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Hydrodynamic interactions in dense active suspensions: from polar order to dynamical clusters

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We study the role of hydrodynamic interactions in the collective behaviour of collections of microscopic active particles suspended in a fluid. We introduce a novel calculational framework that allows us to separate the different contributions to their collective dynamics from hydrodynamic interactions on different length scales. Hence we are able to systematically show that lubrication forces when the particles are very close to each other play as important a role as long-range hydrodynamic interactions in determining their many-body behaviour. We find that motility-induced phase separation is suppressed by near-field interactions, leading to open gel-like clusters rather than dense clusters. Interestingly, we find a globally polar ordered phase appears for neutral swimmers with no force dipole that is enhanced by near field lubrication forces in which the collision process rather than long-range interaction dominates the alignment mechanism.

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Active materials are condensed matter systems self-driven out of equilibrium by components that convert stored energy into movement. They have generated much interest recently, both as inspiration for new smart materials and as a framework to understand aspects of cell motility \cite{1,2,3}. They are characterised by interesting nonequilibrium collective phenomena, such as swirling, alignment, pattern formation, dynamic cluster formation and phase separation \cite{4,5,6,7,8}. Theoretical descriptions of active systems range from continuum models \cite{1,9} to discrete collections of self-propelled active particles \cite{4}. An influential classification of self-propelled active particle systems has been to group them into dry and wet systems \cite{1}. Dry systems do not have momentum conserving dynamics (e.g. Vicsek models \cite{4,9} and Active Brownian particle (ABP) models interacting via soft repulsive potentials \cite{10,11,12}), while wet systems conserve momentum via a coupling to a fluid (e.g. Squirmer driven by surface deformations \cite{13,14,15} and Janus particles driven by surface chemical reactions \cite{16}) leading to hydrodynamic interactions between active particles. Dealing with hydrodynamics leads to significant technical hurdles; as the motion of a self-propelled swimmer is affected by other particles due to both fluid flow and pressure, and even the two-body interaction between spherical squirmers in close proximity (near-field) is non-trivial, requiring sophisticated numerical analyses \cite{17,18,19,20,21,22,23}. Therefore, converting this into an understanding of collective behaviour remains a significant challenge \cite{24}. Because numerical simulations with hydrodynamics require significantly more computational power, studies of these systems have relatively few particles or low resolution of fluid flow \cite{25,26,27,28}. Hence, far-field approximations (swimmers as point multipoles) \cite{29} are often used to account for hydrodynamic interactions \cite{30}. This clearly breaks down when the swimmers are close to one another, limiting the validity of such studies to very dilute suspensions.

The appearance of dynamical clusters \cite{11,31,32} in recent experiments on active particles has generated much interest. This has been linked to a clustered state is observed in two-dimensional (2D) ABP systems called motility-induced phase separation (MIPS) \cite{10,11,12,32} and for squirmers confined between walls \cite{26,33}. A major difference however is finite size clusters in experiments \cite{7,11,31} compared to the infinite cluster formed in MIPS. In addition, recent simulations have shown that clusters are absent in 2D squirmer suspensions and in a 2D squirmer monolayer embedded in a 3D fluid \cite{27,34}. While attractive interactions can lead to clustering \cite{35,36,37}, here we study swimmers with purely repulsive interactions to see role of hydrodynamics in active cluster formation.

It is accepted from continuum arguments that the polar state is generically unstable for wet active systems \cite{1,2,3}, however recent simulations of wet active particles have raised the interesting possibility of other continuum limits in these systems. A polar state has been observed for neutral squirmers with no force dipole with 3D hydrodynamics, but 2D motion \cite{19} and in 3D \cite{38,39,40}. It has been suggested \cite{19} that near-field effects enhance the polar state although there are hints that far-field effects also play a role \cite{19}. These results are limited by relatively few particles so it is natural to ask if the polar state is present in the thermodynamic limit.

In this letter, we systematically construct equations of motion for wet active particles, namely, the dynamics of their position and orientation. One of our aims is to
provide a computationally tractable model of comparable complexity to ABP which takes account of hydrodynamic interactions of particles in close proximity [17, 41]. Using it, we study a suspension of force/torque free repulsive spherical squirmers and obtain the phase behaviour summarized in Fig. 2 as a function of density $\rho_0$. In studying the phase behaviour, we have emphasised the dependence on the sign of the force dipole ($v_2$) and contrasted them to neutral swimmers with force quadrupole and no force dipole ($v_2 = 0$). We find significant differences between the hydrodynamic interactions with and without near-field effects. The phase behaviour of neutral swimmers ($v_2 = 0$) with only far-field interactions are similar to those of ABPs since there are no collision-induced reorientations. Upon including near field effects, we obtain phase diagrams characterised by at low densities, a disordered ‘gas’ state and at higher densities, the emergence of stable ‘static clusters’ except for neutral swimmers [50] which spontaneously develop polar order. Dense static clusters, present in far-field only system, are suppressed by the near-field interactions, leading to open gel-like clusters. In between the gas and static cluster is phases of ‘dynamic clusters’ of finite size that exchange particles with bulk. While the boundaries between different clustered phases are qualitative and threshold-dependent, the boundaries between the polar state and other states has all the features associated with a phase transition.

Each particle (squirmer) is characterized by its position and orientation $(r^{(i)}, p^{(i)})$ with dynamics given by

$$\dot{r}^{(i)} = u^{(i)}; \quad \dot{p}^{(i)} = \omega^{(i)} \times p^{(i)}; \quad |p^{(i)}| = 1 , \quad (1)$$

The translational and angular velocities of each particle, $u^{(i)}$ and $\omega^{(i)}$ respectively, are obtained by solving for the fluid mediated interaction between all pairs of particles. The fluid is taken as incompressible with vanishing $\text{Re}$:

$$\eta \nabla^2 v - \nabla p = 0; \quad \nabla \cdot v = 0 \quad (2)$$

where $\eta$ is viscosity, $v(r)$ is the velocity, and $p(r)$ the pressure. The boundary condition on the swimmer surface is a sum of rigid translation, $u$ and rotational, $\omega$ motion and an active slip flow, $v_s$ driving self-propulsion:

$$v_s = \mathbf{u} + \omega \times \mathbf{R} + v_s \quad (3)$$

for a swimmer with centre at the origin. The fluid velocity vanishes at infinity, $v_s |_{r \to \infty} = 0$, with $\theta$ the polar angle with the $z$-axis and azimuthal, $\varphi$ with the $x$-axis on

\[
\begin{align*}
\mathbf{v}_s &= \sum_{l=1}^{447} \sum_{m=-l}^{l} [v_{lm} \Psi_{lm}(\theta, \varphi) + w_{lm} \Phi_{lm}(\theta, \varphi)],
\end{align*}
\]

FIG. 1: (Colour Online) Schematic interaction between swimmers. Each particle creates a leading order (A) quadrupolar or (B) dipolar flow. (C) When two particles are very close to each other, lubrication flow dominates the interactions.

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FIG. 2: (Colour Online) The state diagram of squirmers with density ($\rho_0$) and dipolar strength ($v_2$) with (A) far field hydrodynamic interactions and (B) both far field and near field with $N = 2048$ particles. Snap shots of (C) the static dense cluster state for a far-field-only system ($\phi = 0.447$, $v_2 = 0$), (D) the dynamic cluster state ($\phi = 0.251$, $v_2 = 0$), (E) the polar state ($\phi = 0.447$, $v_2 = 0$), and (F) the static open cluster state for a near+far system ($\phi = 0.447$, $v_2 = 0.9$).
the $xy$-plane. The slip velocity $v_s$ can be very efficiently expanded in the tangential vector spherical harmonics, $\Psi_{lm}$ and $\Phi_{lm}$ [42, 43]. The second term in (4) represents rotational slip around the swimmer axis associated with spinning motion which we neglect in the following and from now on set $u_{lm} = 0$. The swimmer axis $p$ is a unit vector (see Fig. 1). For uniaxial particles, $v_{lm}$ is a function of a magnitude $v_l$ and the swimmer orientation $p$ [43]. An isolated squirmer moves with the velocity $u^{(i)} = u_0 p^{(i)}$ with $u_0 = - \frac{2}{3} \sqrt{\frac{\pi}{R}} v_l$.

Given two squirmers, separated by $r_{ij}$, the flow field generated by one will affect the other and hence lead to modification of the self-propulsion velocities. We split the problem into two parts, a force and torque, acting on the sphere with: 1st, slip boundary conditions without translational and rotational motion ($F(p), T(p)$, the passive problem), and 2nd with the non-slip boundary conditions undergoing rigid-body motion and $\omega^{(i)} (F^{(a)}, T^{(a)}$, the active problem) [43]. The force and torque-free conditions imply, $F^{(a)} + F(p) = 0$ and $T^{(a)} + T(p) = 0$ which determine $u^{(i)}$ and $\omega^{(i)}$. The problems can be solved exactly for pairs of particles in two asymptotic limits: (1) when their separation, $r_{ij} = |r_{ij}|$ is much less than their radius (near-field) and (2) when their separation is much greater than their radius (far-field). For arbitrary separations between particles, we interpolate between the two limits; far-field and near field, using the tanh function. There is long history of calculation of the passive problem [44, 45]. Here we compute for the first time the active problem for both far-field and near-field in the general setting. Previous near-field active results have been obtained only for axisymmetric surface flowfields [17]. It should be noted that to obtain the velocity and angular velocity for collections of swimmers, one must solve the active/passive problems for all possible relative orientations which has not been achieved before [23].

For a pair of squirmers (labelled $i,j$) with arbitrary positions (and orientation), we define a spherical coordinate system: relative separations $r_{ij} = |r_{ij}|$, polar angles $\theta_{ij}$ and azimuthal angles $\varphi_{ij}$. Using it, a general form for the velocities valid in both far and near field limits is

$$u^{(i)} = u_0^{(i)} + \sum_{j \neq i,l,m} \left[ u_{lm,\parallel}^{(ji)} \Psi_{lm}^{(i)} + u_{lm,\perp}^{(ji)} \Phi_{lm}^{(i)} \right]$$

$$\omega^{(i)} = \sum_{j \neq i,l,m} \left[ \omega_{lm,\parallel}^{(ji)} \Phi_{lm}^{(i)} - \omega_{lm,\perp}^{(ji)} \Psi_{lm}^{(i)} \right].$$

where $\Psi_{lm}^{(i)} = \Psi_{lm}^{(\theta, \varphi)}$, $\Phi_{lm}^{(i)} = \Phi_{lm}^{(\theta, \varphi)}$, $Y_{lm}^{(i)} = Y_{lm}^{(\theta, \varphi)}$ with the $Y_{lm}$ the normal vector spherical harmonics. For the far-field, $u_{1,\parallel}^{(ji)} \sim r_{ij}^{-3}$ (quadrupole), and $u_{2,\parallel}^{(ji)} \sim r_{ij}^{-2}$, and $\omega_{2,\parallel}^{(ji)} \sim r_{ij}^{-3}$ (dipole). For the near-field, $u_{1,\parallel}^{(ji)} \sim \epsilon \log \epsilon$ and $u_{1,\perp}^{(ji)} \sim \epsilon \log \epsilon$ with $\epsilon = (r_{ij} - 2R)/r_{ij}$ [43]. $\lambda^{(i)} = 1$ when the $i$th particle is away from near-field region of any other particles and $\lambda^{(i)} = 0$ otherwise.

Equations (1), (6), and (5) form a closed complete dynamical system. Using them, we performed numerical simulations of $N$ identical particles of radius $R$ with periodic boundary conditions. Figure 2 shows various state points of this model as a function of the density, $\rho_0 = \pi R^2 N/L^2$ and the force dipole strength $v_2$. Most have $N = 2048$ unless specified otherwise. Defining, the average distance between two particles, $\xi = R/\sqrt{\pi/\rho_0}$, we vary $\xi$ from $\xi \simeq 2.65$ to $\xi \simeq 5.30$. We set $v_1 = 1$ for all swimmers and thus $u_0 \simeq 0.32$.

The size of a particle is chosen to be of unit length, thus we set $R = 1$. The time scale is normalised by the time for an isolated squirmer to move a half of its body length, that is, $\tau_0 = R/u_0$. There is a time scale associated with collisions, $\tau_m = \xi/u_0$. We vary the time scale from $\tau_m \simeq 8$ to $\tau_m \simeq 17$. We consider motion restricted to 2D but interacting via 3D hydrodynamic interactions. We neglect the modes with $l \geq 3$. We note that for 3D hydrodynamics projected onto 2D, pushers and pullers are not identical; the interaction at the front and the back is stronger than that at the side. As a result, pullers, on average attract nearby objects. We find global phase separation of active particles with repulsive interactions, i.e. MIP, is suppressed by near-field hydrodynamics and we find instead networks of open clusters for a large range of intermediate densities. We see a gel-like extended state at high enough densities.

Most surprisingly we find that for neutral (quadrupolar) squirmers and squirmers with small dipoles, $|v_2| < 1$, the swimmers self-organise into a polar state with aligned orientations and swimming directions. This polar order vanishes at low density. Screening far-field interactions [46] leads to polar order at lower densities (see Fig. 3(C)). As $|v_2|$ is increased, polar order vanishes. For example, for pushers ($v_2 > 0$), polar order disappears at $v_2^* \simeq 0.15$ ($\beta_{2,1} \simeq 0.58$) in Fig. 3(A). The loss of polar order is accompanied by divergence of fluctuations of the polarity as shown in Fig. 3(A). The position of the phase boundary is not symmetric about $v_2 = 0$, i.e. different $|v_2|$ for pushers and pullers (see Fig. 3(B)).

We check the stability of polar order to fluctuations by adding Gaussian white noise of amplitude $\sigma$ to the rotation in eqn. (1). At a fixed density, $\rho_0$ we find a transition from a gas to a polar state at a critical value of $\sigma_c > 0$. Polar order remains as we increase system size. In Fig. 3(D), polar order is shown as a function of the number of particles, $N$ up to $N = 8192$ for the near-far field system and $N = 32768$ for the near-field only system. Therefore we conclude that the system is asymptotically in a state with macroscopic global polar order. However we are not able to exclude the possibility that it disappears at much larger system sizes, even though we cannot identify an obvious physical mechanism by which that might happen. The mean cluster ratio, $\langle q \rangle$, the fraction of swimmers in large clusters, is nearly zero throughout the polar phase (see Fig. 3(D)), indicating clusters are not associated with polar order.

It is evident from the simulations that collisions be-
For pullers (Fig. 4(D)) and larger reflection angles for pushers (Fig. 4(C)) eventually destroy the polar state for both of them as \(|v_2|\) becomes large. The saturation angle in Fig. 4(C) is due to direct contacts between squirmers (repulsive forces from the interaction potential). This reorientation depends weakly on the contact interaction; big changes of interaction potential lead only to slight shifts of saturation angles. Hence the collective behaviour and phase boundaries are independent of the choice of potential [43].

While we only considered pairwise interactions, combining many of them results in many-body effects which become relevant for a non-dilute suspension. In fact, for a dense suspension, dynamics is dominated by the lubrication interaction between squirmers which is well approximated by a sum of two-body interactions. We note in particular, that \((p)\) is independent of \(N\) even for \(v_2 \neq 0\) (i.e. weak pushers) as shown in Fig. 3(D). To understand how these many-body effects give rise to collective behaviour, we have carried out numerical simulations of a minimal model, in which the only non-zero interactions are rotational near-field: \(\omega_{ij}^{(2)} \sim \epsilon_0\) [43] plus noise. It has two key ingredients: short-range orientational interactions and short-range repulsive interactions. We are able to reproduce the same polar-range orientational transition by increasing the noise amplitude. We conclude that the detailed form of the hydrodynamic interactions are not essential for the development of polar order. In con-
trast to the Vicsek model, here the lack of an alignment rule means excluded volume interactions are required to generate polar order. We observed density bands near the critical point of the order-disorder transition induced by noise for neutral swimmers and the minimal model mentioned above [49]. While this is reminiscent of variants of the Vicsek model, we note that here the polar order is destabilized not only by noise but also by the dipolar interactions.

The existence of polar order is fundamentally surprising because of the apparent contradiction with the well accepted generic instability of polar/nematic order of wet active matter [1, 47]. To understand this we construct a two-fluid model for the system which allows a macroscopic relative flux between swimmers and suspending fluid at small wave numbers, $k$. The suspending fluid (volume fraction, $1 - \phi$) with velocity $\mathbf{v}(\mathbf{r}, t)$ and the active particle (squirmers) 'phase' (volume fraction $\phi$) with local displacement variable $\mathbf{u}(\mathbf{r}, t)$ due to squirmers density variations. Finally we identify a local polar order parameter, $p(\mathbf{r}, t)$. Our analysis highlights collisions of the swimmers as essential for the formation of polar order. An isolated squirmers swims with velocity $u_0 p$ relative to the background fluid. The fluid obeys the Stokes equation with a force density $f^c \sim O(\rho^2)$ due to collisions between squirmers and an active stress $\sigma_{ij}^a = \zeta \rho_a p_{ij}$ where $\zeta \propto v_2$ and $\rho_a = \phi \rho$ is the average density of active particles. Replacing $f^c$ by $\chi (\mathbf{u} - \mathbf{v} - u_0 p)$, and linearising $p, v$ about the homogeneous polar state: $p = p_0$ and $v_0 = u_0 - u_0 p_0$,

$$0 = \eta \nabla^2 \delta \mathbf{v} + \chi \delta \mathbf{v} - \nabla P + \nabla \cdot \mathbf{u}^a : \nabla \cdot \delta \mathbf{v} = 0 \ , \ (7)$$

$$\partial_t \delta p = -u_0 p_0 \cdot \nabla \delta p + \delta \omega \cdot p_0 + \gamma \delta \mathbf{e} \cdot p_0 + K \nabla^2 \delta p \ , \ (8)$$

where $\omega_{ij} = \frac{1}{2}(\partial_i v_j - \partial_j v_i)$, $e_{ij} = \frac{1}{2}(\partial_i v_j + \partial_j v_i)$ and $K$ is the Frank elastic constant in the one constant approximation. A finite screening length $\xi \sim \sqrt{\eta / \chi}$ weakens the generic instability from long-wavelengths to finite wavelengths and stabilises the polar state on long length-scales. Hence a combination between the screening length $\xi$ and the active lengthscale, $\sqrt{K / \zeta} [48]$, allows us to determine the onset of polar order for $|\zeta| < \zeta_c = K / \xi^2$, i.e. for swimmers that are close to neutral. We recover the generic instability as $\phi \rightarrow 0$.

We emphasise that the computational expense of including hydrodynamics in the simulations of collective behaviour of active matter requires trade-offs where accuracy is sacrificed. Navier-Stokes (NS) solvers such as Lattice-Boltzmann compromise on the resolution of the velocity field and hence do not accurately describe fluid flow when the active particles are very close to each other. Here we have developed another scheme whose strengths are exactly where NS solvers are weak, for active particles in close proximity. It is also very accurate when the particles are well separated. Where it is less accurate, however is at intermediate separations. Its other great advantage is the ease with which we can study systems with many more particles. Another nice feature is the ability to switch off different contributions to the motion to identify the dominant mechanisms behind the macroscopic phenomena observed. Using it we have studied the collective behaviour of large numbers of spherical active particles and confirmed and clarified the phenomena observed in smaller simulations. Dense cluster formation is suppressed and we show that it is replaced by open gel-like aggregates at higher densities and most surprisingly, a polar ordered phase is stabilised by hydrodynamic lubrication interactions. We have also provided analytic continuum arguments explaining how such a state can be realised. In addition to the work presented here, we have studied purely 2D systems (2D with 2D interactions) and 3D systems (3D with 3D interactions), and obtained similar results, but at higher densities[49].

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[50] See [43] for the definitions of the polar order and the clusters