{i}} \times \mathbf{G})).
\end{align*}
\]

Appendix C. Boundary integrals for fluid velocity and pressure

Here, we provide explicit expression for the boundary integrals in equation (14). These are given in terms of a Green’s function of Stokes equation, the corresponding pressure vector, and their derivatives \[ \mathbf{G}^{(l)}(\mathbf{r}, \mathbf{R}) = \frac{4 \pi \eta b^{l}}{(l - 2)!(2l - 1)!!} \mathbf{F}_{j}^{l} \mathbf{J}^{(l-1)}_{i} \mathbf{G}(\mathbf{r}, \mathbf{R}), \quad \mathbf{K}^{(l)}(\mathbf{r}, \mathbf{R}) = \frac{2 \pi b^{l-1}}{(l - 2)!(2l - 1)!!} \mathbf{\nabla}^{(l-1)}_{i} \mathbf{P}(\mathbf{r}, \mathbf{R}). \] (C1a) (C1b)

We use the above expression, and the generalized friction tensors, to obtain explicit expression of the fluid flow and pressure. Their explicit expressions, in terms of knowns, are

\[
\begin{align*}
\mathbf{u}(\mathbf{r}) & = -\mathbf{G}^{(1)}_{j}(\mathbf{r}, \mathbf{R}) \cdot \mathbf{F}^{H}_{j} - \mathbf{G}^{(2,0)}_{j}(\mathbf{r}, \mathbf{R}) \cdot \mathbf{T}^{H}_{j} - \sum_{l=2}^{\infty} \sum_{l'=2}^{\infty} \mathbf{G}^{(l',0)}_{j}(\mathbf{r}, \mathbf{R}) \cdot \gamma^{(l',0, l')}_{ij} - \eta \mathbf{K}^{(l,0)}_{j}(\mathbf{r}, \mathbf{R}) \cdot \mathbf{V}^{(l,0)}_{j}, \\
\mathbf{p}(\mathbf{r}) & = -\mathbf{P}^{(1)}_{j}(\mathbf{r}, \mathbf{R}) \cdot \mathbf{F}^{H}_{j} - \sum_{l=2}^{\infty} \mathbf{P}^{(l,0)}_{j}(\mathbf{r}, \mathbf{R}) \cdot \gamma^{(l,0, l)}_{ij} - \mathbf{Q}^{(0)}_{j}(\mathbf{r}, \mathbf{R}) \cdot \mathbf{V}^{(0)}_{j}.
\end{align*}
\]

These are then used to obtain the irreducible expressions of the fluid flow and the pressure in equation (15). The tensors relating the irreducible slip modes to fluid velocity and pressure are
\[ \Pi^{(\alpha \beta)} = -\left( G^{(\alpha \beta)}_i(r, R_i) \cdot \gamma^{(\alpha \beta)}_j(r, R_j) \right), \quad \Lambda^{(\alpha \beta)} = -\left( P^{(\alpha \beta)}_i(r, R_i) \cdot \gamma^{(\alpha \beta)}_j(r, R_j) - Q^{(\alpha \beta)}_j(r, R_j) \right), \]  

In the above expressions for the fluid flow and pressure, we have used the definition of projection operators equation (4) to define the irreducible parts of the boundary integrals as:  

\[ G^{(\alpha \beta)}_i = P^{(\alpha \beta)}_i, \quad K^{(\alpha \beta)}_j = P^{(\alpha \beta)}_j, \]  

etc.

Appendix D. Symbols and notations

A list of symbols and notations used in the paper is given below. The first column describes quantities which have particle indices while those in the second column do not have particle indices. The list has been ordered based on the first appearance of these symbols in the main text. We use abbreviate rigid body motion as RBM and boundary integral equation as BIE.

- \( F_i^H \) hydrodynamic forces on the \( i \)th colloid
- \( F_i^P \) body force on the \( i \)th colloid
- \( \hat{F}_i \) Brownian force on the \( i \)th colloid
- \( T_i^H \) hydrodynamic torque on the \( i \)th colloid
- \( T_i^P \) body torque on the \( i \)th colloid
- \( \hat{T}_i \) Brownian torque on the \( i \)th colloid
- \( V_i \) translational velocity of the \( i \)th colloid
- \( \Omega_i \) angular velocity of the \( i \)th colloid
- \( V_i^A \) active translational velocity of the \( i \)th colloid
- \( \Omega_i^A \) active angular velocity of the \( i \)th colloid
- \( R_i \) coordinate of the centre of the \( i \)th colloid
- \( \rho_i \) orientation of the \( i \)th colloid
- \( v \) boundary velocity on a colloid
- \( \nu^A \) active slip at the surface of the \( i \)th colloid
- \( f \) force per unit area (or the traction)
- \( r_i \) a point on the surface of the \( i \)th colloid
- \( r_{ij} \) \( R_i - R_j \), separation between colloids
- \( S_i \) surface of the \( i \)th colloid
- \( \rho_i \) radius vector of the \( i \)th colloid
- \( Y^{(l)} \) \( l \)th tensorial spherical harmonics
- \( V^{(l)} \) \( l \)th coefficient of slip expansion
- \( F^{(l)} \) \( l \)th coefficient of traction expansion
- \( P^{(\alpha \beta)} \) Projection operator
- \( F_i^{(\alpha \beta)} \) symmetric irreducible part of \( F_i^{(l)} \)
- \( F_i^{(\alpha \beta)} \) antisymmetric part of \( F_i^{(l)} \)
- \( F_i^{(l)} \) trace of \( F_i^{(l)} \)
- \( \mu^{\alpha \beta} \) mobility matrix for \( \alpha, \beta = T, R \)
- \( \pi^{(\alpha \beta)} \) propulsion tensors
- \( \gamma^{(\alpha \beta)} \) generalized friction tensors
- \( G_i^{(l)} \) single-layer integral for fluid flow
\( \mathbf{K}_i^{(l)} \) double-layer integral for fluid flow
\( \mathbf{P}_i^{(l)} \) single-layer integral for fluid pressure
\( \mathbf{Q}_i^{(l)} \) double-layer integral for fluid pressure
\( \mathbf{G}_{ij}^{(l,F)} \) single-layer matrix elements
\( \mathbf{K}_{ij}^{(l,F)} \) double-layer matrix elements
\( \mathcal{F}_i \) operator encoding finite size of colloids
\( b \) radius of the colloid
\( N \) number of colloids
\( \eta \) fluid viscosity
\( \sigma \) fluid stress
\( u \) fluid velocity
\( p \) fluid pressure
\( u^\infty \) externally imposed flow
\( G \) a Green’s function of Stokes equation
\( K \) a Stress function of Stokes equation
\( G \) single-layer operator of the BIE
\( K \) double-layer operator of the BIE
\( \xi \) thermal force acting on the fluid
\( V \) volume of the fluid
\( \phi \) volume fraction
\( \dot{\mathcal{E}} \) power dissipation in the fluid
\( A_T \) translational Activity number
\( A_R \) rotational Activity number
\( B_T \) translational Brown number
\( B_R \) rotational Brown number
\( \Psi \) distribution function for colloids
\( \rho^{(n)} \) \( n \)-body density
\( \Sigma^H \) hydrodynamic suspension stress
\( \Sigma^p \) particle contribution to the stress
\( E \) macroscopic strain rate
\( \Delta^{(l)} \) \( F_i^{(l)} = \Delta^{(l)} \cdot F_i^{(l)} \)
\( \varepsilon \) the Levi-Civita tensor
\( \delta \) the Kronecker delta
\( I \) identity tensor
\( v_s \) self-propulsion speed of the colloid
\( \omega_s \) self-rotation speed of the colloid
\( U \) external potential
\( \epsilon \) strength of the WCA potential
\( k \) stiffness of the harmonic trap trap
\( k^c \) strength of the spherical confinement
\( \tau \) rotational time scale \( \tau_{0}^{(\text{rot})} / b \)
\( \tau_{r} \) rotational time scale in trap \( 8\pi \nu b / k \)
\( k_B \) Boltzmann constant
\( T \) temperature

**ORCID iDs**

Rajesh Singh @ https://orcid.org/0000-0003-0266-9691

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