Dispersion of active particles of arbitrary shape

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Stochastic equations for the 3D active Brownian particle (ABP) model:

$$\dot{x} = U \, e + \left\{ \sqrt{2D_\parallel} \, P^\parallel(e) + \sqrt{2D_\perp} \, P^\perp(e) \right\} \cdot \dot{w}_1$$

$$\dot{e} = -2D_r \, e + \sqrt{2D_r} \, e \times \dot{w}_2.$$  

- translational noises $D_\perp$ and $D_\parallel$ along and perpendicular to the direction of swimming $e$;
- the rotational noise $D_r$ affects the swimming direction;
- diffusivities are related to particle mobility $\times k_B T$;
- $w_i(t)$ are independent standard Wiener processes (5 total);
- angular drift $-2D_r \, e$ ensures unit length $e$.

[Peruani & Morelli (2007); van Teeffelen & Löwen (2008); Baskaran & Marchetti (2008); Romanczuk & Schimansky-Geier (2011); Romanczuk et al. (2012); Kurzthaler et al. (2016); Kurzthaler & Franosch (2017); Ai et al. (2013); Solon et al. (2015); Zöttl & Stark (2016); Wagner et al. (2017); Redner et al. (2013); Stenhammar et al. (2014); Chen & Thiffeault (2021)]
An ABP meanders around, and for long times there is a well-known formula for its effective diffusivity:

$$D_{\text{eff}} = \frac{\|U\|^2}{6D_r}$$

This is a very useful dispersion result that can be measured experimentally.

The ABP model is not quite general: only one vector $e$ is used to denote the orientation. This is fine if the particle has hydrodynamic axial symmetry.

Goal: investigate the general particle and derive $D_{\text{eff}}$.

Secondary goal: avoid using Euler angles or similar parametrization!
For an arbitrary body, instead of $e$ we use an orthogonal matrix $Q$.

In general, need 6 coordinates: $x = (x_1, x_2, x_3)$ and $\phi = (\phi_1, \phi_2, \phi_3)$.

- $\phi$ is some vector of coordinates for $SO(3)$ specifying the particle orientation as $Q(\phi)$ (e.g., Euler angles, quaternions).
- **Notation:** A hat $\hat{\cdot}$ will indicate a matrix or vector with 6 rows and/or columns, denoting position and angles:

  e.g., $\hat{x} = (x, \phi)$
In Stokes flow, force $f$ and torque $\tau$ are \textit{linearly related} to the particle’s velocity $u$ and angular velocity $\omega$:

$$
\begin{pmatrix}
  u \\
  \omega
\end{pmatrix} = \widehat{M} \cdot \begin{pmatrix}
  f \\
  \tau
\end{pmatrix}
$$

$$
\widehat{M} := \begin{pmatrix}
  M_{xx} & M_{x\phi} \\
  M_{\phi x} & M_{\phi\phi}
\end{pmatrix}.
$$

- The $6 \times 6$ symmetric matrix $\widehat{M}$ is called the \textit{grand mobility matrix}.
- $\widehat{M}$ is symmetric, so $M_{x\phi} = M_{\phi x}^\top$.
- Torque (and thus $\widehat{M}$) is defined \textit{w.r.t.} a \textit{reference point}.
The coupling matrix

\[ \hat{M} = \begin{pmatrix} M_{xx} & M_{x\phi} \\ M_{\phi x} & M_{\phi\phi} \end{pmatrix}. \]

- There is a unique reference point called the center of hydrodynamic reaction for which \( M_{x\phi} = M_{x\phi}^\top \) [e.g., Happel & Brenner (1983)].
- If the center of mass differs from the center of reaction, then \( M_{x\phi} \) cannot be symmetric.
- In particular, it cannot be zero.
- Let’s call such a particle wobbly: \( M_{x\phi} \neq M_{x\phi}^\top \).
- Even a sphere with nonuniform density is wobbly.
Hydrodynamic chirality

\[ M_{x\phi} = 0 \]

\[ M_{x\phi} \neq 0 \]

\[ \hat{M} = \begin{pmatrix} M_{xx} & M_{x\phi} \\ M_{\phi x} & M_{\phi\phi} \end{pmatrix}. \]

• If the coupling \( M_{x\phi} \) is nonzero for any choice of reference point, a particle is **hydrodynamically chiral**.
• This can coincide with **geometric chirality**, as above.
Overdamped stochastic dynamics

Kinematics:

\[ \dot{x} = u, \quad \dot{\phi} = \mathbb{L} \cdot \omega. \]

The tensor \( \mathbb{L} \) depends on the specific coordinate representation of \( SO(3) \). [For subtle reasons, it makes sense to choose the center of mass for \( x \).]

Write as deterministic plus stochastic parts:

\[
\begin{pmatrix}
\dot{x} \\
\dot{\phi}
\end{pmatrix} = \begin{pmatrix} 1 & 0 \\ 0 & \mathbb{L} \end{pmatrix} \cdot \left\{ \begin{pmatrix} U \\ \Omega \end{pmatrix} + \sqrt{2\mathbb{D}} \cdot \dot{\mathbb{w}} \right\}
\]

(General ABP model)

where \( \dot{\mathbb{w}} \) is a vector of Wiener increments with correlation

\[
E\{\dot{\mathbb{w}}(t) \otimes \dot{\mathbb{w}}(s)\} = \delta(t - s) \mathbb{1}.
\]

The fluctuation-dissipation theorem (Stokes–Einstein) implies

\[
\hat{\mathbb{D}} = k_B T \hat{\mathbb{M}}.
\]
Remark on the overdamped limit

When passing from the underdamped (Langevin) dynamics to the overdamped limit, there is a well-known stochastic drift \cite{LauLubensky2007, FaragoGrønbech-Jensen2014, FaragoGrønbech-Jensen2014, Farago2017}:

\[
\hat{U} = \begin{pmatrix} U \\ \Omega \end{pmatrix} = \begin{pmatrix} U_{\text{swim}} \\ \Omega_{\text{swim}} \end{pmatrix} + \nabla \hat{x} \cdot \hat{D}
\]

where \( \hat{x} = (x, \phi) \) and

\[
\nabla \hat{x} \cdot \hat{D} = \begin{pmatrix} \nabla x \cdot D_{xx} + \nabla \phi \cdot D_{\phi x} \\ \nabla x \cdot D_{x\phi} + \nabla \phi \cdot D_{\phi \phi} \end{pmatrix}
\]

with \( \nabla \phi \) defined appropriately for SO(3) \( (\nabla \phi := L^\top \cdot \partial \phi) \).

For a free particle in a homogeneous medium \( (\hat{D} = \hat{Q} \cdot \hat{D}^{(0)} \cdot \hat{Q}^\top) \),

\[
\nabla \hat{x} \cdot \hat{D} = \begin{pmatrix} \epsilon : D_{\phi x} \\ 0 \end{pmatrix}, \quad (\epsilon)_{ijk} = \epsilon_{ijk},
\]

which vanishes when the centers of mass and reaction coincide.
Our earlier General ABP model:

\[
\frac{d}{dt} \hat{x} = \begin{pmatrix} 1 & 0 \\ 0 & L \end{pmatrix} \cdot \left\{ \hat{U} + \sqrt{2\mathbb{D}} \cdot \hat{w} \right\}
\]

can be turned into a Fokker–Planck equation for the probability density \( p(\hat{x}, t) = p(x, \phi, t) \):

\[
\partial_t p = -\nabla \hat{x} \cdot \left\{ \hat{U} p - \nabla \hat{x} \cdot (\mathbb{D} p) \right\}.
\]

This equation is hard to solve, being a 6-dimensional PDE.

Our next task is to get rid of the angular dependence by passing to the long time / large scale limit.
For a small parameter \( \delta \), effect the **diffusive rescaling**

\[
\partial_t \rightarrow \delta^2 \partial_t, \quad \nabla_x \rightarrow \delta \nabla_x, \quad \nabla \phi \rightarrow \nabla \phi.
\]

(There is no such things as a “large” angle.)

As usual, we expand

\[
p = p_0 + \delta p_1 + \delta^2 p_2 + \cdots.
\]
The order $\delta^0$ part of the differential operator in the FP equation is

$$\mathcal{L} p = \nabla \phi \otimes \nabla \phi : (D_{\phi \phi} p) - \nabla \phi \cdot (\Omega p).$$

At order $\delta^0$ we must solve

$$\mathcal{L} p_0 = 0.$$ 

To keep things simple, assume $\mathcal{L} p_0 = 0$ only has solutions which are independent of $\phi$ (isotropic). Then we may write

$$p_0 = P(\mathbf{x}, t),$$

that is, our leading-order solution is some as-yet unknown function of the large-scales variables $\mathbf{x}$ and $t$. 

(i.e., at long times our particle randomizes its orientation completely.)
At the next order in $\delta$, we must solve

$$\mathcal{L} p_1 = \nabla_x \cdot (V P), \quad V := U - 2\nabla \phi \cdot D \phi x.$$ 

By linearity, if we can solve

$$\mathcal{L} \chi = V$$

cell problem for $\chi$

then we can write

$$p_1 = \nabla_x \cdot (V \chi)$$

Let’s assume for now that we’ve solved this.
As is common in this type of problem, we don’t actually need to solve for $p_2$. We just need to apply a solvability condition at order $\delta^2$ to obtain a heat equation

$$\partial_t P = \nabla_x \otimes \nabla_x : (D_{\text{eff}} P)$$

where the effective diffusivity is

$$D_{\text{eff}} = \langle D_{xx} \rangle - \text{sym} \langle U \otimes \chi \rangle$$

and angle brackets denote an average over $SO(3)$.

So, in principle we can find the effective diffusivity, as long as we can solve $\mathcal{L}\chi = V$. 

Order $\delta^2$
The elliptic problem

We want to solve $\mathcal{L} \chi = V$:

$$\mathcal{L} \chi = \nabla \phi \otimes \nabla \phi : (\mathbb{D}_{\phi \phi} \chi) - \nabla \phi \cdot (\Omega \chi) = V$$

When the particle is of axially-symmetric shape, $\mathbb{D}_{\phi \phi} = D_r \mathbb{1}$ and the second-order operator is the spherical Laplacian. (In that case ignore the $\psi$ Euler angle.)

We can then solve the problem by expanding $V$ in terms of spherical harmonics, which are eigenfunctions of the Laplacian.

[See for example Cates & Tailleur (2013); Sandoval (2013).]

But in general $\mathbb{D}_{\phi \phi}$ can be essentially arbitrary.
Avoiding spherical harmonics

We can completely avoid spherical harmonics by using the magic relation

\[ \mathcal{L} \mathbf{Q} = -\mathbf{Z}^{-1} \cdot \mathbf{Q} \]

where we define the positive-definite matrix

\[ \mathbf{Z}^{-1} = (\text{Tr} \, \mathbb{D}_{\phi \phi}) \, 1 - \mathbb{D}_{\phi \phi} - \mathbf{\Omega} \cdot \mathbf{\epsilon} \]

\[ = k_B T \{ (\text{Tr} \, \mathbb{M}_{\phi \phi}) \, 1 - \mathbb{M}_{\phi \phi} \} - \mathbf{\Omega} \cdot \mathbf{\epsilon} \]

[Obvious? It wasn’t to us! Harder calculation than it looks.]

Solution to \( \mathcal{L} \chi = \mathbf{V} \) is thus \( \chi = -\mathbf{Z} \cdot \mathbf{Q} \).
The effective diffusivity

We omit a lot of details, but eventually we find our sought-after isotropic effective diffusivity:

\[ D_{\text{eff}} = \frac{1}{3} \text{Tr} \, D_{xx} + \frac{1}{3} U \cdot Z \cdot (U - 2\nabla \phi \cdot D_{\phi x}) \]

where recall that \( U \) is the total velocity of the particle, including noise-induced drift, and

\[ Z^{-1} = (\text{Tr} \, D_{\phi \phi}) \mathbb{1} - D_{\phi \phi} - \Omega \cdot \epsilon. \]

For a hydrodynamically isotropic particle (\( D_{\phi \phi} = D_r \mathbb{1} \)) with \( \Omega = 0 \), reduces to the ‘traditional’ result

\[ D_{\text{eff}} = \frac{1}{3} \text{Tr} \, D_{xx} + \frac{1}{6D_r} U \cdot (U - 2\epsilon : D_{\phi x}) \]

with potentially a correction \( \epsilon : D_{\phi x} \).
There is an insight here for the most classical of all particles, a thermal passive particle \((U_{\text{swim}} = \Omega_{\text{swim}} = 0)\), which still has a noise-induced drift:

\[
U = \nabla \phi \cdot \mathbb{D} \phi x = \epsilon : \mathbb{D} \phi x, \quad \Omega = \nabla \phi \cdot \mathbb{D} \phi \phi = 0.
\]

The term \(\epsilon : \mathbb{D} \phi x\) can only be nonzero if the centers of mass and reaction don’t coincide.

When subjected to noise, such a ‘wobbly passive particle’ behaves a bit like an ABP, with an effective noise-induced swimming velocity!

**Does this have consequences?**
Passive thermal particle: Effective diffusivity

Part of the original motivation for this work was a claim in a recent preprint that the noise-induced drift $\nabla \phi \cdot \mathbb{D}_{\phi x}$ for a wobbly particle leads to an ‘enhanced’ effective diffusivity

$$D_{\text{eff}} = \frac{1}{3} \text{Tr} \mathbb{D}_{xx} \times (1 + \frac{1}{2} \| \Delta x \|^2 / a^2) \quad \text{wobbliness correction}$$

where $\mathbb{D}_{xx} = k_B T \mathbb{M}_{xx}^h$ is the diffusivity tensor defined from the center of hydrodynamic reaction, i.e., as if the particle was not wobbly.

However, accounting for the coupling terms between the rotational and translational degrees of freedom reveals

$$D_{\text{eff}} = \frac{1}{3} \text{Tr} \mathbb{D}_{xx}^h$$

Conclude: even though the small-scale dynamics differ, the large-scale dynamics of a wobbly particles are the same as an unwobbly one.
Other results and future work

- Previous work called ‘chiral’ a particle with a net rotation $\Omega$, but the particle was axially symmetric.
- Here we can involve chirality through shape as well.
- Can derive a more complex, general formula for particles subjected to external field (Sevilla, 2016).
- Can also allow dependence large-scale variables that can lead to a novel large-scale drift.
- Our work actually focuses also on non-thermal active particles, involving a generalized fluctuation-dissipation theorem.
- We derive the overdamped limit from the Langevin equation, which is involves subtleties regarding the interpretation of multiplicative noise.


