Motility-induced phases: out-of-equilibrium droplets, surfaces, and survival

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Abstract

Active matter represents a unique kind of out-of-equilibrium matter that is endowed with *motility*, the ability of each individual unit to move according to its own self-propulsion force. Objects of study in active matter include entities like birds and cells, which become particularly interesting at scales larger than the individuals, where we find emergent collective behavior like flocking and morphogenetic self-organization, respectively. One can study both microscopic and macroscopic behaviors of these particles using theory, simulation, and experiment, but largescale simulations are critically important to understanding some of the underlying statistical mechanical properties of active matter.

Motility-induced phase separation (MIPS) is a unique example of out-of-equilibrium emergent behavior, in which a fluid of active particles with repulsive-only interactions use their motility to spontaneously separate into coexisting dense and dilute phases. In this emergent collective behavior, particles nucleate stable clusters that eventually coarsen and coalesce into system-spanning bulk phases that stabilize in a steady state, much like nucleation and spinodal decomposition in liquid-gas phase coexistence. This dissertation covers the work I have done studying the fundamental physics of MIPS.

The spontaneous aggregation of MIPS results from the random and uncoordinated efforts of many particles that are driven by non-Markovian, randomized forces at the level of individual units. Building off of ideas of classical Brownian motion, in this thesis I first review some basics of Langevin dynamics and the Fluctuation-Dissipation Theorem (FDT) in order to describe how run-and-tumble particles (RTPs) and active Brownian particles (ABPs) break from equilibrium by utilizing their short-time ballistic motion, which becomes diffusive on long timescales. I review some continuum descriptions that provide an effective equilibrium picture of MIPS as well as experimentally engineered synthetic systems that exhibit life-like self-assembly and mesoscopic clustering.
My work studying MIPS has relied on simulations of large ensembles of Active Brownian Particles (ABPs). Using these, I can directly measuring quantities like pressure, surface tension, density, currents, and cluster growth exponents of systems of ABPs. All of these quantities can be compared to continuum models and experiments. As described by the main chapters of this thesis, my work has focused on studying the pressure and kinetics of MIPS, and more recently, its uniquely out-of-equilibrium surface “tension”. Additionally, I have worked in collaboration with biologists to study the self-assembly of a solid-dwelling bacteria, *Myxococcus xanthus*, whose cells utilize collective behavior to form aggregates known as fruiting bodies. Fruiting bodies are nascent structures that are critical to the survival of *M. xanthus* colonies, and while bacteria are inherently more complex than ABPs, we have shown that the onset of fruiting body formation is remarkably similar to MIPS at large scales. Overall, this work is part of a larger discussion about the unique out-of-equilibrium nature of MIPS, seeking to answer big questions about universality in living matter.
Motility-induced phases
out-of-equilibrium droplets, surfaces, and survival

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Right shows the normal current density $I^n_n$ measured in the local frame and averaged along the interface. Again, solid (open) symbols correspond to particles in the dense (gas) phase. The positive, outward-moving average current just inside the dense phase is balanced by the negative inward-moving current just inside the gas phase. Within each phase, the normal average current is finite within a surface layer of thickness $\xi^d$ (dense) and $\xi^g$ (gas) and decays exponentially. The solid lines are fits to the exponential decay. The top right inset shows $\xi_{d,g}$ extracted from those fits as functions of $\ell_p$. The solid lines are fits that show the linear growth of $\xi_{d,g}$ with persistence. The bottom left inset shows that the signed tangential current density $I^t_t$ is zero at the interface because particles travel without preference in either direction tangent to the interface. Here we see the fluctuations in the tangential current are much larger inside the cluster where tangential are most dominant (compare to Fig. 4.4).

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High-density clusters nucleate when colliding particles are caged by their surrounding neighbors before being able to reorient.

5.8 Static structure factor calculated for time intervals used to produce $L(t)$ ($\phi = 0.5, P_{e_{r}}^{-1} = 0.01$, $L = 10^3$). This is an example of spinodal phase decomposition, where this distribution is expected to tighten around smaller $k$ as dense regions coarsen with time.
5.9  (A) Examples of WT FB size evolution at different densities. The top two conditions stimulate nucleation and growth as fruiting bodies form at varying times. The bottom two conditions stimulate spinodal decomposition where the emergence of fruiting bodies is synchronized. (B) Histogram of the variance in the time when fruit bodies reach half-maximal size. When this time variance is smaller than 1h, we conclude that the system is undergoing spinodal decomposition. (C) The fraction of clusters observed over time is shown for 100 simulations at varying packing fractions (Pe−1 = 0.01). The resulting deviation of the nucleation times shows how as density is decreasing from just inside the coexistence region, significantly longer mean times and wider variances in times of nucleation occur. This provides a clear distinction between spinodal decomposition and nucleation and growth, allowing for a quantitative threshold between the two.

5.10 Local density distributions for three different mean densities, φ = {0.45, 0.55, 0.65}, at three different rotational Peclet numbers, Pe−1 = {0.005, 0.0025, 0.00125}. The peaks of these distributions are used to draw the boundary in the simulation phase diagram.

A.1 Normal (top) and tangential (middle) pressure broken down into swim, interaction and total parts. The bottom panel shows the various contributions (total, swim and interaction) to the difference \( p_n - p_t \) as a function of the distance \( x \) from the dense phase center of mass, for \( \ell_p = 140 \). This difference is the integrand in the expression for the tension, Eq. (4.11).
C.1 (top) The difference in the integrand in the mechanical definition surface tension for local slices beyond \((\delta h(s) > 0)\) or within \((\delta h(s) < 0)\) the average position of the interface, as a function of distance from the interface in that slice. The integrand is less negative in the valleys than the mountains, consistent with the results in Fig. 4.5. (bottom) The difference in transverse currents for the same sets of local slices. The excess transverse currents in the “mountains” suggests a Marangoni-like effect connecting local geometry and shear flow along the interface.
Chapter 1

Introduction

Collective behavior can be seen at all scales of life, in the form of patterns, order, and structures that serve important biological functions. Examples range from the flocking of birds [Ballerini et al. 2008] to the self-organization of cells in morphogenesis [Friedl and Gilmour 2009]. In recent decades, the new field of active matter has emerged from strong evidence that physical models can capture many aspects of this complex organization with minimal sets of physical rules or interactions [Toner and Tu 1998, Vicsek et al. 1999, Marchetti et al. 2013]. Active matter is a distinct category of nonequilibrium matter in which energy consumption, motility, and dissipation occur at the level of its microscopic constituents. The active matter paradigm bridges analytic, numerical, and experimental work in an effort to build a new understanding of the underpinnings of emergent collective behavior.

systems, the main subject of this dissertation, these efforts come together to connect the microscopic and macroscopic behavior of active matter, with the goal of providing a solid basis for phenomenological continuum models that try to formulate an effective equilibrium-like theory. A realization of active systems to which minimal models apply are active colloids, micron-size spheres partly coated with a catalyst that promotes the decomposition of one of the components of the ambient fluid, propelling the colloid forward [Paxton et al. 2006, Howse et al. 2007, Palacci et al. 2010, Poon 2013]. In the lab, these active synthetic particles can spontaneously assemble in coherent mesoscale structures with remarkable life-like properties [Palacci et al. 2013, Soto and Golestanian 2015], validating the theoretical biomimicry of emergent motility-induced phases. From the perspective of active matter, we hope to extrapolate from these simple systems to more universal physics of complex emergent patterns found in groups of living, motile entities.

Living matter has motility which is the ability to move itself. This is distinctly different from mobility, which is the well-established concept for quantifying the ability to be moved. Motility represents a new force, so it is reasonable to ask what new states emerge in motile matter? States of matter like solid, liquid, and gas are classically formed by a large number of interacting particles. The properties of atoms and molecules and the nature of their interactions determine the macroscopic behavior of the material. Living systems are generally composed of entities no smaller than cells, so ensembles of living matter may contain significantly smaller numbers of constituents. For instance, the average square inch of skin contains $N \sim 10^5$ cells while a single grain of salt contains $N \sim 10^{18}$ molecules. For comparison, a murmuration of starlings may contain hundreds to thousands of birds. One may then ask whether the tools of statistical mechanics can be used for living systems. Are there effective analogs to equilibrium material properties like temperature, pressure, and surface tension? What is the role of finite size in active systems? How do boundary conditions affect bulk behavior? By developing a robust framework for thinking about active systems, we hope to gain useful insight for future studies of order, structure, and pattern formation in living
Active systems exhibit large-scale spatial or temporal organization in states with unique macroscopic properties. For instance, a dense swarm of bacteria may flow as a living fluid with novel rheology \cite{Lopez15, Marchetti15}, self-organize into complex regular patterns \cite{Budrene95}, exhibit turbulent motion \cite{Dombrowski04}, or rigidify in a solid-like biofilm \cite{Hall-Stoodley04}. Inert matter is known to exhibit transitions between phases upon the external tuning of temperature or the introduction of boundary forces like shear stress or perhaps through globally applied forces via an electric or magnetic field. No longer just inert, active systems are tuned out of equilibrium by the self-propulsion of its constituents, generating new axes of unexplored phase space. The study of active matter is fundamentally directed toward describing and classifying the behaviors of this new class of nonequilibrium systems, and it does so by drawing on our understanding of familiar states of matter and the transitions between them, as controlled by its microscopic units. Our goal is to understand how new states of matter arise when we put together many units that are individually driven or motile. How much of this behavior is controlled solely by local interactions among the active particles? Is additional information transfer, like chemical signalling, required to understand the emergence of these new states? If we can classify and describe them can we then control the transitions between such states as we know how to do with familiar inert matter?

\subsection{Types of active matter}

One of the easiest ways to start classifying different types of active matter is to consider the local symmetries of its units, building on notions from equilibrium soft matter \cite{Chaikin00, Marchetti13}. The addition of self-propulsion provides an
additional tuning knob that generates emergent behavior. From this perspective, the three main categories are polar, nematic, and spherical active matter. In this section, I provide some brief background for each.

**Polar active matter**

Polar active particles are elongated motile particles with a head and a tail such as birds or bacteria. In the presence of polar aligning interactions, these can form polar or ferromagnetic states with a broken rotational symmetry where all particles are moving in the same direction on average. The orientational order that results is thus analogous to that of a ferromagnet, in which system spanning order means all particles point in the same direction on average.

This behavior is captured by the Vicsek model [Vicsek et al. 1995], in which point particles have a fixed speed and align their direction of motion with their neighbors according to a local noisy alignment rule. When the strength of alignment is large enough compared to noise they form a flock with a finite mean velocity.

The transition to the flocking state is first order [Grégoire and Chaté 2004] and the ordered state exhibits giant number fluctuations (GNF) across space [Ramaswamy et al. 2003, Narayan et al. 2007]. By GNF, we mean that the variance in the standard deviation...
\[ \Delta N = \langle (N - \langle N \rangle)^2 \rangle^{1/2} \] of the number of particles \( N \) grows linearly with the mean \( \Delta N \sim \langle N \rangle \).

This contrasts homogenously distributed systems, which obey the Central Limit Theorem \( \Delta N \sim \sqrt{N} \), a central tenet of equilibrium statistical mechanics. GNF are ubiquitous in active matter and have been observed in systems ranging from flocks\cite{Vicsek1995} to bacteria\cite{Lega2006} to vibrated granular rods\cite{Narayan2007}.

**Active nematics**

A second class of active systems is composed of particles that, although active, are head-tail symmetric and go nowhere on average, i.e., are apolar. An example is certain types of cells that move back and forth along their long axis, such as melanocytes\cite{Giomi2014}—the cells that spread pigment in our skin. These systems form nematic states that exhibit orientational order, but they show no preferred direction and no mean motion. Active nematics are essentially out-of-equilibrium nematic liquid crystals, which make them an exciting topic of study, with rich dynamical behavior\cite{Ramaswamy2003,Keber2014,Bertin2013}.

While there are many examples of active nematics, one that has drawn significant attention is a suspension of bundles of microtubules and molecular motors\cite{Keber2014}, which are of particular interest for the potential role they play in developmental and cellular processes. Special interest is given to the formation and dynamics of defects, which may help answer important questions about how structure is generated \textit{in vivo}.

**Spherical active matter**

Spherical active particles with no alignment interactions do not form states with any orientational order, but they still reveal a rich and surprising nonequilibrium dynamics. A realization are active colloids—colloidal particles propelled by self-catalytic reactions\cite{Theurkauff2012,Palacci2013}. A surprising finding is that purely repulsive active colloids can spontaneously form condensed liquid\cite{Fily2012a}, solid\cite{Bialke2013}.
and glassy states Berthier and Kurchan 2013, Berthier 2014 with properties akin to those induced in passive matter by attractive interactions.

At high enough densities, persistent motility, i.e., the ability to move ballistically in a straight line before a noisy precess randomizes one’s direction of motion, generates an effective attraction that allows the system to phase separate at intermediate densities. This effective attraction can be controlled by the persistence of self-propulsion. Beyond a critical threshold of persistence, systems of active colloids phase separate into dense and dilute phases, showing behavior akin to nucleation and spinodal decomposition observed at a conventional liquid-gas transition. This phenomenon is known more generally as motility-induced phase separation (MIPS) and is the primary topic of interest in this dissertation, so many more details of this will be fleshed out in the following sections and chapters.

1.2 Minimal models of active dynamics

Brownian motion is the random motion of a particle in fluid that arises at any finite temperature from the constant kicks from the solvent molecules. It is the simplest example of a nonequilibrium phenomenon that allows us to quantify the relation between the response of a system slightly displaced from equilibrium by an external force and the properties of fluctuations in the equilibrium state. This relation, known as the Fluctuation-Dissipation Theorem (FDT), is at the foundation of nonequilibrium physics. We will show here how active particle dynamics is markedly different since it does not obey a fluctuation-dissipation relation. This leads to some really surprising behavior. Active particles do not fill a container, but accumulate in corners and at walls Bechinger et al. 2016, Elgeti and Gompper 2013, Yang et al. 2014 with a strong dependence on curvature Fily et al. 2014a, Nikola et al. 2016, generate active Casimir forces Ray et al. 2014, and nucleate clusters in the absence of explicit interparticle attraction Tailleur and Cates 2008, Fily and Marchetti 2012a, Redner et al. 2013a, Bialké et al. 2015, Cates and Tailleur 2015.
Robert Brown, a botanist, first investigated what we now call Brownian motion in the form of jittering motion of small particles ejected from pollen grains [Brown 1828], which he found to move erratically and without physical explanation. Since these particles came from pollen, Brown’s interest in the problem came down to whether or not this motion was ironically a result of a “life force” [Brown 1828], which he ultimately ruled out. Einstein famously solved this problem in his seminal paper, *Theory of Brownian Movement* in 1905 [Einstein 1905]. The Einstein relation $D = \mu k_B T$ is the simplest example of a fluctuation-dissipation relation. A brief review of Brownian dynamics and the Einstein relation is presented in Sec. 2.1.

Self-propelled active particles do not, however, perform conventional Brownian motion. Now, we want to consider active particles, breaking from equilibrium. Several models have been developed to describe active dynamics. Two of the simplest and most widely used are the run-and-tumble particle (RTP) and the active Brownian particle (ABP), which are each schematically represented in Fig. 1.2. While it has been shown that their differences lead them to segregate when put in mazes [Khatami et al. 2016], when unconfined by boundaries, they function in much the same way [Solon et al. 2015d] in both dilute and finite density systems with open boundaries Here, I would like to briefly describe each, leaving mathematical details of ABPs, the primary particle of interest, for Chapter 2.

**Run-and-tumble particles**

This model is most appropriate to describe the dynamics of motile bacteria that continuously dissipate energy to propell themselves destroying microscopic time-reversibility. A canonical example is *Escherichia coli*, that performs [Cates et al. 2010, Cates and Tailleur 2013, Solon et al. 2015a] a sequence of runs and tumbles: a single cell “runs” or travels in a straight line at nearly constant speed $v_0$ for intervals punctuated by random reorientations or *tumbles*, which occur at a rate of $\alpha$ (Fig. 1.2). While these values vary a lot in populations of *E. coli*, $\alpha \approx 0.1 \text{s}^{-1}$ and $v_0$ ranges approximately from $0.1 \mu\text{m/s}$ to $50 \mu\text{m/s}$ [Matthus et al. 2011].
Figure 1.2: (Left) Run-and-tumble particles (RTPs) and active Brownian particles (ABPs) have characteristically different step styles. RTPs “run” in the same direction until they experience a “tumble” in which they reorient and move in a new direction until the next tumble, while ABPs continuously change their direction due to rotational diffusion $D_r$. (Right) Mean-square displacements for dilute simulations of ABPs, where the increasing value of $v_0$ indicates more persistent motion, breaking from the diffusive case where $v_0 = 0$. Here, we have set translational diffusion $D_t = 0.1 \mu m^2/s$ and rotational diffusion $D_r = 0.1s^{-1}$, varying persistence using self-propulsion speed $v_0 = 0, 1, 2, 4, 8 \mu m/s$. Note the long-time diffusive goes as $t$ while the shorter-time ballistic behavior grows as $t^2$. At even shorter times, the system is back to its diffusive regime at its shortest time-scales. Details about the use of these parameters in the ABP model can be found in Chapter 2.
While much slower, eukaryotic mammary epithelial cells have \( v_0 \approx 15 \mu m/hr \) and tumbling rates and a reorientation rates \( \alpha \approx 0.2 \text{min}^{-1} \) [Potdar et al. 2008]. At long times and at scales much larger than the average run length, these particles diffuse with \( D = v^2/\alpha d \), where \( d \) is the dimensionality. However, at short times on length scales lower than the average run length, these particles move ballistically, with a direction set by the previous tumble.

**Active Brownian particles**

As indicated by the name, the ABP model builds off the idea of Brownian motion, but integrates the persistent motion of RTPs. ABPs have a persistent motility of fixed speed and direction randomized by noise. Like RTPs, ABPs are ballistic at short times and diffusive on long timescales (Fig. 1.2). The equivalence of their fluctuating hydrodynamics has been demonstrated analytically and numerically. Rather than long straight runs and random tumbles, ABP directors get random kicks that result in small changes of direction at every time step. This results in time-dependent correlations of the propulsive force that are not matched by the constant particle mobility breaking the FDT. A more detailed description of ABPs motility is provided Chapter 2.

1.3 Motility-induced phase separation

In the relatively brief history of active matter, *flocking* is the model collective behavior of study, first formulated in a discrete-time agent-based point-particle model, known as the Vicsek Model [Vicsek et al. 1995] (introduced in Section 1.1.1). Two main drawbacks of the Vicsek model are its rule-based alignment mechanism and its lack of excluded volume, leading many to wonder about emergent effects of active particles that occupy space. Spherical ABPs with repulsive-only potentials represent perhaps the simplest model one can use to study the many-body physics of active matter. With this model, one can tune the density and persistence length of active particles so that they amazingly undergo complete spinodal
Figure 1.3: Schematic of nucleation in dense system of active Brownian particles (ABPs). High-density clusters nucleate when colliding particles are caged by their surrounding neighbors before being able to reorient.

phase decomposition in the absence of any attractive interactions in a process aptly called motility-induced phase separation (MIPS).

MIPS is essentially a traffic jam where due to their persistent dynamics, active particles effectively run into one another and get stuck—something like colloidal bumper cars whose dynamics can be tuned by the persistence of their directed motion. Those bumper-car-like traffic jams can become frequent enough in space and time, increasing the likelihood that more accumulate as part of a nucleation and growth process, as shown schematically in Fig. 1.3. At even higher persistence, systems separate into dense and dilute phases upon quenching, and this process grows to the size of the system in clear analogy with liquid-gas condensation. However, as droplets form, their interfaces are much more rough and only appear smooth and well-defined when averaged on long timescales. At short times, dense phases sometimes seem to just barely hold themselves together, fluctuating wildly, despite reproducing many attributes of classical phase separation: the normal pressure remains constant across interfaces, corresponding to a net zero interphase flux and bubbles form inside the dense phase where gaseous regions open up. MIPS is a genuinely fascinating

1Spherical cow jokes aside, this is perhaps the simplest way to imagine life-like individuals that remember their orientation longer than the traditional random or “drunken” walk. Thanks to Michael Cates for the bumper car analogy. He somehow managed to invoke live-action role playing as well.
phenomenon.

1.3.1 Theoretical descriptions and simulations

While MIPS has been observed in many simulations, it has proven difficult to demonstrate it quantitatively in experimental systems where aggregation in clusters of finite size (instead of bulk phase separation) is more often observed. Several striking similarities to equilibrium systems have been shown, like nucleation kinetics Stenhammar et al. [2013a], Redner et al. [2016], Patch et al. [2017a], spinodal phase decomposition Cates et al. [2010], Fily and Marchetti [2012a], Bialké et al. [2013], Wittkowski et al. [2014], Cates and Tailleur [2015], Marchetti et al. [2016a], Ostwald ripening Stenhammar et al. [2013a], Gonnella et al. [2015], bubble formation Tjhung et al. [2018], and a fascinating new out-of-equilibrium mechanism that operates like surface tension Bialké et al. [2015], Lee [2017], Patch et al. [2018]. While some of these features can be captured using equilibrium-like effective theories, many elements are still not understood well with current analytic predictions, like the anomalously negative surface tension Tjhung et al. [2018], Patch et al. [2018] found at the interface of MIPS phases. For this reason, the study of MIPS relies heavily on the simulation of microscopic equations of motion, which is relatively simple to implement using molecular dynamics Frenkel and Smit [2002], Fily and Marchetti [2012a] or Monte Carlo Levis and Berthier [2014].

Using simulations, one can directly measure physical quantities of interest that are otherwise both difficult to incorporate in phenomenological models and to tease of statistically limited experimental data. A primary goal of my work is thus to test the validity of these equilibrium-like descriptions and to help build a fuller picture of previously unexplored aspects of collective behavior of active systems. In particular my work studying interfacial tension in MIPS has led to interesting conversations about current limitations of our best continuum descriptions (Chapter 4). In my collaborative work studying the onset of fruiting body formation in M. xanthus, while we could show the quantitative similarity of single-
particle dynamics between reversing and non-reversing ABPs, without simulations we would have been unable to clearly show the resulting phase behavior at the larger scale (Chapter 5).

### 1.3.2 Synthetic active matter

Several synthetic systems have been engineered to show the emergent behavior of active systems. These include autophoretic colloids [Howse et al. 2007, Palacci et al. 2010, Theurkauff et al. 2012], rollers [Bricard et al. 2013], and droplets [Thutupalli et al. 2011]. The simplest realization of spherical microswimmers are Janus colloids, which are first created by coating half of a gold bead with platinum, and then activated by immersion in a solution of hydrogen peroxide ($H_2O_2$) [Paxton et al. 2006]. The difference in the consumption rate of $H_2O_2$ at the gold and platinum sides generates an asymmetric concentration of solute on the two hemispheres, which results in propulsion along the concentration gradient.

For these and other colloidal swimmers, interactions and propulsion can be tuned in a controlled way, allowing systematic studies up to moderate densities. One remarkable phenomenon shown by these systems is spontaneous assembly in mesoscopic clusters [Theurkauff et al. 2012, Palacci et al. 2013]. This phenomenon is distinct from equilibrium assembly, which arises from attractive interactions between the particles. Rather, this phenomenon has been shown to be driven by the nonequilibrium interplay of motility and crowding. The nonequilibrium pressure equation of state of active colloids has also been probed experimentally via sedimentation measurements [Ginot et al. 2015], showing a motility-induced effective adhesion that strongly suppresses pressure at moderate density. Many more experimental findings can be found in Ref. [Cates and Tailleur 2015], and theoretical work with ABPs has in many cases predicted and qualitatively reproduced aspects of these findings.

### 1.3.3 *M. xanthus* aggregation

Single-cell organisms such as bacteria and amoeba are capable of spontaneously organizing into complex multicellular structures [Laub and Loomis 1998, Dormann et al. 2002], mak-
Nucleation and growth: we compare *M. xanthus* at a concentration of $3 \times 10^9$ cell/ml (A) and simulations of ABPs with inverse Péclet number $\text{Pe}^{-1} = 0.01$ and packing fraction $\phi = 0.29$ (C). In both cases, small clusters form at random times and positions, only some of which develop into large nucleation centers. Spinodal decomposition: for larger densities, particle aggregates form everywhere at the same time and coarsen. This is shown for *M. xanthus* at a concentration of $1 \times 10^{10}$ cell/ml (B) and for ABPs with $\text{Pe}^{-1} = 0.01$ and $\phi = 0.5$ (D). Simulation snapshots have a linear size $L = 1000a$. 

Figure 1.4: Aggregation in *M. xanthus* (A-B) and in simulations of ABPs (C-D). The snapshots for both systems are taken at equal times in terms of the single-particle reorientation time ($\tau_r \approx 10$ min for the bacteria). If the density is sufficiently low, no aggregation occurs.
ing them generally interesting in the study of emergent phenomena. A striking example of this kind of collective behavior is the starvation-induced organization of the rod-shaped, soil-dwelling bacterium *Myxococcus xanthus* whose lifecycle includes the formation of macroscopic, multicellular droplets known as “fruiting bodies” (FBs) Zusman et al. [2007]. *M. xanthus* cells move by gliding on solid surfaces using both tank-tread-like transport motors and the retraction of extruded filaments called pili and can modulate their speed in a continuous manner Balagam et al. [2014], Hodgkin and Kaiser [1979]. The cells also have the ability to reverse their direction of motion typically every several minutes, and can modify the reversal frequency in different situations Wu et al. [2009], Blackhart and Zusman [1985], Thutupalli et al. [2015]. When nutrients are scarce, starving *M. xanthus* cells undergo a multicellular process of self-organization to form dome-shaped droplets, comprising hundreds of thousands of those cells. A subset of cells at the center of each droplet differentiate to form metabolically quiescent spores that can survive long periods of starvation Zusman et al. [2007], Starruß et al. [2012], Shimkets [1990], meaning individual cells have evolved to utilize this collective survival mechanism.

The striking phenotypic similarity between FB formation in *M. xanthus* and in the amoeba *Dictyostelium discoideum* has led to the longstanding hypothesis that *Myxococcus* FB formation is driven by long-range chemical signaling mechanisms, as it is in the amoeba. Despite decades of research, questions remain on the the links between spatial chemical cues and the direct mechanisms of FB formation. Although *M. xanthus* cells are known to employ chemical communication to initiate FB formation (termed A-signaling) Kuspa et al. [1992], to potentially synchronize reversal frequency (termed C-signaling) Lobjedanz and Søgaard-Andersen [2003], Shimkets and Rafiee [1990], and to communicate through the production of mucopolysaccharide “slime trails” that other cells can sense and follow Burchard [1982], a quantitative understanding of the mechanisms that drive aggregation has remained elusive.

Using experiments and insight from theory, I have worked in collaboration with biologists in the Welch Lab at Syracuse University and biophysicists in the Shaevitz Lab at Princeton
University to demonstrate that the onset of *M. xanthus* FB formation can be described as a phase separation process driven, at least initially, by changes to the motility of individual cells. Importantly, this appears to happen in the absence of complex signaling mechanisms and interactions between cells, and requires no real-time control at the cellular level. While the ability to actively change motility ultimately leads to a phase transition, cells do not have to implement a complicated feedback mechanism to alter motility in response to specific chemical or mechanical cues. Rather, cells need only speed up and suppress reversals upon starvation and the collective mechanics then naturally induces phase separation of the entire population.

This particular system provides an example of MIPS, where purely repulsive Active Brownian Particles (ABPs) spontaneously aggregate due to density-dependent inhibition of motility [Fily and Marchetti 2012a, Cates and Tailleur 2015, Marchetti et al. 2016c]. While there are important differences between the MIPS process of bulk phase separation and the droplet formation seen in *M. xanthus* populations, important findings of this work suggest drawing a connection between their aggregation dynamics to those understood in reversing ABPs provides a powerful way to understand the multiscale nature of *M. xanthus* aggregation.

### 1.4 Overview

The remainder of this dissertation is divided into four main chapters followed by general conclusions. Chapter 2 gives an introduction to the minimal models of ABPs we use to study MIPS and active systems more generally. It also introduces phenomenological continuum models that have been used to describe MIPS physics.

Chapter 3 uses simulations to examine the notion of pressure in fluids of ABPs. It examines finite-size effects in the kinetics of phase separation for both closed systems and periodic boundary conditions. We show that in ABPs pressure is generically a function of
density and that this is not the result of finite-size effects. We correlate the time evolution of
the mean pressure towards its steady state value with the kinetics of motility-induced phase
separation. For parameter values corresponding to phase separated steady states, we identify
two dynamical regimes. The pressure grows monotonically in time during the initial regime
of rapid cluster formation, overshooting its steady state value and then quickly relaxing
to it, and remains constant during the subsequent slower period of cluster coalescence and
coarsening. The overshoot is a distinctive feature of active systems. This work has been
published in Physical Review E: Kinetics of motility-induced phase separation and swim
pressure, Adam Patch, David Yllanes, and M.Cristina Marchetti, Phys. Rev. E, 95, 012601
(2017), Ref. Patch et al. [2017a].

Chapter 4 reports on more recent work aimed at quantifying the emergent properties
of the interface of coexisting motility-induced phases. Remarkably, the mechanical tension
measured along the interface between these phases is negative. In equilibrium this would im-
ply an unstable interface that wants to expand, but these out-of-equilibrium systems display
long-time stability and have intrinsically stiff boundaries. By studying this phenomenon in
detail using active Brownian particle simulations and a novel frame of reference. We have
shown that an out-of-equilibrium, Marangoni-like effect generates stability at the interface.
By shifting from the global (or laboratory) frame to a local frame that follows the dynamics
of the phase boundary, we observe correlations between the local curvature of the interface
and the measured value of the tension. Importantly, our analysis reveals that curvature
drives sustained local tangential motion of particles within a surface layer in both the gas
and the dense regions. The combined tangential current in the gas and local ”self-shearing”
of the surface of the dense phase suggest a stiffening interface that redirects particles along
itself to heal local fluctuations. These currents restore the otherwise wildly fluctuating in-
terface through an out-of-equilibrium Marangoni effect. We discuss the implications of our
observations on phenomenological models of interfacial dynamics. This work has been ac-
cepted for publication with minor revisions by Soft Matter: Curvature-dependent tension and

Chapter 5 discusses collaborative work with the experimental groups of Joshua Schaevitz (Princeton University) and Roy D. Welch (Syracuse University) aimed at understanding self-organization in *M. xanthus*, which we have modeled as a modified ABP whose single-cell parameters can be measured experimentally for direct comparison. By combining high-resolution single cell tracking experiments with numerical simulations, we show that starvation-induced fruiting body (FB) formation in *Myxococcus xanthus* is a phase separation driven by the cells by tuning their motility over time. The phase separation can be understood in terms of cell density and a dimensionless Péclet number that captures the cell motility in terms of its speed and reversal frequency. Our work suggests that *M. xanthus* evolved to take advantage of a self-driven non-equilibrium phase transition that can be controlled at the single cell level. This work is currently under review at Physical Review Letters: *A self-induced phase transition drives Myxococcus xanthus fruiting body formation*, authored by Guannan Liu, Adam Patch, Fatmagül Bahar, David Yllanes, Roy D. Welch, Shashi Thutupalli, M. Cristina Marchetti, and Joshua W. Schaevitz, arXiv:1709.06012 (2017), Ref. Liu et al. [2017].

I finally conclude with Chapter 6 giving an overview of this work.
Chapter 2

Minimal models of active particles

This section introduces the model of Active Brownian Particles (ABPs) that is studied in all the work presented in this dissertation. After reviewing the behavior of Brownian particles in a thermal bath [Zwanzig 2001], I will describe the Langevin dynamics of ABPs that is used in the numerical work, consider the role of fluctuation and dissipation in this simple system, and show how this minimal model exhibits collective behavior.

2.1 Brownian Particles

As introduced in Sec. 1.2, Brownian motion is perhaps the simplest nonequilibrium phenomenon that obeys a Brownian motion that obeys a fluctuation-dissipation relation. For a Brownian particle, there is a direct relationship between thermal fluctuations in equilibrium and response to a small perturbation from equilibrium (e.g. \( \mu \)). It is useful to first review Brownian motion to understand how we characterize the behavior of ABPs.

The key insight of Brownian motion is that one can split the effect of the random bombardment on the Brownian particle by the atoms in the surrounding fluid into a mean drag resulting in friction \( \zeta \) and a random force \( F(t) \) that has zero mean and is uncorrelated in time. The dynamics of a Brownian particle of mass \( m \) in two dimensions is then governed
by a Langevin equation

\[ m\dot{v} = -\zeta v + F_{\text{ext}} + F(t) \]  

(2.1)

with \( v = \dot{r} \) and \( F_{\text{ext}} \) an external force. The random force \( F \) has zero mean and uncorrelated variance

\[ \langle F(t) \rangle = 0 \]  

(2.2)

\[ \langle F_\alpha(t) F_\beta(t') \rangle = 2B\delta_\alpha\beta \delta(t - t') , \]  

(2.3)

where \( \alpha, \beta \) denote Cartesian components. The variance \( B \) of the noise is determined by requiring that, at long times and when \( F_{\text{ext}} = 0 \), the particle is in equilibrium and velocity fluctuations are described by a Maxwell-Boltzmann distribution, i.e., \( \lim_{t \to \infty} \langle [v(t)]^2 \rangle = \sqrt{k_B T / m} \). This gives \( B = 2\zeta k_B T \).

For simplicity, let us consider the case of large friction when inertia can be neglected for \( t \gg m/\zeta \) and the equation of motion is

\[ \zeta \dot{r} = F_{\text{ext}} + F(t) \]  

(2.4)

A constant \( F_{\text{ext}} \) yields a mean velocity \( \langle v \rangle = \mu F_{\text{ext}} \), with \( \mu = 1/\zeta \) the mobility.

In equilibrium (\( F_{\text{ext}} = 0 \)) the particle goes nowhere on average (\( \langle x(t) \rangle = 0 \)), but fluctuations can be quantified by the mean-square displacement (MSD) \( \langle [\Delta r(t)]^2 \rangle \) with \( \Delta r(t) = r(t) - r(0) \), which is found to be

\[ \langle [\Delta r(t)]^2 \rangle = 2Dt \]  

with \( D = \frac{k_B T}{\zeta} = k_B T \mu . \)  

(2.5)

This \( \sim t \) linear growth corresponds to diffusion. It should be contrasted to ballistic motion of a particle of constant velocity \( v \) where \( \langle [\Delta r]^2 \rangle = v^2 t^2 \).

The Einstein relation \( D = \mu k_B T \) represents the simplest example of the Fluctuation-Dissipation Theorem (FDT). The FDT is a general statistical mechanical result that one
can use to describe the direct relationship between fluctuations in the equilibrium state ($\sim k_B T$) and linear response to an external force (e.g. $\mu$) in systems at thermal equilibrium. Thus, Brownian motion represents an important foundation for relating diffusion to thermal equilibrium fluctuations.

### 2.2 Active Brownian Particles

#### 2.2.1 Single ABP

To model self-propulsion, we endow a Brownian particle of diameter $2a$ with an internally generated propulsive force $F_s = F_0 \hat{n}$ of fixed magnitude $F_0$ and direction identified by a unit vector or director $\hat{n} = (\cos \theta, \sin \theta)$, where $\theta$ is the angle of the director with the $x$-axis (see Fig. 2.1). Considering again overdamped dynamics, the equation of motion for a single ABP is given by,

$$\zeta \dot{v} = F_0 \hat{n} + \delta F(t) .$$ (2.6)

The direction of $\hat{n}$ is randomized by rotational noise

$$\dot{\theta} = \eta(t) .$$ (2.7)
Active Brownian particles (ABPs) display ballistic behavior $MSD \sim t^2$ at times $t < D_r^{-1}$ short compared to the reorientation time $\tau_r$ and diffusive behavior $MSD \sim t$ for $t > D_r^{-1}$. Here $\ell_p$ is varied and MSD results collapse when made dimensionless with proper length ($\ell_p$) and time ($D_r^{-1}$) scales.

where $\eta(t)$ is Gaussian white noise with zero mean and correlations $\langle \eta(t)\eta(t') \rangle = 2D_r \delta(t-t')$, with $D_r$ the rotational diffusion rate. It is convenient to rewrite the equation of motion in terms of velocity as

$$\dot{\mathbf{r}} = v_0 \hat{\mathbf{n}} + \eta^T(t) , \quad (2.8)$$

we have defined the self-propulsion speed $v_0 = \mu F_0$ and $\eta^T(t) = \mu \delta \mathbf{F}(t)$ with $\langle \eta^T_\alpha(t)\eta^T_\beta(t') \rangle = 2\mu k_B T \delta_{\alpha\beta} \delta(t-t')$, where mobility $\mu$ is the inverse of friction $\mu = \zeta^{-1}$.

The mean-square displacement (MSD) of a single ABP can be calculated analytically with the result

$$\langle |\Delta \mathbf{r}(t)|^2 \rangle = 4D_t t + \frac{2v_0^2}{D_r} \left[ t - \frac{1}{D_r} \left( 1 - e^{-D_r t} \right) \right]. \quad (2.9)$$

Here, we can see that at short times, $t \ll D_r^{-1}$ the MSD grows ballistically as $\langle |\Delta \mathbf{r}(t)|^2 \rangle \sim v_0^2 t^2$, and at long times, $t \gg D_r^{-1}$, it grows diffusively as $\langle |\Delta \mathbf{r}(t)|^2 \rangle \sim 4D_t + \frac{v_0^2}{2D_r} t$. The
quantity

\[ D_{\text{eff}} = D_t + \frac{v_0^2}{2D_t} \]  

(2.10)

defines an effective diffusivity. The active contribution \( D_a = v_0^2/(2D_t) \) to the diffusivity also has an intuitive interpretation in terms of the persistence length \( \ell_p = v_0/D_t \) and persistence time \( \tau_r = 1/D_t \) of the single particle dynamics as it can be written as \( D_a = \ell_p^2/(2\tau_r) \). ABPs travel ballistically over the length \( \ell_p \), with direction randomized over times \( \tau_r \), resulting in diffusive behavior at long times. This long-time diffusivity means these particles are equilibrium-like on long timescales. The short-time ballistic nature of these particles results in all kinds of new physics in the presence of interactions. Results from molecular dynamics simulations are precisely comparable to this prediction, as shown in Fig. 2.2.

It is interesting to note that the equations for translational and angular dynamics of each particle, can be rewritten as one equation with colored or non-Markovian noise of the form

\[ \dot{\mathbf{r}}_i = \mu \left[ \sum_{j \neq i} \mathbf{F}_{ij} + \mathbf{\xi}_i(t) \right], \]  

(2.11)

where the noise \( \mathbf{\xi}_i(t) \) has zero mean and variance

\[ \langle \xi_{i\alpha}(t)\xi_{i\beta}(t') \rangle = 2 \left[ D_t \delta(t-t') + \frac{v_0^2}{2} e^{-D_t|t-t'|} \right] \delta_{\alpha\beta} \delta_{ij}. \]  

(2.12)

This expression shows the key role of the persistence time \( \tau_r = 1/D_t \). ABPs do not obey the FD theorem because noise correlations are non-Markovian while the friction is constant.

It also allows us to define the notion of effective temperature. Using \( \lim_{D_t \to \infty} D_t e^{-|t|/D_t} = 2\delta(t) \), we can see that the contribution to Eq.(2.11) from the propulsive forces becomes white noise when \( D_t \to \infty \) for fixed \( \frac{v_0^2}{2D_t} \) as \( \frac{v_0^2}{4D_t} D_t e^{-|t|/D_t} \to \frac{v_0^2}{2D_t} \delta(t) \). This suggests the definition of an effective temperature \( T_{\text{eff}} = \frac{v_0^2}{2\mu D_t} \). It is, however, important to keep in mind that even in this limit the speed distribution of ABPs is not Maxwellian, as in equilibrium, since the particles have fixed self-propulsion speed.
2.2.2 Interacting ABPs

We now consider a collection of $N$ ABPs in an area $A = L^2$. Assuming the particle interacts via a pairwise additive force $F_{ij}$, the equations of motion are given by [Fily and Marchetti 2012a, Yang et al. 2014, Fily et al. 2014d]

\[
\dot{r}_i = v_0 \hat{n}_i + \mu \sum_{j \neq i} F_{ij} + \eta^T_i(t) ,
\]

(2.13)

\[
\dot{\theta}_i = \eta_i(t) ,
\]

(2.14)

$r_i$ is position of the $i$th particle, $\hat{n}_i = (\cos \theta_i, \sin \theta_i)$ its director. Both $\eta^T_i(t)$ and $\eta_i(t)$ are Gaussian white noise with zero mean and correlations are $\langle \eta^T_i(\tau) \eta^T_j(\tau') \rangle = 2D_t \delta_{ij} \delta_{\alpha \beta} \delta(t - t')$ and $\langle \eta_i(t) \eta_j(t') \rangle = 2D_r \delta_{ij} \delta(t - t')$ respectively, where $D_t$ is translational diffusion and $D_r$ is rotational diffusion, which may be related via effective temperature meaning $D_t = v_0^2 / (2D_r)$. Alternatively, one may model an athermal system by setting $D_t = 0$ and taking $D_r$ as a control parameter, as we have done throughout my work.

In the minimal ABP model, the interactions are purely repulsive. It has been shown that the large scale behavior does not depend on the specific form of the repulsive interaction. In our work we have used two types of interactions. The first is a simple harmonic force,

\[
F_{ij} = \begin{cases} 
  k(2a - r_{ij})\hat{r}_{ij} & \text{if } r_{ij} < 2a \\
  0 & \text{else} 
\end{cases}
\]

(2.15)

where $r_{ij} = |r_i - r_j|$ is the distance between particles $i$ and $j$ and $a$ is the particle’s radius. Throughout this work, we use monodisperse disks. Alternatively, one can use a potential with a harder core, like the Weeks-Chandler-Anderson (WCA) potential, a modified Lennard-
Jones potential, cut off at its energy minimum,

$$\Phi_{ij} = \begin{cases} 
\epsilon\left(\frac{a}{r_{ij}}\right)^{12} - 2\left(\frac{a}{r_{ij}}\right)^6 & \text{if } r_{ij} < r_m \\
0 & \text{else} \end{cases}$$

(2.16)

with $F_{ij} = -\nabla \Phi_{ij}$. An even simpler and more general solution to this is to simply take $F_{ij} \sim r_{ij}^{-\alpha}$, which allows tuning of softness by $\alpha$. In my work, I have used both harmonic and WCA potentials in order to create strong steric repulsion and minimal overlaps. In the case of harmonic potentials, this means setting spring forces that are very strong with respect to self-propulsion.

### 2.2.3 Summary of model parameters

It is useful to summarize the parameters of the model. The first is the packing fraction\footnote{Here packing fraction used does not account for overlaps which may result from softness of a chosen potential.}$\phi = n \pi a^2$, where $n = N/L^2$ is the number density. In 2D, $\phi_{2\text{DCP}} = \pi \sqrt{3}/6 = 0.9069$ for hexagonal close packing of disks and $\phi_{\text{RCP}} \approx 0.64$ for random close packings of monodisperse disks\cite{Luding2016}.

The rotational Peclet number, on the other hand, is a dimensionless number that is useful for comparing the energy of the system to its disordering. It is also nicely proportional to the persistence length and is given by

$$\text{Pe}_r = \frac{v_0}{a D_r} = \frac{\ell_p}{a}.$$ 

(2.17)

A more general summary of important length and time scales are shown in Table 2.1.
Length scales | Time scales
---|---
particle radius $a$ | persistence time $\tau_r = 1/D_r$
interaction length $\ell_p$ | interaction time (harmonic) $\tau_R = 1/(\mu k)$
system size $L$ | mean free time between collisions $\tau_c = 1/(2av_0n)$

Table 2.1: Summary of length and time scales.

### 2.3 Motility-induced phase separation

Purely repulsive active colloids form gas, liquid, and solid phases, with transitions between such phases observed upon tuning their density and motility. As discussed in Sec. 1.3, the most striking phenomenon exhibited by a minimal model of spherical active colloids is motility-induced phase separation (MIPS), in which an active fluid of particles with purely repulsive interactions spontaneously phase separates into dense and dilute phases [Fily and Marchetti 2012b] at densities well below close-packing. This effect follows from the slowing down of the active particles’ speed due to crowding. This yields an effective propulsive speed $v(\rho) < v_0$ resulting in the particles accumulating in the regions where they slow down. In other words, steric repulsions and crowding slow down the particles, increasing their density. This leads to further slow down, resulting in an effective traffic jam that manifests itself as complete phase separation, as shown in Fig. 1.3. MIPS occurs provided the persistence time is the longest time scale in the problem. Specifically, it requires $\tau_r$ to exceed the mean-free time between collisions, $\tau_c = 1/2av_0n$. The condition $\tau_r > \tau_c$ or $Pe_{r}na^2 > 1/2$ gives a simple criteria for the onset of MIPS [Matas-Navarro et al. 2014]. The MIPS regime therefore exists in an intermediate range of packing fraction $\phi$ and large persistence $Pe_{r} \gg 1$ (where $\ell_p \gg a$). In the case of soft repulsion, at low-intermediate densities a high-density fluid cluster emerges surrounded by a gas, and this changes continuously when moving to the region of higher densities where bubbles of gas phase emerge inside a densely packed liquid [Fily et al. 2014d].

A mean-field theory of MIPS can be developed as follows. From Eq. (2.13) it is clear that repulsive interactions suppress the self-propulsion velocity with $v_0 \rightarrow v(\rho) = v_0 + \mu \langle \hat{n}_i \cdot \rangle$.
Figure 2.3: A comparison of nucleated dense phase droplets in simulations of different system sizes, where interaction radius $a = 1$ is the unit length. Note that in the smallest system, $L = 100$, the dense droplet is less stable than those found in the larger simulations, which qualitatively corresponds with our quantitative measurements of finite size effects (Ch. 3).

$$\sum_{j \neq i} F_{ij} \approx v_0(1 - \rho/\rho^*)$$ for $\rho < \rho^*$ to leading order in $\rho$, where $\rho^*$ generally depends on $\text{Pe}_r$. One can then write effective dynamical equations of the form given in Eqs. (2.13) and (2.14) with $v_0 \to v(\rho)$. We can then construct continuum equations for the active fluid using found tools of statistical mechanics. We obtain equations for the density $\rho(r, t)$ and the polarization density $p(r, t)$ which describe the local orientation of the particles’ axis of self-propulsion, given by Fily and Marchetti [2012b]

$$\partial_t \rho = -\nabla \cdot [v(\rho)p - D_t \nabla \rho] \quad ,$$

$$\partial_t p = -D_t p - \frac{1}{2} \nabla [\rho v(\rho)] + K \nabla^2 p \quad ,$$

(2.18)  
(2.19)
Figure 2.4: Snapshot of an ABP simulation with inset zooming in on the shaded region of the background image. The inset shows particles in the dense liquid phase tagged with green (dark green for inward-pointing boundary particles, light green for particles in the bulk of the dense liquid), while gas particles are tagged with white. The self-propulsion vector is shown originating from the center of each disk. The full system shown in the background has periodic boundaries, $\phi = 0.5$ and $L = 200$. The parameters are $\mu = k = 1$, so $\tau_k = (\mu k)^{-1}$ is the time unit, $v_0 = 0.01$ and $a = 1$. The rotational diffusion has been set to give a Péclet number of $\mathrm{Pe}_r = 50$.

For times much longer than the reorientation time $t \gg \tau_r$, one can neglect the time derivative of the polarization in Eq. (2.19) relative to the damping term and eliminate $\mathbf{p}$ to obtain a nonlinear diffusion equation for the density, given by

$$
\partial_t \rho = \nabla \cdot \left[ D(\rho) \nabla \rho \right],
$$

(2.20)
with effective diffusivity

\[ D(\rho) = D_t + \frac{v^2(\rho)}{2D_r} \left( 1 + \frac{d\ln v}{d\ln \rho} \right). \]  

(2.21)

The linear stability of a homogeneous state of constant density \( \overline{\rho} \) can be examined by considering the dynamics of density fluctuations, \( \delta \rho = \rho - \overline{\rho} \), which is governed by

\[ \partial_t \delta \rho = D(\overline{\rho}) \nabla^2 \delta \rho. \]

The decay of fluctuations is controlled by a diffusive mode with diffusivity \( D(\overline{\rho}) \) that becomes negative when the \( D(\overline{\rho}) < 0 \), signaling the instability of the uniform state.
The onset of this instability corresponds to $D(\rho) = 0$. Using $v(\rho) = v_0(1 - \rho/\rho_*)$, we find $D(\rho) = D_t + \frac{v_0^2}{2D_t}(1 - 2\frac{\rho}{\rho_*})$ and $D(\rho) < 0$ for $\rho > \rho_c$, with $\rho_c = \rho_*/2 + \rho_* D_t D_r/v_0$. To express $\rho_c$ in terms of $\text{Pe}_r$ we need to estimate the density of $\rho_*$ that controls the suppression of motility. We do so via a kinetic argument [Stenhammar et al. 2013b, Fily et al. 2014d] which follows the fact that at finite density particles can be stuck for times $\tau_c$ of collisions with other particles. At low density, where the mean free time between collisions $\tau_f = (v_0 2a\rho)^{-1}$ exceeds $\tau_c$, the mean speed can be written as $v(\rho) = v_0(1 - \tau_c/\tau_f)$. The collision time $\tau_c$ was treated as a constant fitting parameter in Ref. [Stenhammar et al. 2013b], but generally depends on $\text{Pe}_r$, and can be controlled by two delay mechanisms: the time $a/v_0$ it takes two interacting particles to move around each other and the reorientation time $\tau_r = D_r^{-1}$. Since the collision time $\tau_c$ will be controlled by the faster of these two processes, we add the two rates to obtain $\tau_c^{-1} \sim v_0/a + D_r$. This gives $v(\rho) = v_0 [1 - \rho/\rho_*(\text{Pe}_r)]$, with

$$\rho_*(\text{Pe}_r) = \frac{c}{a^2} \left( 1 + \frac{1}{\text{Pe}_r} \right), \quad (2.22)$$

where $c$ is a number of order unity. In our simulations studying the pressure, which will be explained in more detail in the next section and in Chapter 3, we have found that this expression works well for $\text{Pe}_r > 2$, with $c \approx 0.26$ (Fig. 2.6).

We then obtain a mean-field estimate of the spinodal line as

$$\rho^\pm_c(\text{Pe}_r) = \frac{\rho_*(\text{Pe}_r)}{4} \left( 3 \pm \sqrt{1 - \left( \frac{\text{Pe}_r^c}{\text{Pe}_r} \right)^2} \right), \quad (2.23)$$

where $\text{Pe}_r^c = 4\sqrt{D_t/(a^2D_t)}$ represents the minimum $\text{Pe}_r$ required for phase separation to occur. One takeaway is that thermal diffusion suppresses phase separation and drives this critical $\text{Pe}_r$ to larger values. The condition $\text{Pe}_r > \text{Pe}_r^c$ for phase separation can also be written as $D_t < v^2/a^2D_t$, indicating that the observed spinodal decomposition requires a sufficiently small $D_t$ or long persistence time $\tau_r$.  

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The expression given in Eq. (2.23) corresponds to the dotted line in Fig. 2.5. It is, however, unclear whether thermal diffusion is really the mechanism responsible for the lower bound on $\text{Pe}_r$, given the simulations leading to Fig. 2.5 were carried out with $D_t = 0$ and do show a finite threshold. Although simulations find a finite value of $D_t$ can be generated by interactions even if $D_t = 0$, this discrepancy suggests that other mechanisms not yet understood may be at play here.

The critical density $\rho_c(\text{Pe}_r)$ for phase separation has been estimated several ways in the literature. Naively, one assumes that phase separation occurs when $\tau_f < \tau_t$, since a particle with large persistence experiences many collisions before changing direction, leading to caging by other particles. This results in $\rho_c(\text{Pe}_r) \sim (a^2 \text{Pe}_r)^{-1}$. Redner et al. obtain the same estimate via a kinetic argument that flux $\rho_{\text{gas}} v_0$ of particles that enter the cluster to the flux out of the cluster, $\kappa D_r/a$, with $\kappa$ a fitting parameter, where the dependence on $D_t$ follows the fact that particles must be turned around and pointing outward before they can leave the cluster, as shown schematically in Fig. 2.4. By equating these fluxes one can estimate the density of the gas as $\rho_{\text{gas}} = \kappa D_r/(av_0)$ and calculate the fraction $f_c$ of particles in the cluster as a function of $\rho$ ($\phi$) and $\text{Pe}_r$. The condition $f_c = 0$ gives a criterion for the phase separation, corresponding to $\rho_c(\text{Pe}_r) = \kappa/(a^2 \text{Pe}_r)$. These naive estimates predict that even at the lowest densities, some large $\text{Pe}_r$ will generate phase separation, but this behavior is not recovered in simulations which find a minimum threshold of density below which there is no phase separation.

2.4 Pressure of active Brownian particles

There has been a lot of interest in characterizing the pressure of active systems. Pressure can be defined in terms of the forces transmitted across a unit bulk plane of material. In an active fluid there is a unique contribution to pressure that measures the flux of propulsive forces across a bulk plane of material.
This contribution, named swim pressure in recent literature [Takatori et al. 2014], can be expressed via a virial-type formula as [Yang et al. 2014], [Takatori and Brady 2014], [Marchetti et al. 2016c]

\[
p^{\alpha\beta}_s = \frac{1}{dL^2} \sum_i F^{s,\alpha}_i R^{\beta}_i ,
\]

(2.24)

with indices \(\alpha\) and \(\beta\) for components of \(d\)-dimensional pressure tensor \(p^{\alpha\beta}\). Here \(d = 2\), and the scalar pressure is given by the trace \(p = p^{\alpha\alpha}\). In a closed box \(R_i = r_i\), but with periodic boundary conditions \(R_i\) is the position of the particle in an infinite system, accounting for winding numbers as the particle crosses the periodic boundary [Louwerse and Baerends 2006], [Winkler and Huang 2009]. The virial expression estimates the swim pressure as the propulsive force carried over a distance of the order of the persistence length, in analogy to the kinetic pressure of an ideal gas or the radiation pressure of a photon gas. Note, however, that in spite of the one-body expression given in Eq. (3.3), the swim pressure depends on interactions that lead to suppression of the persistence length. In addition to the swim pressure, there is also the contribution describing the direct transmission of interaction forces across the bulk plane.

A numerically more useful expression for the local swim pressure is given in terms of the flux of “active impulse” [Fily et al. 2017b], as

\[
p^{\alpha\beta}_s = \frac{1}{dA} \sum_i J^{\alpha}_i v^{\beta}_i ,
\]

(2.25)

where \(v_i\) is the velocity of particle \(i\) and \(J_i[\theta_i(t)] = F^s_i(t)\tau_i\) is the active impulse introduced in Ref. [Fily et al. 2017b], where it was shown that the expression given in Eq. (4.13) for the swim pressure is equivalent to the virial one proposed in previous work [Yang et al. 2014], [Takatori and Brady 2014]. The form in terms of active impulse is more convenient for numerical studies because it avoids the strong finite size effects that arise in calculations of the virial expression for the swim pressure [Patch et al. 2017b].
Figure 2.6: Pressure as a function of packing fraction $\phi$ from (a) our simulations of self-propelled particles with soft repulsive interactions for increasing values of $\text{Pe}_r = \ell_p/a$, obtained by decreasing $D_r$ at fixed $v_0$, and (b) sedimentation experiments of active Janus colloids by Ginot et al. [2015]. These experiments use gold microsphere half-coated with platinum and immersed in a bath of $\text{H}_2\text{O}_2$. The suspension is at an ambient temperature $T_0 = 300\text{K}$ and the various curves corresponding to increasing concentration of $\text{H}_2\text{O}_2$ resulting in increasing self-propulsion speed (from bottom to top). The pressure is extracted form measured density profiles in colloids sedimenting under gravity in a slightly tilted geometry that allows to control and reduce the strength of gravity. The lines in frame (a) are a guide to the eye. In both (a) and (b) the insets show the pressure for a dilute active gas. In both insets the straight lines are a fit to Eq. (2.27) (augmented by the thermal ideal gas pressure) with no adjustable parameters. Frame (c) shows the swim pressure versus $\phi$ for $\text{Pe}_r = 10, 20$. The solid lines are fits to Eq. (3.6) using $v(\rho) = v_0(1 - \rho/\rho^*)$ (for each $\text{Pe}_r$, $\rho^*$ is the only fit parameter). The inset shows $\rho^*(\text{Pe}_r)$ and a fit basic to (2.22). Frame (d) displays the various contributions to the pressure for $\text{Pe}_r = 20$. In all the simulations, $\tau_k = (\mu k)^{-1} = 1$ is chosen as the time unit, $a = 1$, $v_0 = 0.01$ and $D_r$ is varied to obtain the desired $\text{Pe}_r$. We follow the system for a time of $10^6\tau_k$, or equivalently at least $2 \times 10^3\tau_r$, more than enough to reach the steady state, and average over several dozen runs to obtain our error estimates.
As in passive fluids, there is an additional contribution to the pressure due to interactions. For pairwise forces $F_{ij}$, as relevant to our collection of ABPs, this can be calculated from the familiar virial expression

$$p_{D}^{\alpha\beta} = \frac{1}{dL^2} \sum_{i,j} F_{ij}^\alpha \cdot r_{ij}^\beta.$$  \hfill (2.26)

It has been shown that for the minimal model of spherical ABPs considered here the total pressure $p = p_s + p_D$ defined from Eqs. (3.3) and (3.4) coincides with the force per unit area on the walls of a container [Yang et al., 2014] and represents a state function of the active fluid [Solon et al., 2015c], independent of the properties of the walls. For an ideal active gas the swim pressure can be calculated analytically and it is given by

$$p_s^0 = \rho \frac{v_0^2}{2 \mu D_r} = \rho k_B T_a$$ \hfill (2.27)

where we now have an expression for the swim pressure of an ideal active gas that is simply the pressure of an ideal gas at temperature $T_a$.

For sufficiently large values of $\ell_p$ and $\text{Pe}_r$ the total pressure of the active fluid is non-monotonic with density (shown and discussed in Chapter 3). This arises from the strong suppression of the swim pressure following motility-induced aggregation. Ref. [Solon et al., 2015c] showed that the suppression of $p_s$ can be captured by a simple expression, given by

$$p_s = \rho \frac{v_0 v(\rho)}{2 \mu D_r},$$ \hfill (2.28)

with $v(\rho) = v_0 + \mu \langle \hat{n}_i \cdot \sum_{j \neq i} F_{ij} \rangle$, again, the effective velocity of a particle along its direction of self-propulsion. To leading order in the density, this has a linear decay $v(\rho) = v_0(1 - \rho/\rho_*)$, where $\rho_* \equiv \rho_*(\text{Pe}_r)$ provides a cutoff, above which $v(\rho)$ goes to zero. Inserting this form of $v(\rho)$ into Eq. (3.6) yields a quadratic form for the swim pressure, $p_s = k_B T_a \rho(1 - \rho/\rho_*)$ [Solon et al., 2015c], which works well for low persistence, as shown in Fig. 2.6. At higher persistence, this lends itself to an interesting non-monotonicity in pressure as a function of density, which
will be more fully discussed in my study of MIPS kinetics in Chapter 3.
Chapter 3

Kinetics and swim pressure of motility-induced phase separation

3.1 Introduction

Progress has recently been made in formulating the nonequilibrium statistical mechanics of active matter using a minimal model of active Brownian particles (ABPs) consisting of purely repulsive self-propelled spherical colloids with overdamped dynamics [Fily and Marchetti 2012a]. Perhaps the most remarkable property of this simple system is that it spontaneously phase separates into a dense liquid phase and a gas phase in the absence of any attractive interactions [Tailleur and Cates 2008, Fily and Marchetti 2012a, Fily et al. 2014c, Redner et al. 2013a, Cates and Tailleur 2015, Marchetti et al. 2016c]. This phenomenon arises from the persistent dynamics of self-propelled particles when the time for particles to reorient after a collision exceeds the mean free time between collisions, hence the name of motility-induced phase separation (MIPS). Additionally, in a sort of reverse MIPS, confined ABPs spontaneously accumulate at the walls of the container [Elgeti and Gompper 2013, Yang et al. 2014, Fily et al. 2014b], a behavior which is at odds with fundamental properties of gases and fluids in thermal equilibrium. These findings have raised a lot of interest
in understanding whether active matter can be characterized in terms of equilibrium-like properties, such as effective temperature and pressure. A broad class of active particles can exert persistent forces on the walls of a container. These forces have been quantified recently in terms of a new contribution to the pressure, dubbed swim pressure, that measures the flux of propulsive forces across a unit bulk plane of material Yang et al. [2014], Takatori and Brady [2014]. Remarkably, it has been shown that in generic active systems the mechanical force exerted on the walls of the container depends on the detail of particle-walls interactions, making it impossible to define the pressure of an active fluid as a state function Solon et al. [2015c].

This result, however, does not apply to the special case of spherical ABPs. For this minimal model it has been shown that the pressure is indeed a state function that characterizes the bulk materials properties of this simple active fluid Solon et al. [2015c]. On the other hand, the behavior of pressure of spherical ABPs is unusual, in that simulations have reported a non-monotonic behavior of pressure versus density, arising from the suppression of motility (and therefore of swim pressure) associated with particle caging at the onset of phase separation Yang et al. [2014], Winkler et al. [2015]. Experimental measurements of pressure of active colloids have similarly shown a strong suppression of pressure at intermediate density Ginot et al. [2015]. In spite of extensive work, some open question remains concerning the quantitative role of finite-size effects in active systems and the origin and robustness of the non-monotonic behavior of pressure.

In this chapter we use molecular dynamics simulations to investigate the pressure of spherical ABPs with soft repulsive forces and correlate its non-monotonic behavior with density with the kinetics of cluster size growth in phase separating systems. By examining both systems bounded by confining walls and ones with periodic boundary conditions, we show that finite-size effects are consistent in both cases with the behavior expected for a rarefied thermal gas. The pressure calculated in confined systems is strongly suppressed by the presence of boundaries if the persistence length of the particles dynamics is comparable to
the linear size of the container, with a linear dependence on system size and a behavior that resembles that of a rarified Knudsen gas. With periodic boundary conditions the convergence to the large system size limit is exponential, again as expected in a thermal system.

The non-monotonic behavior of pressure as a function of density for repulsive ABPs with large persistence lengths was first reported in simulations of systems in closed boxes [Yang et al. 2014] and has been seen in sedimentation experiments of active colloids [Ginot et al. 2015]. More recently it was confirmed in numerical studies of hard active particles with periodic boundary conditions in three dimensions [Winkler et al. 2015]. Our work provides a systematic study of finite size effects in both bounded and periodic systems and shows that the nonmonotonicity is not a finite size effect, but a bulk properties of ABPs. It arises becasue in phase separted systems the aggregate effectively provides a bounding wall for the active gas, which suppresses the swim pressure. Additionally, by considering soft repulsive disks, we complement previous work on hard particles and show that this effect does not depend on the details of the interparticle interaction. Finally, we examine the kinetics of coarsening and establish a strong correlation between the aggregation dynamics and the relaxation of the pressure towards its steady state value. We identify two dynamical regimes that control the relaxation of a disordered initial state towards the nonequilibrium steady state of the system. For system parameters that produce a homogeneous steady state, pressure grows monotonically in time to its steady state value, which it reaches on a timescale controlled by the persistence time of the self-propelled dynamics. In contrast, for parameters that produce a phase-separated steady state, the time evolution of the pressure is not monotonic and there are two dynamical regimes. Initially, small clusters form and break up rapidly and the pressure quickly builds up and overshoots its asymptotic value. Eventually, the dynamics crosses over to a coarsening regime with a slower cluster growth and the pressure relaxes to its steady-state value.

The rest of the paper is organized as follows. Section II described the ABP model and provides details of our simulations. Section III examines the swim pressure and its behavior
at low and high density. It also discusses the relevance of finite-size effects. Section IV examines correlations between the pressure relaxation and the kinetics of MIPS of swim pressure. Finally, we close in Section V with some concluding remarks.

3.2 Active Brownian Particles Model

In this chapter, we consider the minimal model of monodispersed ABPs in two dimensions [Fily and Marchetti 2012a], consisting of $N$ self-propelled disks of radius $a$ in a square box of area $L^2$, as introduced in section 2.2.2. The dynamics is described by Eqs 2.13 and 2.14 but here we neglect noise in the translational dynamics, which is less important than the orientational one in both synthetic active colloids and swimming bacteria [Marchetti et al. 2016c, Liu et al. 2017]. The equations of motion are then simply

\[ \dot{\mathbf{r}}_i = v_0 \hat{e}_i + \mu \sum_{j \neq i} \mathbf{F}_{ij} , \]  
\[ \dot{\theta}_i = \eta_i(t) , \]  

(3.1)  
(3.2)

where the pair forces between particles $i$ and $j$, $\mathbf{F}_{ij}$, are obtained from a repulsive-only harmonic potential as described in section 2.2.2.

Our simulations employ a conventional Brownian dynamics algorithm [Brańka and Heyes 1999]. We take the interaction timescale, $\tau_D = (\mu k)^{-1}$, as the unit time ($\mu = k = 1$) and the particle radius $a$ as the unit of length ($a = 1$). To prevent particles from passing through each other, we set $v_0 = (a\mu k)/100$. We choose a time step sufficiently small to handle many-body interactions at high density, $\delta t \ll \tau_D$.

As discussed in the introduction, purely repulsive ABPs exhibit macroscopic phase separation, or MIPS, where the dense phase grows to the size of the system and nearly free particles in the gas phase try to explore lengths or order $\ell_p$. For this reason, ABPs are subject to strong finite-size effects that we quantify here by considering simulation boxes of
Figure 3.1: Phase diagram of ABPs for a system of size $L = 200$ in the plane of packing fraction $\phi$ and persistence length $\ell_p$. The phase diagram is constructed by examining the probability distribution $P(\phi_W)$ of the local densities, computed by dividing the system in $N_W$ windows of size $20 \times 20$ and calculating the packing fraction $\phi_W$ in each window. The heat map shows the values of the variance of this distribution, defined as $\sigma^2_W = \frac{1}{N_W} \sum_{i=1}^{N_W} (\phi_W - \phi)^2$. In phase-separated states, the probability distribution of $\phi_W$ is bimodal (see, e.g., Redner et al. [2013a]) and $\sigma^2_W$ is large. We show with circles those values of the parameters that result in a $P(\phi_W)$ with two peaks (gas density and aggregate density).

All simulations have been run for a time $t_f$ of $10^6$ time units or longer, which in all cases is several orders of magnitude longer than the time scale of the particles’ rotational diffusion: $t_f \gtrsim 500D_r^{-1}$. To examine the time evolution of our observables, we average the relevant physical quantities over exponentially increasing time windows. In particular, we use the so-called log$_2$-binning procedure by following the evolution of averages in time intervals $I_n = (2^{-(n+1)}t_f, 2^{-n}t_f]$. The error bars are estimated from the fluctuations between several
Figure 3.2: Time evolution of the pressure of a dilute gas ($\phi = 0.01$) of ABPs for various values of the persistence length for periodic (a) and closed (b) square boxes of linear size $L = 200$. At this density the pressure is determined entirely by the swim pressure. The solid lines show the large-$L$ ideal gas prediction of Eq. (3.5), which matches the numerical results for periodic boundary conditions even at very large $\ell_p$. For closed systems the pressure is significantly suppressed by accumulation of particles at the walls as soon as $\ell_p$ becomes comparable to $L$. The insets show our estimate for the steady-state pressure $\langle p \rangle$ in the dilute limit divided by the ideal gas steady state value $p_0 = \rho v_0 \ell_p / 2 \mu$ as a function of persistence length for various systems sizes $L$. For periodic boundary conditions (a)-inset the system is self-averaging and $\langle p \rangle^{(L)} \approx p_0$ within errors for $L = 200$. For the closed box (b)-inset, there are strong finite-size effects due to the walls. We show that $\langle p \rangle / p_0$ versus $\ell_p / L$ can be fit to $1 - A \ell_p / L$ for $\ell_p / L \leq 0.1$ (black line) with a result of $A = 1.78(3)$, $\chi^2 = 7.16$ for 6 degrees of freedom (d.o.f.).

We also use this time binning procedure to assess convergence to the steady state. We consider the simulation to have reached a steady state if the averages in at least the first three $I_n$ are compatible with each other within errors. If this condition is not met, we double the simulation time. In particular for parameter values where the system phase separates, simulations have been run for $10^7$ time units.

Finally, we shall denote our estimate of the steady state ensemble average of an observable $O$ by $\langle O \rangle$, which we compute by taking the value for the $I_0$ time interval (i.e., the last half of the simulations).
3.3 Swim pressure

There has recently been a lot of interest in characterizing the pressure of active systems. Pressure can be defined in terms of the forces transmitted across a unit bulk plane of material. In an active fluid there is a unique contribution to pressure that measures the flux of propulsive forces across a bulk plane of material [Yang et al. 2014, Takatori and Brady 2015a, Mallory et al. 2014]. This contribution, named swim pressure in recent literature [Takatori et al. 2014], can be expressed via a virial-type formula as [Yang et al. 2014, Takatori and Brady 2014, Marchetti et al. 2016c]

\[ p_s = \frac{1}{dL^2} \sum_i F_{i}^s \cdot R_i , \]  
(3.3)

with \( d \) the spatial dimension (here \( d = 2 \)). In a closed box \( R_i = r_i \), but with periodic boundary conditions \( R_i \) is the position of the particle in an infinite system, accounting for winding numbers as the particle crosses the periodic boundary [Louwerse and Baerends 2006, Winkler and Huang 2009]. The virial expression estimates the swim pressure as the propulsive force carried over a distance of the order of the persistence length, in analogy to the kinetic pressure of an ideal gas or the radiation pressure of a photon gas. Note, however, that in spite of the one-body expression given in Eq. (3.3), the swim pressure depends on interactions that lead to suppression of the persistence length. In addition to the swim pressure, there is also the contribution describing the direct transmission of interaction forces across the bulk plane. For pairwise forces \( F_{ij} \), as relevant to our collection of ABPs, this can be calculated from the familiar virial expression

\[ p_D = \frac{1}{dL^2} \sum_{i,j} F_{ij} \cdot r_{ij} . \]  
(3.4)

Recent work has shown that for the minimal model of spherical ABPs considered here the total pressure \( p = p_s + p_D \) defined from Eqs. (3.3) and (3.4) coincides with the force per
unit area on the walls of a container [Yang et al. 2014] and represents a state function of the active fluid [Solon et al. 2015c], independent of the properties of the walls.

In the remainder of this section we examine the importance of finite-size effects in the calculation of the pressure of active systems and demonstrate that the predicted [Yang et al. 2014] and observed [Ginot et al. 2015] non-monotonicity of pressure versus density is an intrinsic property of these nonequilibrium fluids, not a finite-size effect.

### 3.3.1 Pressure of a dilute active gas

For a dilute gas of ABPs, the dominant contribution to the pressure comes from the swim pressure \( p_s \). Neglecting interactions, and in the large-size limit, Eq. (3.3) can be calculated exactly, with the result [Mognetti et al. 2013, Yang et al. 2014]

\[
p(t) = p_0 \left(1 - e^{-D_r t}\right), \quad p_0 = \rho \frac{v_0^2}{2\mu D_r},
\]

where \( \rho = N/L^2 = \phi/(\pi a^2) \) is the number density. The pressure \( p_0 \) of an active ideal gas can be naturally interpreted in terms of an active temperature, with \( p_0 = \rho k_B T_a \), and \( T_a = v_0^2/(2\mu k_B D_r) \). This active temperature also coincides with the one set by the ratio of translational diffusion \( D_s = v_0^2\tau_r/2 \) to the friction coefficient \( \mu^{-1} \), as required for thermal Brownian particles satisfying the Stokes-Einstein relation. Indeed, the stochastic propulsive force becomes \( \delta \)-correlated in time in the limit \( \tau_r \to 0 \), with \( k_B T_a = \text{constant} \) [Fily and Marchetti 2012a]. In other words, noninteracting ABPs behave like thermal colloid at temperature \( T_a \) in the limit of vanishing persistence time, \( \tau_r \). Here we consider the pressure of active particles for finite values of the persistence time, away from this Brownian limit.

The time evolution of \( p_s \) of a dilute active gas (\( \phi = 0.01 \)) is shown in Fig. 3.2 for a system in a box of linear size \( L = 200 \) with periodic boundary conditions (a) and one enclosed by bounding walls (b), for various values of the persistence length, \( \ell_p \). Flat repulsive walls are implemented using the same harmonic forces that describe interparticle interactions.
Figure 3.3: Pressure as a function of packing fraction for \( \ell_p/a = 25 \) (a) and \( \ell_p/a = 50 \) (b) for a system with periodic boundary conditions. The box size must be several times larger than \( \ell_p \) to obtain results representative of the large-\( L \) limit.

For periodic boundary conditions, Eq. (3.5) provides an excellent fit to the calculated pressure over the entire time range and for all \( \ell_p \) considered. In confined systems, however, a second time scale comes into play, that is the time \( \tau_L = L/v_0 \) it takes an ABP to travel the size of the box. The finite-size corrections to pressure are negligible only when \( \tau_L \gg \tau_r \), or equivalently \( L \gg \ell_p \) (here with \( L = 200 \) this only holds for the smallest \( \ell_p = 5 \)). In all other cases the presence of confining walls strongly suppresses the asymptotic long-time value of the pressure as compared to the value \( p_0 \) given by Eq. (3.5).

To quantify the finite-size corrections, the insets of Fig. 3.2 show the ratio \( \langle p \rangle / p_0 \) of the calculated steady-state pressure to the ideal active gas pressure as a function of \( \ell_p \) for various systems sizes \( L \). With periodic boundary conditions (a), the finite-size corrections are essentially independent of \( \ell_p \) and converge very quickly to \( p_0 \) with increasing system size—in fact for \( L = 200 \) \( \langle p(\ell_p) \rangle / p_0 \) is indistinguishable from 1 with our errors.

In the case of the closed box the finite-size effects are much more pronounced and depend strongly on \( \ell_p \), as shown in the inset of Fig. 3.2b. Our \( \langle p \rangle / p_0 \) for different \( L \) can be nearly collapsed when plotted versus \( \ell_p/L \). In fact, for \( \ell_p/L \lesssim 0.1 \), the behavior can be fitted by a linear form, \( \langle p \rangle / p_0 \approx 1 - A \ell_p/L \). This functional dependence supports the idea that the box boundary has a simple surface effect on the pressure for \( \ell_p/L \ll 1 \). At higher persistence
Figure 3.4: Swim pressure $\langle p_s \rangle$ (a) and cluster fraction $\langle N_c \rangle/N$ (b) versus packing fraction $\phi$ for several values of $\ell_p$ (we use $L = 200$ for $\ell_p/a \leq 25$ and $L = 400$ for $\ell_p/a = 50, 100$). In (a), solid lines show fits to $\langle p_s \rangle = p_0(\phi)(1 - \phi/\phi_0)$, where $\phi_0$ is the only adjustable parameter (see Ref. Marchetti et al. [2016c]). This quadratic behavior breaks down for higher $\ell_p$. In both frames, dashed lines provide a guide for the eye.

lengths, collective effects, such as clustering of particles at the corners of the box, yield further deviations from the linear behavior.

In summary, we find that at low density the swim pressure is clearly suppressed in systems that restrict the mean free path below $\ell_p$, so that the dynamics remains ballistic at all times. When particles are confined, there are strong boundary effects reminiscent of those seen in a Knudsen gas, where the density is so low that the mean-free path from inter-particle collisions exceeds the box size. Specifically, the convergence to the large-$L$ limit is exponential for the periodic box and linear for the closed box. This is precisely the type of the finite-size effects one expects in a thermal system whose free energy would have bulk and surface contributions, with the former the only relevant one in periodic systems at large $L$.

### 3.3.2 Pressure at finite density

At finite density, both the swim contribution and the direct contribution from interactions are appreciable. The direct contribution form interactions, $p_D(\phi)$, grows monotonically with density as in passive systems and depends only weakly on self propulsion Yang et al. [2014]. In contrast the swim pressure is non-monotonic in density and strongly suppressed at inter-
mediate density due to the decrease of particle motility. This leads to an overall non-convex density dependence of the total pressure $\langle p(\phi) \rangle$, which has been seen in simulations \[ \text{Yang et al. } 2014, \text{ Solon et al. } 2015c, \text{ Winkler et al. } 2015 \] and experiments \[ \text{Ginot et al. } 2015 \].

For larger $\ell_p$ simulations show a non-monotonic $\langle p(\phi) \rangle$.

When the system phase separates, the macroscopic aggregate effectively provides a bounding wall to the gas phase, leading to strong finite-size effects in the swim pressure even in systems with periodic boundary conditions. This is shown in Fig. 3.3. Here, the bottom frame ($\ell_p/a = 50$) shows a system which undergoes phase separation for intermediate densities, while in the top frame ($\ell_p/a = 25$) the phase separation is still incipient (recall Fig. 3.1). We note that in the former case we need box sizes of $L \geq 200$ to achieve convergence \[ \text{1} \].

All the results reported in the remainder of the paper are for box sizes $L = 200, 400$, and are free of finite-size effects within our error bars.

For sufficiently large values of $\ell_p$ the pressure of the active fluid is non-monotonic with density. This behavior, shown in Fig. 3.3b, is not a finite-size effect and is associated with the strong suppression of the swim pressure arising from motility-induced aggregation. Ref. \[ \text{Solon et al. } 2015c \] showed that the suppression of $p_s$ can be captured by a simple expression, given by

$$
p_s = \rho v_0 v(\rho) \frac{\mu \langle \hat{e}_i \cdot \sum_{j \neq i} F_{ij} \rangle}{2 \mu D_r}, \tag{3.6}
$$

with $v(\rho) = v_0 + \mu \langle \hat{e}_i \cdot \sum_{j \neq i} F_{ij} \rangle$ the effective velocity of a particle along its direction of self-propulsion. To leading order in the density, this has a linear decay $v(\rho) = v_0(1 - \rho/\rho_*)$, where $\rho_* \equiv \rho_*(\ell_p)$ provides a cutoff, above which $v(\rho)$ goes to zero. Inserting this form of $v(\rho)$ into Eq. \[ 3.6 \] yields a quadratic form for the swim pressure, $p_s = k_B T a \rho (1 - \rho/\rho_*)$ \[ \text{Solon et al. } 2015c \].

As we can see in Fig. 3.4a, this ansatz works well only for moderate $\ell_p$ ($\ell_p/a \lesssim 25$ with our parameters). For large $\ell_p$, i.e., for systems exhibiting phase separation (see Fig. 3.1), the

\[ \text{1} \] At very high packing fraction, approaching or even surpassing the closed-packed limit, there are larger discrepancies, but we do not consider this regime here.
swim pressure displays a much stronger dependence on density and drops abruptly at the onset of phase separation. Phase separating systems always evolve to contain a single large dense region.

The relation between pressure and particle aggregation is shown in Fig. 3.4. The bottom frame displays the total number of particles $N_c$ that belong to clusters above a certain threshold size (we use a cutoff of 100) \footnote{A particle $i$ is part of cluster $C$ if it is interacting with any other disks in that cluster (i.e., if $r_{ij} \leq 2a$ for any $j \in C$).} The sharp drop in pressure shown in Fig. 3.4a at large values of $\ell_p$ corresponds to a jump in the value of $N_c$, signaling the onset of phase separation. A similar result has been obtained for hard repulsive spheres in $d = 3$ Winkler et al. [2015].
Figure 3.6: Snapshots (a–e) show a single run, for times that correspond to the points in the (f), labeled respectively. Time evolution of pressure $p$ (red squares), total cluster fraction $N_c/N$ (blue lines), and largest cluster fraction $N_1/N$ (blue circles) for a system with size $L = 400$, $\phi = 0.35$, and $\ell_p = 50$. $N_c/N$ is shown in (f) for 10 individual runs to emphasize its robust steady state. The overshoot in pressure results from the onset of clustering, which dampens the swim and total pressure in the long-time limit. The steady state of pressure corresponds with the steady state of $N_c$, while $N_1$ continues to grow as the system coarsens.

3.4 Kinetics of Motility-Induced Phase separation

The time evolution of the pressure from a random initial condition towards the final steady state reveals a non-monotonic dynamics directly related to the kinetics of MIPS. This is displayed in Fig. 3.5. For parameter values corresponding to homogeneous steady states, the pressure evolves monotonically in time, reaching its steady state value in a time of the order of the persistence time, $\tau_r$. For parameter values corresponding to phase separated steady states, however, the convergence to the steady state is delayed by cluster nucleation and aggregation, which operate on a density-dependent timescale much longer than rotational diffusion. This results in the pressure temporarily overshooting its steady-state value, as
shown in Fig. 3.5. The total time required for the pressure to reach steady state depends on density and, not surprisingly, is longest for the lowest densities that exhibit phase separation. The growth of the dense phase is associated with the formation of clusters of jammed particles. If the reorientation time $\tau_r$ required for particles to turn and escape from the cluster is longer than the mean free time between collisions, more particles will accumulate and the cluster will grow. This process continues as long as the rate for escaping from the cluster ($D_r$), is slower than the rate at which particles absorb at the boundary, which is controlled by the collision rate. Systems with lower densities take a longer time to reach the final state due to lower nucleation and adsorption cross-sections. This is in agreement with recent work examining the kinetics of MIPS in terms of classical nucleation theory [Redner et al. 2016].

By comparing the evolution of pressure and of density correlations in our largest phase-separating systems ($L = 400$), we can identify two distinct dynamical regimes. Once the pressure reaches its steady state value, clusters begin to coarsen at a slower rate, corresponding to previous observations [Redner et al. 2013b, Stenhammar et al. 2013a].

By the time pressure equilibrates, multiple clusters have formed and there is an average zero net flux between phases, shown in Fig. 3.6 where we additionally quantify the number of particles in the largest cluster $N_1$ alongside all clustered particles $N_c$. At this stage, the dense aggregates move very slowly, having caged the motility of most aggregated particles, only allowing those at the boundary to de-adsorb, while large clusters coalesce into one another until system-spanning phase separation occurs. We find a division in time regimes of growth for $N_c(t)$ and $N_1(t)$ and show snapshots of this process in Fig. 3.6. The largest cluster continues to grow long after the cluster fraction saturates, which corresponds with pressure reaching its steady state.

In particular, for the data shown in Fig. 3.6 we have fit the number of particles in the largest cluster to

$$N_1(t) \sim t^\gamma.$$  \hspace{1cm} (3.7)

In the first time regime (up to and including point B in the figure, that is, for $tD_r < 20$) this
fit gives $\gamma_1 = 1.3(2), \chi^2$/d.o.f. = 0.12/1. In the coarsening regime, for $t_{D_f} > 20$, we obtain $\gamma_2 = 0.29(4), \chi^2$/d.o.f. = 1.65/3. These fits are plotted with dotted lines in the figure.

We note that the coarsening regime has been studied in previous work [Redner et al. 2013b, Stenhammar et al. 2013a] using the growth of a length scale computed from the structure factor: $L(t) \propto t^\alpha, \alpha \approx 0.28$. If we reproduce the analysis of $L(t)$ in Stenhammar et al. 2013a with our data we obtain $\alpha = 0.26(2), \chi^2$/d.o.f. = 1.82/3 for $t_{D_f} > 20$.

Finally, we compare the approach to the steady state to the dynamics of pressure fluctuations in the steady state by computing the time autocorrelation function of the instantaneous mean pressure, given by

$$C_p(t) = \frac{\langle p(s)p(s+t) \rangle - \langle p \rangle^2}{\langle p^2 \rangle - \langle p \rangle^2}. \tag{3.8}$$

This function is shown in Fig. 3.7 for $\ell_p = 25$ and $\phi = 0.4$. The correlation function

---

Figure 3.7: Correlation time $\tau$ for $\ell_p = 25, 50, 100$ computed for the correlation function, Eq. (3.8). Inset shows an example of the steady-state correlation function, for $\phi = 0.4$ and $\ell_p = 25$. Even though the time needed to reach the steady state depends on $\phi$ (Figure 3.5), once the steady-state has been reached $\tau$ is equal to the single particle persistence time, $\tau = \tau_r = \ell_p/v_0$. 

We note that the coarsening regime has been studied in previous work [Redner et al. 2013b, Stenhammar et al. 2013a] using the growth of a length scale computed from the structure factor: $L(t) \propto t^\alpha, \alpha \approx 0.28$. If we reproduce the analysis of $L(t)$ in Stenhammar et al. 2013a with our data we obtain $\alpha = 0.26(2), \chi^2$/d.o.f. = 1.82/3 for $t_{D_f} > 20$.
decays exponentially, allowing us to extract a relaxation time $\tau$,

$$C_p(t) \simeq e^{-t/\tau},$$

(3.9)

We find that $\tau$ does not depend on density and coincides with $\tau_r$ for all our runs (Fig. 3.7). Therefore, even though the time evolution of the pressure in the approach to the steady state is density dependent, once the steady state has been reached the only time scale for pressure fluctuations is given by the single-particle rotational diffusion.

### 3.5 Conclusions

We have examined the effects of finite system size and of the kinetics of MIPS on the pressure of ABPs in two dimensions. In a dilute gas of ABPs the finite-size effects on pressure for both open (periodic) and closed (particles in a square box) boundary conditions are quantitatively similar to what one would expect for a thermal gas. At finite density, finite-size effects are pronounced even for the periodic case. We find that the box size has to be several times larger than the persistence length of the particles to obtain results representative of bulk behavior. This has implications for studies in strip geometries [Bialké et al. 2015], where a careful control is needed to avoid spurious anisotropies in the stresses.

For parameter values corresponding to MIPS, we have examined the correlation between the relaxation of the mean pressure to its steady state value and the kinetics of clustering and phase separation. In this regime, the interplay between the decreasing swim pressure and the increasing direct pressure from interaction in the incipient clusters results in long, density-dependent time scales for approach to the steady state. The phase separation process shows two distinct dynamical regimes: a rapid growth corresponding to the formation of small clusters, followed by a slower regime of cluster coalescence and coarsening. These two regimes are reflected in the time evolution of the pressure that first grows rapidly during the small cluster nucleation when it is mainly controlled by the swim pressure of the gas,
even overshooting its steady state value, and then remains constant during coarsening when
the net flux of particles between the dense and dilute phases vanishes. The overshoot of the
swim pressure upon phase separation is a distinctive feature of active particles associated
with crowding of the gas at the boundary of the dense phase. The cluster provides an
effective bounding wall to the active gas, strongly suppressing the swim pressure. This effect
has no analogue in thermal systems where the kinetic contribution to the pressure is not
affected by interactions nor by interfaces.

The total pressure, has, however, been shown to remain equal across the two phases,
confirming that this minimal model can be described in terms of an effective thermodynamics.
The relationship between the kinetics of MIPS and the relaxation of pressure to its steady
state value additionally validates the use of equilibrium-like ideas as done in Ref. [Redner
et al.] [2016] to describe the coarsening kinetics.
Chapter 4

Tension and dynamics at the interface of motility-induced phases

4.1 Introduction

Pierre-Gilles de Gennes wrote [2005] that “the interfaces between two forms of bulk matter are responsible for some of the most unexpected actions... the overlap region is mobile, diffuse, and active.” This description is particularly apt as applied to the emergent behavior of dense collections of active Brownian particles (ABPs), in which purely repulsive particles are driven out of equilibrium via self-propulsive forces in an overdamped environment [Fily and Marchetti 2012a]. Even in the absence of attractive interactions, such systems can spectacularly phase separate into a dense liquid phase coexisting with a dilute gaseous phase [Tailleur and Cates 2008, Fily and Marchetti 2012a, Redner et al. 2013a, Cates and Tailleur 2015]. This motility-induced phase separation (MIPS) is heuristically understood by considering the persistent dynamics of an individual particle, and it occurs when the time for a particle to re-orient after a collision becomes long relative to the typical mean free time between those collisions. The occurrence of MIPS has also been described through an approximate mapping onto an effective equilibrium system undergoing conventional vapor-
liquid phase separation. Wittkowski et al. [2014], Fily et al. [2014d], Solon et al. [2015c,b], Cates and Tailleur [2015], Winkler et al. [2015], Takatori and Brady [2015a], Marini Bettolo Marconi et al. [2016], Solon et al. [2018]. It has been demonstrated in numerical simulations in two and three dimensions for various minimal models and repulsive potentials. Fily and Marchetti [2012a], Redner et al. [2013b], Bialké et al. [2013], Yang et al. [2014], Takatori and Brady [2015b]. Experiments in active colloids and bacterial suspension, however, generally observe the formation of finite-size clusters rather than bulk phases, suggesting that non-generic phenomena may be at play and arrest the phase separation. Theurkauff et al. [2012], Buttinoni et al. [2013], Palacci et al. [2013], Mallory et al. [2014], Bechinger et al. [2016], Liu et al. [2017].

A typical snapshot of such an out-of-equilibrium phase separation is shown in Fig. 4.1, where the enormous fluctuations characteristic of MIPS are readily seen. Large fluctuations occur both at the interface and in the bulk of the dense phase, where bubbles of the dilute phase spontaneously nucleate and travel to the phase boundary, breaking at the surface (see SI for a video). Despite the wildly fluctuating nature of the interfaces, connections with interfacial properties of equilibrium phases have been identified. Bialké et al. [2015], Speck [2016], Lee [2017], Marini Bettolo Marconi et al. [2016], Paliwal et al. [2017], Solon et al. [2018], Tjhung et al. [2018]. For example, as we describe in more detail below, the scaling of the interfacial stiffness with system size is found to be consistent with equilibrium arguments. There is, however, a major caveat: the measured interfacial tension $\gamma$ is negative [Bialké et al. 2015, Lee 2017, Solon et al. 2018], and the equilibrium arguments connecting it to the interface stiffness require one to take $|\gamma|$ as the relevant quantity [Speck 2016].

How can we reconcile a stable, equilibrium-like interface with negative values of surface tension, especially in a system driven by far-from-equilibrium dynamics? In this work we rely on extensive simulations to study the structure of the MIPS interface. We show that for such strongly fluctuating interfaces – where the instantaneous deviation of the interfacial height from its average value is decidedly not small – a transformation to the local coordinate
frame along the interface (illustrated in Fig. 4.1) reveals surprising additional structure. An important finding is a strong correlation between the local curvature and the magnitude of the surface tension. By examining in detail the local dynamics near the fluctuating interface, we demonstrate the existence of surface layers with large local tangential particle motion in both the dilute and the dense regions. While such tangential currents in the interfacial region within the gas have been highlighted before, the local “self-shearing” of the surface layer in the dense phase is a new result. This observation suggests a new mechanism for the stabilization of active interfaces where the interface directs particles along itself to heal fluctuations.

Below, we first describe our simulation model and study the properties of the interface in a global reference frame, in line with other studies. Bialké et al. [2015], Speck [2016], Lee [2017], Solon et al. [2018] In addition to expanding on previous results for the scaling of the interfacial width with simulation size, we directly study the spectral density of the interface fluctuations. Despite the non-equilibrium character of the system, this spectrum is surprisingly equilibrium-like. In this global frame we also study the (mechanically defined) value of the interfacial tension over a wide range of parameters, varying the persistence length of the particles. We confirm the unusual result that the interfacial width scales with a stiffness proportional to the absolute value of a negative $\gamma$.

We then shed new light on the mechanism of interfacial stability by shifting to a local frame defined along the interface, utilizing an algorithmically traced interface contour with single particle resolution. In this frame, it is straightforward to define normal and transverse particle fluxes and forces. Using this technique we quantify the strong correlations between the local curvature of the interface and the value of the surface tension: although we find that the tension is always negative, it is closest to zero in regions of large positive curvature (inward “valleys”) and most strongly negative in regions of large negative curvature (outward “mountains”).

We close by presenting a heuristic picture of the emergent collective behavior of self-
Figure 4.1: Top: A snapshot of a system composed of roughly $2 \times 10^5$ active Brownian particles of radius $r_0$ undergoing spontaneous separation into dense and dilute phases. The persistence length of the particles (defined in the text) is $\ell_p = 100r_0$; the area fraction is $\phi = 0.5$ and phase separation into a strip geometry is attained by choosing the aspect ratio of the simulation box to be $L_x/L_y = 2$, with periodic boundary conditions (here $L_x = 1600r_0$). Bottom left: A demonstration of two methods for identifying the interface, with black scale bar of length $\ell_p = 100r_0$. The pixelated red curve results from a contour-finding algorithm that captures all overhangs and allows for local curvature measurements, while the smoother blue curve considers only the outermost particles at each value of $y$ and can be used to obtain the spectrum of the interface height fluctuations (see Appendix A for details). Bottom right: A schematic of the the local frame we use to measure dynamical quantities near the interface (see Appendix B for details).

propelled particles near the interface, highlighting how their dynamics produces a negative interfacial “tension”, resulting in an effectively “extensile” interface that tends to grow longer, while maintaining its integrity. This is in sharp contrast with familiar equilibrium interfaces that are “contractile” in the sense that the positive tension always tends to shorten the
interface. These interfaces have been phenomenologically interpreted in terms of Edwards-Wilkinson-like growth processes [Edwards et al. 1982, Lee 2017] but the dependence of the surface tension on local curvature naturally leads to simple KPZ-like equations [Kardar et al. 1986]. We attempt different scaling collapses of our data for interfacial roughening in an effort to discriminate between these scenarios.

4.2 Model and Methods

We simulate a system of active Brownian particles (ABPs) in a regime in which they are known to undergo motility-induced phase separation (MIPS). We choose a strip geometry for ease of identification of the interface. We identify the dense phase as all members of the largest set of touching particles, while the gas phase is composed of those remaining particles.

4.2.1 Active Brownian Particle Model

A minimal model of monodisperse, purely repulsive ABPs [Fily and Marchetti 2012a] consists of $N$ self-propelled particles with interaction radius $r_0$, as introduced in Section 2.2.2.

We place these in a rectangular simulation domain of sides $L_x$ and $L_y$ with periodic boundary conditions, setting $L_x/L_y = 2$ so that the bulk phases yield a quasi-1D interface (see Fig. 4.1). The dynamics is described by Eqs 2.13 and 2.14, but here again we neglect noise in the translational dynamics, which is less important than the orientational one in relevant experimental systems [Marchetti et al. 2016c, Liu et al. 2017]. Particles are thus simply governed by the overdamped Langevin equations

$$\dot{\mathbf{r}}_i = \mu \left( \mathbf{F}^s_i + \sum_{j \neq i} \mathbf{F}_{ij} \right), \quad (4.1)$$

$$\dot{\theta}_i = \sqrt{\frac{2}{\tau_r}} \eta_i(t). \quad (4.2)$$
where the pair forces between particles $i$ and $j$, $F_{ij}$, are obtained from a repulsive Weeks-Chandler-Anderson potential, as described in section 2.2.2.

### 4.2.2 Capillary waves and interface width

Taking a mesoscopic view of the interface, one can characterize the fluctuations in terms of the deviations of the instantaneous location of the surface along the $x$ direction from its mean value. Rowlinson and Widom [2013] $\delta h(y, t) = h(y, t) - \bar{h}(t)$, with $\bar{h}(t) = \frac{1}{L_y} \int_0^{L_y} dy \ h(y, t)$. The mean interfacial width can then be written as,

$$w^2 = \frac{1}{L_y} \int_0^{L_y} dy \ \langle |\delta h(y)|^2 \rangle = \sum_q \langle |\delta h(q)|^2 \rangle , \quad (4.3)$$

where

$$\delta h(q) = \frac{1}{L_y} \int_0^{L_y} dy \ \delta h(y)e^{-i q y}. \quad (4.4)$$

In thermal equilibrium, interfaces carry an excess free energy $E_s = \gamma \ell$ determined by the constant interfacial tension, $\gamma$, and the length $\ell$ of the interface, with

$$\ell = \int_0^{L_y} dy \ \sqrt{1 + |\nabla_y h(y)|^2} \approx L_y \left[ 1 + \frac{1}{2} \sum_q q^2 |\delta h(q)|^2 \right] . \quad (4.5)$$

The interfacial height mode amplitudes are then determined by the equipartition theorem as

$$\langle |\delta h(q)|^2 \rangle = \frac{2}{L_y} \frac{kT}{\gamma q^2} . \quad (4.6)$$

Substituting Eq. (4.6) into Eq. (4.3), we can immediately calculate the interface width as

$$w^2 = w_0^2 + \sum_{q > 0} \langle |\delta h(q)|^2 \rangle = w_0^2 + \frac{L_y}{12 \sigma} ; \quad (4.7)$$
where $w^2_0$ describes the fluctuations of the $q = 0$ mode and $\sigma = \gamma/kT$ is the interfacial stiffness that measures the cost of deformations along the entire length $L_y$.

Note that the preceding analysis is carried out under the assumption that $\delta h \ll 1$. Even though the interfaces in MIPS systems are characterized by extremely large fluctuations, we will find that the scaling of the width the system size and the Fourier spectrum of the interfacial modes are surprisingly well-described by these equilibrium, small-fluctuation expressions.

### 4.2.3 Interfacial tension

A mechanical definition of the interfacial tension $\gamma$ can be obtained by examining the work $\delta W = \gamma \delta \ell$ needed to change the length of the interface by an amount $\delta \ell$. Here we follow the standard quasi-thermodynamic treatment of Ref[Rowlinson and Widom 2013], where in equilibrium it is shown that this mechanical definition yields the same value as that obtained from the interfacial fluctuations. As we will see, this is not, however, the case for ABPs [Bialké et al. 2015], [Lee 2017].

Working in two dimensions, we consider a one-component system confined to a box of area $A = \ell^2$ and separated into two bulk phases, with a vertical interface at some position $0 < x_0 < \ell$. If the area of the system is changed isotropically by an amount $\delta A$, the associated work is controlled by the pressure, with $\delta W = -p \delta A$. To define the tension we imagine isothermally and reversibly deforming the sides of the confining box so that the interface increases in length by an amount $\delta \ell$, while maintaining fixed area. This requires an anisotropic deformation of the box, but the symmetry of the interface ensures that the pressure tensor can only have non-zero components $p_{xx}$ and $p_{yy}$. The tangential work done to the system in increasing the length of the interface is then

\[ \delta W_t = -\delta \ell \int_0^\ell dx p_{yy}(x) \quad (4.8) \]
and the normal work done in keeping the area fixed is

$$\delta W_n = \ell \delta \ell p_{xx}. \quad (4.9)$$

In writing the above expression we have assumed mechanical stability of the interface, $\nabla \cdot p = 0$. This, together with the symmetry of the system ensures that $p_{xx}$ is not itself a function of $x$ or $y$. The total work done is then

$$\delta W = \delta W_n + \delta W_t = \delta \ell \int_0^\ell dx \left( p_{xx} - p_{yy}(x) \right). \quad (4.10)$$

Comparing this expression with $\delta W = \gamma \delta \ell$ we recover the Kirkwook-Buff expression\cite{Kirkwood1949},

$$\gamma = \int_{\text{dilute}}^{\text{dense}} dx \left[ p_n - p_t(x) \right], \quad (4.11)$$

where we have more generally replaced Cartesian components of the tensor with normal and tangential components, and assumed that the two phases we are considering are a dense and dilute phase.

Equation \ref{eq:gamma} quantifies the surface tension as the total anisotropy in pressure across the interface. It assumes of course that the local pressure tensor has a mechanical definition. In the next section we outline how we measure the local pressure tensor for our active system and compute Eq. \ref{eq:gamma}.

### 4.2.4 Pressure in Active Matter

The pressure $p$ of a system of overdamped torque-free active particles involves two contributions: a contribution due to direct particle interactions, $p_d$, and a “swim” contribution that represents the flux of propulsive forces across a unit area, $p_s$, with $p = p_d + p_s$. The components of the interaction pressure tensor $p_d^{\alpha \beta}$, where $\alpha$, $\beta$ denote Cartesian indices, are
given by a virial expression in terms of particle pair interactions,

\[ p^{\alpha\beta} = \frac{1}{dA} \sum_{i,j} F^{\alpha}_{ij} r^{\beta}_{ij}. \]  

(4.12)

The swim pressure can also be calculated via a virial-like expression. Yang et al. [2014], Marchetti et al. [2016b], Patch et al. [2017a] A numerically more useful expression for the local swim pressure is given in terms of the flux of “active impulse”, as

\[ p^{\alpha\beta}_s = \frac{1}{dA} \sum_i J^{\alpha}_i v^{\beta}_i, \]  

(4.13)

where \( v_i \) is the velocity of particle \( i \) and \( J_i[\theta_i(t)] = F^s_i(t)\tau_r \) is the active impulse introduced in Ref. Fily et al. [2017b], where it was shown that the expression given in Eq. (4.13) for the swim pressure is equivalent to the virial one proposed in previous work. Yang et al. [2014], Takatori and Brady [2014] The form in terms of active impulse is more convenient for numerical studies because it avoids the strong finite size effects that arise in calculations of the virial expression for the swim pressure. Patch et al. [2017a]

Since there are many quantities considered in this work, we summarize them in table 4.2.4.

<table>
<thead>
<tr>
<th>name</th>
<th>symbol</th>
<th>normal</th>
<th>tangential</th>
</tr>
</thead>
<tbody>
<tr>
<td>swim pressure</td>
<td>( p_s \sim \mathbf{e} \cdot \mathbf{v} )</td>
<td>( p_n \sim e_n v_n )</td>
<td>( p_t \sim e_t v_t )</td>
</tr>
<tr>
<td>current density</td>
<td>( \mathbf{I} = \phi \mathbf{v} )</td>
<td>( I_n )</td>
<td>( I_t )</td>
</tr>
<tr>
<td>swim current density</td>
<td>( \mathbf{I}^s = \phi v_0 \mathbf{e} )</td>
<td>( I^s_n )</td>
<td>( I^s_t )</td>
</tr>
</tbody>
</table>

Table 4.1: A summary of the various quantities used in the text and figures.

### 4.2.5 Simulations

Simulations of large phase-separating systems of ABPs require total running times \( t \gg \tau_r \) and integration timesteps \( \Delta t \ll \tau_r \). Patch et al. [2017a] This, in addition to the wildly fluctuating nature of the interfaces and the need to sweep different \( \ell_p \) values, means that
Figure 4.2: Top: density profiles in the interface frame, for varying persistence. [Inset] Scaling of \( w^2 \) as a function of \( L_y/\ell_p \), for comparison to Eq. 4.7. Bottom: Interface height fluctuation spectrum for systems with \( L_y = 400 \), collapse for several values of \( \ell_p \). The equilibrium expectation \( \langle |\delta h(q)|^2 \rangle \sim q^{-2} \) is shown as a solid black line. The collapse of \( \ell_p |\delta h(q)|^2 \), signifies the clear dependence of interfacial fluctuations on persistence. [Inset] Best fit exponent characterizing the initial decay of the interface modes as a function of \( \ell_p \).

considerable computational effort must be made to gather an adequate statistical sample. Most of our simulations have been carried out on GPUs using the HOOMD-blue\textsuperscript{Anderson et al.} \textsuperscript{2008}, hoo \textsuperscript{2017} simulation package, complementing with CPUs for the smaller system sizes.

To suppress transients, we nucleate the phase-separated strip by initially placing particles in a triangular lattice with interparticle distance \( r_{ij} = 2r_0 \). We performed parameters sweep by using persistence lengths \( \ell_p = \{60, 80, 100, 120, 140, 200\} \) and system sizes \( L_x = \{200, 400, 600, 800, 1600\} \). The area fraction was fixed at \( \phi = 0.5 \), with \( \mu = 1 \) and \( v_0 = 100 \). We have simulated for \( t \geq 1 \times 10^3 \tau_r \), with a number of independent runs \( N_{\text{runs}} = 100 \) for our smallest systems, down to \( N_{\text{runs}} = 8 \) for \( L_x = 800 \). Unless stated otherwise, the results in this chapter refer to \( L_x = 800 \) systems. In order to study the interface growth, we have also run shorter simulations \( (t = 100\tau_r) \) that do not reach the steady state, for system sizes ranging from \( L_x = 400 \) (\( N_{\text{runs}} = 400 \)) to \( L_x = 2400 \) (\( N_{\text{runs}} = 28 \)).

We note that previous studies have found strong finite-size effects when studying the pressure of phase-separated systems with \( \ell_p \sim L \), even at low density\textsuperscript{Patch et al.} \textsuperscript{2017a}. Measuring the swim pressure with the active impulse flux, Eq. (4.13), rather than with a
Figure 4.3: The main figure correlates the interfacial tension (defined via Eq. 4.11) and the stiffness (defined via Eq. 4.7), showing that the tension becomes more negative with increasing stiffness. This suggests that the same dynamics that produces a positive stiffness yields a negative interfacial tension. The black points correspond to measurements in the global frame (described in Sec. 4.3), while the blue points correspond to measurements in the local frame of the interface (described in Sec. 4.4). The solid lines are linear fits to $\gamma(\sigma) = A + B\sigma$. The inset shows the difference between normal and tangential pressure that determines the integrand in Eq. 4.11, broken down into cluster and gas contributions, highlighting the significant net-negative contributions from both cluster and gas particles. The dotted line marks the location of the mean interface determined using Eq. 4.14.

virial expression, mitigates this effect. Nevertheless, we have kept the values of $\ell_p$ smaller than $L_x$ in our work.

### 4.3 Measurements in the global frame

The MIPS interface is characterized by wild fluctuations (as seen in Fig. 4.1). Upon time-averaging, however, the interface appears deceptively well-behaved and equilibrium-like. In this section we show this by characterizing the average density profile, determining the stiffness of the interface from the scaling of the interfacial width, and investigating the spectrum of interface fluctuations. We will often refer to the system as divided into two sets
of particles: those belonging to the largest connected cluster (dense) and those in the gas (dilute).

4.3.1 Interface width and stiffness

Working in the global frame, we first consider the average area fraction projected onto the $x$-axis, $\phi(x) = \frac{1}{L_y} \int_0^{L_y} dy \phi(x, y)$. The resulting profile is well described by

$$\phi(x) = \frac{\phi_+ + \phi_-}{2} + \frac{\phi_+ - \phi_-}{2} \tanh \left( \frac{x - x_0}{2w} \right), \quad (4.14)$$

where $\phi_\pm$ denotes the area fraction for the dense and dilute phases, $x_0$ is the mean position of the interface, and $w$ is a measure of the interfacial width. In broad agreement with previous observations, Cates and Tailleur [2015] we find that with increasing $\ell_p$ the interfacial width decreases and the difference in density between the coexisting phases increases, as we show in the upper panel of Fig. 4.2. The inset to the upper panel of Fig. 4.2 also shows that $w^2 \sim L_y$ for a variety of $\ell_p$.

We note that the finite intercept at $L_y = 0$ provides an estimate of the minimum system size needed to observe MIPS in these systems. In the following, $\gamma$ is calculated from its mechanical definition (Eq. (4.11)), while $\sigma$ is extracted from Eq. (4.7) by fitting the scaling behavior of the interface width with system size $L_y$ (i.e., it is obtained from the slope of the linear curves in the inset of the top frame of Fig. 4.2).

4.3.2 Interface fluctuations

Although the MIPS interface is extremely rough and is characterized by frequent overhangs, to quantify the structure of the interfacial fluctuations we approximate the interface by constructing a height map $h(y)$ as described in Appendix A. We then examine the Fourier spectrum of $\delta h(y) = h(y) - \bar{h}$, where $\bar{h}$ is the instantaneous value of the mean position of the height map, and average the resulting mode spectrum $\langle |\delta h(q)|^2 \rangle$ over time in the
steady state. The spectrum of interfacial fluctuations shown in Fig. 4.2 is well described by
\( \langle |\delta h(q)|^2 \rangle \sim \ell_p^{-1} q^{-\beta} \), in keeping with our finding that the interfacial width itself scales with \( \ell_p \). We do observe small deviations from the strictly equilibrium expected scaling of \( \beta = 2 \) (see inset of lower panel of Fig. 4.2).

Interfacial fluctuations have recently been measured in a mixture of active and passive disks with attractive interactions\( \text{[del Junco and Vaikuntanathan 2018]} \). In that case the attractions stabilizes the interface and activity enhances the stiffness that grows linearly with \( \ell_p \).

### 4.3.3 Mechanical surface tension

As discussed in Section 4.2.3, a straightforward mechanical measurement of the surface tension involves integrating differences in the local pressure tensor across the interface,\( \text{[Bialké et al. 2015]} \) via
\[
\gamma = \int dx \left( p_n - p_t \right) \text{ (Eq. 4.11)}. \]

In the global frame one simply has \( p_n = p_{xx} \) and \( p_t = p_{yy} \). At an equilibrium liquid-gas interface the positive surface tension arises from the lowering of tangential pressure associated with the weaker binding of liquid surface molecules as compared to bulk liquid molecules. In our active system, in contrast, we find a large increase of tangential pressure at the interface, as shown in the inset of Fig. 4.3 (see also Fig. A.1 for further details). This large tangential pressure is responsible for the negative value of the interfacial tension. It arises not only from the swim pressure of the gas (Fig. A.1), as shown in previous work\( \text{[Bialké et al. 2015], [Lee 2017], [Solon et al. 2018]} \) but also from continuous tangential self-shearing motions of particles at the surface of the dense phase. In other words, particles moving tangentially in the interfacial region both inside and outside the dense phase contribute the negative sign of the tension, as explicitly demonstrated below.

Interestingly, we also find that the magnitude of the surface tension increases with persistence length, meaning that in these systems stiffer, sharper interfaces correspond to more negative values of tension. Classically, a negative interfacial tension would indicate an inter-
face that prefers to grow; here the opposite occurs. Refs. Bialké et al. 2015, Speck 2016 conjectured a “housekeeping work” $w_{hk}$ that accounts for the work done by the particle as opposed to the work done to the particle $w_{ex}$, resulting in a relationship between tension and stiffness of the form $\gamma = -\sigma F^* \ell_p$. Earlier we found that $\sigma$ is linear in $\ell_p$, implying a quadratic dependence of $\gamma$ on $\ell_p$ (or on $\sigma$). However, the fact that $\sigma(\ell_p)$ has a non-zero axis intercept (Fig. 4.2) makes it very difficult to verify this relation quantitatively. Indeed, while our results show qualitative agreement with the idea that $F^* \ell_p$ sets an energy scale relevant to determining interfacial stability, with our error bars $\gamma$ seems to be adequately represented by a linear dependence on $\sigma$ (Fig. 4.3).

4.4 Measurements in the local frame

It is well known that even in an ideal gas of ABPs the geometry of any confining wall has a strong influence on both the structure and dynamics of the system. Fily et al. 2014a, 2017a In MIPS the gas particles interact with an emergent, self-generated boundary that continuously absorbs and releases particles, with zero net flux at steady state, but substantial local tangential currents. It is therefore illuminating to probe the interfacial structure and dynamics with respect to a local frame at each point of the interface. As described in more detail in Appendix A, we use a contour-finding algorithm to move beyond the height-map representation $h(y)$ to the position of the interface with respect to the contour length along the interface itself, $h(s)$. To do so, we define a local frame whose origin is set at points along the contour and whose orientation is set by the local normal $n$ (which is also calculated using this contour by fitting a region around a given point to a quadratic function and evaluating its curvature). We then compute the average value of the pressure tensor in slices of fixed width at each point along the interface. Although our results are quantitatively sensitive to the choice of slice width and curvature coarse-graining scale, we have confirmed that our qualitative results are robust to any sensible choice of these parameters.
Figure 4.4: We use the local frame to illuminate the complex dynamics near the interface. The top figure shows the anisotropy of current densities. The $x$-axis shows the distance from the interface measured along the normal, as shown in Fig. 4.1. Near the interface, the average swim current density field shows an excess of inward-pointing swim force current inside the cluster and an excess of transverse swim current just inside the gas. The anisotropy of the resulting motion shows that the gas moves with its swim current, but the cluster self-shears as its surface particles move tangentially. The bottom figure shows the square tangential current density $[I_t]^2$ of gas and cluster particles, as well as their sum. Tangential currents are largest in the dense phase. Results shown for $\ell_p = 100$.

4.4.1 Dynamics along the interface

We first illustrate our findings by looking at the structure and dynamics of the particles in this local frame of measurement. The stability of MIPS interfaces has often been heuristically explained by particles pointing “inward” at the outer edge of the dense cluster, with a rotational re-orientation time for these particles that is longer than the typical time for a gas particle to arrive at the interface. The condition of net zero flux then suggests large transverse currents in the gas phase outside the dense cluster. In Fig. 4.4 we can see this dynamics at play in the local frame of the interface. The square points show the average anisotropy of the swim current density, $(I_n^s)^2 - (I_t^s)^2$, and show that indeed particles in the dense cluster but near the interface preferentially point inwards, while particles in the gas phase preferentially point tangent to the local interface. An examination of the current density field itself, $I_n^2 - I_t^2$, reveals, however, an unexpected behavior. In the gas phase the current and the swim current point in the same direction since
interactions are negligible, but these two quantities are distinctly different for particles at the interface within the dense phase. One clearly sees that there is a local transverse current even in the dense phase, as the projection of the swim force in the tangential direction causes the clustered particles to slide along the very boundary they are defining.

The transverse swim current is also enhanced by persistence, which we have shown corresponds to stiffer interfaces. Additional details are given in Section 4.4.3 where we examine the behavior in the interfacial layers both within the dense and the gas phases. One observes tangential particle motions in both regions and an associated local stiffening of the interface, with the fluctuating boundary providing a local guiding effect on active gas particles similar to that observed for curved solid walls. Fily et al. [2017a].
4.4.2 Interfacial curvature and local surface tension

The findings above, showing correlated flows in both the dense and dilute phases on either side of the interface, qualitatively suggest a Marangoni-like local mass transport that stabilizes the interface. Inspired by this, we explicitly investigate the connection between the curvature of $h(s)$ and the local mechanical measurement of surface tension.

In the local frame, we still find on average a negative value for the surface tension. In this frame, though, we find that the spatial profiles of $p_n - p_t$ are much sharper, and individual slices can be binned according to the local value of curvature. These measurements have enormous fluctuations (with a variance at least an order of magnitude larger than the mean), but careful averaging allows us to distinguish a clear correlation between local curvature and local tension, which we show in Fig. 4.5 (restricting ourselves to values of the local curvature within one standard deviation of the mean to ensure sufficient statistics). Although the mean surface tension is negative for all values of curvature, we find that the outward mountains of the interface (i.e., regions of negative curvature) are quantitatively more unstable than the inward valleys. This gradient of stability provides a mechanism for the interface to dynamically stabilize itself as transverse currents from the mountains on average fill in the gaps in the valleys.

4.4.3 Dynamics near the interface

Previous work has focused on measuring the direction of the particles’ propulsive force (polarization), identifying the excess of particles with inward polarization just inside the dense cluster as the main stabilizing mechanism. In this chapter, we have closely examined the local dynamics in the interfacial region both within the dense and the gas phases. While we do observe an excess of particles with inward-pointing polarization at the surface of the dense phase (see Fig. 4.6), the most intriguing observation is of sustained local tangential motion of particles in both regions. This dynamics is displayed in more detail in Fig. 4.6, where we show that the anisotropy of current extends into each phase within a surface layer.
Figure 4.6: The left frame shows the normal swim current density $I_n$ measured in the local frame and averaged along the interface. The solid (open) symbols correspond to particles in the dense (gas) phase. The data show that there is an excess of inwardly polarized particles in a surface layer of thickness $\xi_s$ in the dense phase, but not in the gas. The length $\xi_s$ has been extracted with an exponential fit to the decay of $I_n$ and is shown in the inset as a function of $\ell_p$. It is found to decay slightly with increasing persistence. The net tangential component of swim current density $I_t$ (not shown) remains zero at the interface because particles travel without preference in either direction tangent to the interface. Right shows the normal current density $I_n$ measured in the local frame and averaged along the interface. Again, solid (open) symbols correspond to particles in the dense (gas) phase. The positive, outward-moving average current just inside the dense phase is balanced by the negative inward-moving current just inside the gas phase. Within each phase, the normal average current is finite within a surface layer of thickness $\xi_d$ (dense) and $\xi_g$ (gas) and decays exponentially. The solid lines are fits to the exponential decay. The top right inset shows $\xi_{d,g}$ extracted from those fits as functions of $\ell_p$. The solid lines are fits that show the linear growth of $\xi_{d,g}$ with persistence. The bottom left inset shows that the signed tangential current density $I_t$ is zero at the interface because particles travel without preference in either direction tangent to the interface. Here we see the fluctuations in the tangential current are much larger inside the cluster where tangential are most dominante (compare to Fig. 4.4).

Additionally, while the thickness of the region of inward-pointing swim current in the dense phase decreases slightly with increasing persistence (see inset of Fig. 4.6), the thickness of the layers with finite current increases with persistence in both the gas and the dense phases. In these regions we observe both normal currents that balance each other and tangential currents that average to zero (Fig. 4.6), but greatly exceed in magnitude the normal currents (Fig. 4.4). This suggests that the stiffening of the interface results from the combined effect of inward polarization excess at the cluster’s surface, local tangential flows in the gas, and the self-shearing of the interface in the dense cluster that heals fluctuations and enhances
stability. This observation may provide additional intuition in constructing simple models of MIPS systems, such as the Active Matter Model B+[Tjhung et al. 2018].

4.5 Discussion

In a model of purely repulsive active Brownian particles undergoing motility-induced phase separation, we have explored the surprising dichotomy of an interface that on one hand exhibits some equilibrium-like properties (a well-behaved time-averaged density profile and a nearly $q^{-2}$ spectrum of interfacial fluctuations), but on the other is governed by strong fluctuations driven by non-equilibrium physics, resulting in negative interfacial tension. It is tempting to try to write down a phenomenological model of the interface. Some authors have in fact proposed mapping the MIPS interface to an Edwards-Wilkinson (EW) growth model[Lee 2017]. On the other hand, our finding that the surface tension is itself a function of the local curvature naturally leads to additional terms, such as those in the Kardar-Parisi-Zhang (KPZ) description[Kardar et al. 1986].

Although it is clear that in the presence of a negative surface tension additional terms would be needed to stabilize the KPZ equation, we have tried to discriminate between different universality classes for the MIPS interface by measuring the critical exponents characterizing interfacial growth and steady state fluctuations. For 1d interfaces both the EW and KPZ models are characterized by the same roughening exponent, $\alpha = 1/2$, characterizing the growth of the steady-state interface width with system size $L_y$. They differ, however, in the exponent $\beta$ that controls the interface growth at short times, $w(t) \propto t^\beta$, with $\beta_{KPZ} = 1/3$ and $\beta_{EW} = 1/4$. Discriminating between the two scenarios is numerically challenging for two reasons. First, the values of $\beta$ differ only slightly in the two models. Second, as previously pointed out, there is a smallest system size necessary to observe MIPS, so achieving even a single decade in linear system size $L_y$ requires very large computational effort. Nevertheless, we have run many short simulations, starting from a flat interface, to try to characterize
Figure 4.7: The growth of the interfacial width \( w(t) \) starting from a flat configuration for several \( \ell_p \) and system sizes is shown. We present a scaling collapse according to the Edwards-Wilkinson (top) and Kardar-Parisi-Zhang (bottom) critical exponents. Here \( \alpha = \frac{1}{2}, z = \alpha/\beta, \beta_{\text{KPZ}} = \frac{1}{3}, \beta_{\text{EW}} = \frac{1}{4} \).

Surprisingly, we find that the EW dynamical exponent seem to better collapse the data than the (from a symmetry standpoint) more natural KPZ model. In particular, fits to \( w(t) = At^\beta \) for our largest systems suggest \( \beta = 0.23(3) \).

The key features of the MIPS interface seem to be a negative value of surface tension coupled with a scale-free, nearly equilibrium-like spectrum of interfacial fluctuations. The negative value of \( \gamma \) suggests that either additional terms in \( \partial_t h \) or the coupling of \( h \) to another field is required. Natural candidates, such as \( \nabla^4 h \) terms or the coupling of the interfacial dynamics to a scalar field describing the flux of particles in and out of the two phases, select either a length scale or a time scale, and neither is seen in our system (we have confirmed that in our data the power spectrum of the interface \( S(q, \omega) \) does not have any apparent time scale). The slight deviations from the equilibrium scaling of \( \langle |\delta h(q)|^2 \rangle \) may indicate that either a fundamentally nonlinear phenomenological model is required, or that simulations of much larger systems would reveal a long characteristic length scale in the problem. We view such simulations as a natural object of future study to resolve this issue.
Chapter 5

Motility-induced aggregation of

*Myxococcus xanthus*

Unicellular organisms such as bacteria and amoeba are capable of spontaneously organizing into complex multicellular structures [Laub and Loomis 1998, Dormann et al. 2002]. A striking example of such collective behavior is the starvation-induced organization of the rod-shaped, soil-dwelling bacterium *Myxococcus xanthus* into macroscopic, multicellular droplets known as “fruiting bodies” (FBs) [Zusman et al. 2007]. When nutrients are scarce, starving *M. xanthus* cells undergo a multicellular process of self-organization during which cells move to form dome-shaped droplets comprising hundreds of thousands of cells. A subset of cells at the center of each droplet differentiate to form metabolically quiescent spores that can survive long periods of starvation [Zusman et al. 2007, Starruß et al. 2012, Shimkets 1990].

The striking phenotypic similarity between FB formation in *M. xanthus* and in the amoeba *Dictyostelium discoidium* has led to the longstanding hypothesis that *Myxococcus* FB formation is driven by long-range chemical signaling mechanisms, as it is in the amoeba. Despite decades of research, questions remain on the the links between spatial chemical cues and the direct mechanisms of FB formation. Although *M. xanthus* cells are known to employ chemical communication to initiate FB formation (termed A-signaling) [Kuspa et al. 1992],
to potentially synchronize reversal frequency (termed C-signaling) Lobedanz and Søgaard-Andersen [2003], Shimkets and Rafiee [1990], and to communicate through the production of mucopolysaccharide “slime trails” that other cells can sense and follow Burchard [1982], a quantitative understanding of the mechanisms that drive aggregation has remained elusive.

*M. xanthus* cells move by gliding on solid surfaces using both tank-tread-like transport motors and the retraction of extruded filaments called pili and can modulate their speed in a continuous manner Balagam et al. [2014], Hodgkin and Kaiser [1979]. The cells also have the ability to reverse their direction of motion typically every several minutes, and can modify the reversal frequency in different situations Wu et al. [2009], Blackhart and Zusman [1985], Thutupalli et al. [2015].

In this chapter, using experiments and insight from theory, we demonstrate that *M. xanthus* FB formation can be described as a phase separation process driven, at least initially, by changes to the motility of individual cells. Importantly, this appears to happen in the absence of complex signaling mechanisms and interactions between cells, and requires no real-time control at the cellular level. While the ability to actively change motility ultimately leads to a phase transition, cells do not have to implement a complicated feedback mechanism to alter motility in response to specific chemical or mechanical cues. Rather, cells need only speed up and suppress reversals upon starvation and the collective mechanics then naturally induces phase separation of the entire population.

The theoretical inspiration for this work comes from the phenomenon of Motility-Induced Phase Separation (MIPS), where purely repulsive Active Brownian Particles (ABPs) spontaneously aggregate through a jamming-based phase transition Fily and Marchetti [2012a], Cates and Tailleur [2015], Marchetti et al. [2016c]. While there are important differences between the MIPS process of bulk phase separation and the droplet formation seen in *M. xanthus* populations, the scaling of the MIPS aggregation dynamics for reversing ABPs with a rotational Péclet number provides a powerful way to understand *M. xanthus* aggregation data.
Figure 5.1: Aggregation in *M. xanthus*. (A) Low cell densities result in phase separation via a nucleation and growth process. (B) Larger densities result in the formation of droplets everywhere at the same time via spinodal decomposition. (C) Radial component of the Fourier transform of images from *M. xanthus* phase separation at high density. Solid lines are fits to a Gaussian function added to a decaying exponential for time \( t = 1, 2, 3, 5, \) and 10 h after starvation. (D) Growth of the dominant length scale with time for spinodal decomposition experiments and ABP simulations. Time is written in units of the reversal time \( \tau_r \approx 10 \text{ min} \). The length scale of droplets coarsens as a power law in time with an exponent of \( \alpha_{\text{experiment}} = 0.30 \pm 0.02 \). Results from a simulation of reversing ABPs is shown in red, which result in an exponent of \( \alpha_{\text{simulation}} = 0.281 \pm 0.002 \).

We first investigated the dynamics of FB formation using different cell densities (experimental details in the SM). Time-lapse, bright-field images are used to quantify the resultant dynamics (Fig. 5.1). Pixel intensity is indicative of local population density (height) with darker regions corresponding to the FBs and the lighter pixels corresponding to the low density regions of bacteria. When the cell density is very low (5×10^8 cells/ml), no large-scale structure formation is seen. Over the first few hours, cells largely move independently, reversing frequently and with minimal cell-cell contacts and interactions. This results in the
formation of spatially stable nematic streams at later times (∼ 6-8 hours) Thutupalli et al. [2015]. At these low densities, starving cells do not form fruiting bodies on the time-scale of our observations. As the density is increased to $1.5 \times 10^9$ cells/ml, FBs form randomly in space and time (Fig. 5.1A). In a field of view of 3 mm by 2.5 mm, approximately 10 FB droplets were observed after 24 hours, although in some cases as few as 2 droplets formed. This spatio-temporally random appearance of FBs is similar to a phase separation process via nucleation and growth in which an energy barrier between two phases causes small fluctuations in the population density (height) to die out. The later stages of coarsening involve significant flux between neighboring FBs, seen directly in some experiments where the cell movement is evident and in others where small droplets are observed to dissolve into nearby larger ones. This is reminiscent of an Ostwald ripening-like process and indeed recent work used a model of Ostwald ripening to predict the disappearance and persistence of FBs Bahar et al. [2014].

When the density is further increased to $1 \times 10^{10}$ cells/ml and above, we observed that FBs formed via a different dynamical mechanism (Fig. 5.1B). Rather than spatially random nucleation and slow growth, high-density cultures spontaneously and immediately begin to phase separate over the entire field of view. Within the first 6 hours after plating, we observed the formation of a global instability in the cell density that resulted in small, mesh-like structures that cover the plate. This kind of spontaneous phase separation, similar to spinodal decomposition, classically arises when microscopic fluctuations in the local density are inherently unstable, lacking an energy barrier to separate the homogeneous and more favorable phase-separated regimes. As the mesh coarsened over time, small droplets appeared that were connected by thinner layers of cells. Finally, a subset of these droplets grew and turned into round FBs.

A hallmark of spinodal decomposition is a well-defined length scale of the phase-separated domains that increases with time as a power-law as the domains grow in a self-similarity manner Bray [2002], Chaikin and Lubensky [2000]. We found that at high inoculation
densities, a single dominant length scale emerges in the organization of the bacterial domains (Fig. 5.1C) and grows in time as a power law with an exponent $\alpha = 0.30 \pm 0.02$ (Fig. 5.1D).

We next sought to define the phase diagram of *M. xanthus* FB formation by varying both the overall density of cells and their motility. Active agents are often modeled as self-propelled particles with propulsion speed $v_0$ and direction of motion randomized by rotational noise at rate $D_r$. The nature of the dynamics and fluctuations is then quantified in terms of the inverse rotational Péclet number given by the ratio of the cell size $\ell_c$ to the persistence length of the motility paths $\ell_p = v_0/D_r$ [Patch et al. 2017a, Redner et al. 2013b, Stenhammar et al. 2013a, Bialké et al. 2015],

$$\text{Pe}_r^{-1} = \ell_c/\ell_p = \ell_c D_r/v_0 . \quad (5.1)$$

As *M. xanthus* cells can spontaneously reverse their direction of motion at rate $f_{\text{rev}}$, we sought to understand how these reversal events might change the Péclet number. Introducing a reversal frequency adds a new timescale, the effects of which can be incorporated in an
effective rotational diffusion coefficient (see SM and Grossman et al. [2016])

\[ D_{\text{eff}} = D_r + 2f_{\text{rev}} . \] (5.2)

Adding reversals as a Poisson process with mean frequency \( f_{\text{rev}} \) in a model of ABPs Fily and Marchetti [2012a], Marchetti et al. [2016c], Tailleur and Cates [2008], we indeed found (Fig. 5.2) that the single-particle MSDs for different reversal frequencies collapse when time is scaled by the reorientation time \( \tau^{-1} = D_{\text{eff}} \). Moreover, the crossover between ballistic and diffusive motion occurs at \( \tau^{-1} \) for all \( f_{\text{rev}} \) at both low and high particle density. This shows that \( D_{\text{eff}} \) is the appropriate parameter to use when calculating the inverse Péclet number for reversing cells. Additionally, we find that the exponent characterizing the temporal growth of the length scale of MIPS spinodal decomposition, \( \alpha = 0.281 \pm 0.002 \) (Fig. 5.1D), is close to our measurement for \( M. xanthus \) cellular droplet formation and in agreement with previous results from non-reversing ABPs Stenhammar et al. [2013a], Redner et al. [2013b], Patch et al. [2017a].

To control the Pe\(^{-1}\) of \( M. xanthus \) cells experimentally, we took advantage of the non-reversing mutant \( \Delta FrzE \), which does not change its velocity over time even in starvation conditions (Fig. 5.4). We then varied Pe\(^{-1}\) by altering the propulsion speed \( v_0 \) using the drug nigericin Sun et al. [2011]. The inverse Péclet number for each experiment was measured in separate tracking experiments to probe \( D_r \) and cell speed for each nigericin concentration. We mixed a small number of fluorescently-labeled cells with non-fluorescent cells at a ratio of 1:400 and tracked only the fluorescent cells to measure the speed. We estimated \( D_r \) by tracking the motion of cells at very low density such that cells do not physically interact with each other and move purely in two-dimensions on the substrate. The decay of the temporal velocity autocorrelation function yields \( D_r = 0.04 \pm 0.02 \text{ min}^{-1} \) (Fig. 5.5A. We note that the typical reversal frequency of wild-type (WT) \( M. xanthus \), \( f_{\text{rev}} \sim 0.05 - 0.17 \text{ min}^{-1} \), is

---

\(^1\)While \( \Delta FrzE \) cells have been reported to reverse at a very low frequency, we did not observe any reversals using this strain in our analysis.
Figure 5.3: (A) Experimental phase diagram for *M. xanthus* phase separation. For each experiment, we determined whether the system is phase separated (red squares) or homogeneous (black circles). The phase boundary is drawn by hand as a guide to the eye. Dashed horizontal lines represent $Pe^{-1}$ for wild-type cells before (black) and after (red) starvation. (B) The phase diagram for reversing ABPs showing the spinodal boundary, obtained as described in the SM. The spinodal points correspond to the peaks of a bimodal distribution of local density in systems with different values of $f_{rev}$ (different symbols) and different mean packing fraction (different colors). The dashed line is a guide to the eye. The horizontal axis is the particle packing fraction $\phi$.

Of the same order of magnitude but slightly larger than the rotational diffusion coefficient. This implies that subtle changes in $f_{rev}$ can have appreciable affects on $Pe^{-1}$. We combine the measured speed and $D_r$ with the average cell size of $\ell_c = 2.5 \mu m$ (half a cell length) to calculate the Péclet number for each condition.

The experimentally derived phase diagram of *M. xanthus* phase separation is shown in Fig. 5.3A. As predicted, at low density or high $Pe^{-1}$, the system does not form droplets (black circles). At high density or low $Pe^{-1}$, the system phase separates via spinodal decomposition (red squares). The estimated spinodal line is denoted on the phase diagram. In Fig. 5.3B, we show the phase diagram obtained from simulations of the model of reversing ABPs described in the SM. The scaling of the data for different values of $f_{rev}$ and mean density confirms that the effect of reversals can be incorporated entirely in a $Pe^{-1}$ defined with the effective
Figure 5.4: Cell tracking over time after starvation. (A) Speed, (B) reversal frequency, and (C) the resulting $Pe_r^{-1}$ are shown for wild-type (blue) and \( \Delta \text{FrzE} \) cells (red).

rotational diffusion $D_r^{\text{eff}}$.

$Pe_r^{-1}$ depends on four parameters, two of which \textit{M. xanthus} potentially has the ability to control during FB formation. Cells do not grow during aggregation due to the starvation conditions and $D_r$ is presumably set by thermal fluctuations of the cell body and molecular noise in the motility process. However, both the cell speed $v_0$ and the reversal frequency $f_{\text{rev}}$ are known to be under cellular control, suggesting that the cells might have adapted to take advantage of such control.

We tracked individual fluorescent wild-type cells at a population density of $1 \times 10^9$ cell/ml for 11 hours after starvation and found cells changed both their gliding speed and reversal frequency (Fig. 5.4). In the first 3-4 hours, cells exhibited low gliding speed, $\approx 1.5\mu\text{m/min}$, with fewer than 10 percent of the cells actively moving, in agreement with previous reports of an initial “resting” phase upon starvation \textcolor{green}{Jelsbak and Sogaard-Andersen 2002}. After being starved for over 5 hours, \textit{M. xanthus} cells sped up to $\approx 2.5\mu\text{m/min}$ (Fig. 5.4A), almost
doubling their initial speed. At these times, over 90 percent of the cells were actively moving (Fig. 5.5). \(^2\) We also found that reversal frequency decreased significantly from 0.128 min\(^{-1}\) to 0.055 min\(^{-1}\) over the 11-hour experiment (Fig. 5.4B). A combination of increased \(v_0\) and decreased \(f_{rev}\) produces a reduction in \(Pe_{r}^{-1}\) from 0.60 to 0.16 (Fig. 5.4C). This reduction in \(Pe_{r}^{-1}\) is sufficient to drive phase separation and FB formation. Before starvation, cells move very slowly and reverse frequently, favoring a homogeneous population on the surface. Upon starvation, wild-type cells speed up and reverse less often, producing a situation favorable for phase separation and FB formation (Fig. 5.3).

Many of the details that we purposely left out of our analysis most likely play a role in the specific evolution and shape of the final fruiting bodies. These include cell-cell alignment, the effects of “slime following,” and cell-cell communication via the C- and A-signaling mechanisms. For example, ΔFrzE cells do not form stable fruiting bodies. While initially stable, droplets typically fall apart at the end of 24 hours, towards the end of traditional FB development. This potentially indicates that additional biological or chemical mechanisms may play a role in FB stability over long times. More complicated models of \(M. xanthus\) aggregation may uncover the role of these additional parameters \[see e.g. Ref. Cotter et al., 2017\], but it is unlikely that they will change the basic features we have observed here.

Future work tracking cells and monitoring droplet shape in three dimensions should lead to a more accurate theory of the phase separation which might be viewed as the dewetting of an active bacterial fluid layer into 3D droplets, which eventually form the FBs.

\(\Delta FrzE\) cells do not change speed when starving, indicating a link between the Frz pathway and gliding speed.
5.1 Methods

5.1.1 *Myxococcus xanthus* culture and development conditions

Liquid cultures of wild-type *M. xanthus* strain DK1622 and ∆FrzE were grown at 32°C in agitating CTTYE medium (1.0% Casitone, 0.5% yeast extract, 10.0 mM Tris-HCl at pH 8.0, 1.0 mM $KH_2PO_4$, and 8.0 mM $MgSO_4$). Kanamycin (40 µg/ml) was added only to liquid cultures of ∆FrzE. Starvation assays were performed using non-nutritive Tris phosphate medium (TPM) agarose (10.0 mM Tris-HCl at pH 7.6, 1.0 mM $KH_2PO_4$, 8.0 mM $MgSO_4$, and 1.5% agarose). To induce development, growing cells were harvested from liquid culture at mid-log phase and resuspended to a final concentration of various densities in TPM: $5 \times 10^8$, $1.5 \times 10^9$, $2.5 \times 10^9$, $5 \times 10^9$, $2.5 \times 10^{10}$ cells/ml. 10 µl spots were plated on a TPM agarose slide complex and allowed to dry as described previously [Bahar et al. 2014]. To modulate velocity, cells suspensions and TPM agarose was supplemented with nigericin sodium salt at concentrations of 0, 1, 2, 4, and 10 µM which changed the speed by a factor of about 4 (Fig. 5.5B).

5.1.2 Imaging and tracking

Cells were imaged at 20× and 100× magnification to record the behavior of both single cells and aggregates. For 100× magnification, cells were imaged on a modified Nikon TE2000 inverted microscope with an oil-immersion objective (NA 1.49). To capture an enlarged 110×110µm field of view, we used a tiling strategy and imaged a 3×3 grid of 100× fields. Details of this imaging and auto-focusing strategy were reported previously [Thutupalli et al. 2015]. Images were recorded at a rate of one frame per 10 seconds. Cell tracking using bright field images was performed using our previously published BCTracker algorithm [Thutupalli et al.] 2015).

For high cell density fluorescence cell tracking, a 1:400 mixture of Alexa Fluor 594 carboxylic acid succinimidyl ester labeled DK1622 or ∆FrzE cells to non-labeled cells was used.
to record the behavior of individual cells in large groups. To stain cells, cells were grown to mid-log phase, harvested by centrifugation and resuspended in MC7 buffer. 2 µl of dye (10 mg/ml, dissolved in DMSO) and 5µl of 1M NaHCO$_2$ was added to 100 µl of cells and shaken vigorously at 100 RPM for 1 hour in the dark at room temperature. Cells were then pelleted by centrifugation, washed 3 times in TPM and microscopically examined. Fluorescent microscopy images were taken at a rate of one frame per min for the first 15 min in each hour to minimize the amount of laser exposure for cells. Experiments lasted 11 hours. Fluorescent cells were tracked using a particle tracking algorithm developed by Crocker, Grier and Weeks [Crocker and Grier 1996]. In our analysis, a cell is counted as actively moving if during each tracked hour it glides with a mean speed greater than 0.5µm/min (Fig. 5.6). A reversal event is defined as occurring when the velocity vector between two successive time points changes sign.

To calculate the rotational diffusion constant, we tracked 50 ΔFrtzE cells and observed their motion for at least 35µm. The diffusion constant was then calculated from the decay of the velocity temporal autocorrelation function and assumed to be the same for all experiments (Fig. 5.5A). To measure the length-scale growth displayed in Fig. 5.1C,D, we imaged WT M. xanthus (1 × 10$^{11}$cells/ml) on a 20× magnification home-built bright field microscope at a rate of one frame every 10 seconds for 24 hours.

At higher densities, it is experimentally difficult to break up cell clumps that have formed in the liquid culture. Thus, at the beginning of a movie we sometimes see isolated aggregates that are not fruiting bodies and which very quickly dissolve as cells migrate out of them. We start our analysis from the point where the initial visible aggregates have dissolved.

### 5.1.3 Simulation details and parameters

Each reversing ABP is modeled as a disk of radius $a$, with dynamics governed by overdamped Langevin equations of motion (Fig. 5.7), which is a slightly modified version those introduced
Figure 5.5: (A) Top: Examples of ∆FrzE cell tracking results. Bottom: Velocity temporal autocorrelation function. An exponential fit to the data yields a rotational diffusion coefficient $D_r = 0.04 \pm 0.02 \, \text{min}^{-1}$. (B) Tracking of ∆FrzE cells under various Nigericin concentrations. Cell velocity is averaged over $\approx 50$ tracks each.

in section 2.2.2

\[
\begin{align*}
\dot{r}_i &= v_0 \kappa_i(t) \hat{n}_i + \mu \sum_j F_{ij}, \\
\dot{\theta}_i &= \sqrt{2D_r} \eta_i(t), 
\end{align*}
\]

where we incorporate directional reversals through a function $\kappa_i(t)$, which takes the values $\pm 1$, changing sign at times given by a Poisson process with a mean reversal frequency $f_{rev}$. The force $F_{ij}$ is purely repulsive harmonic force, as introduced in section 2.2.2.

We simulated Equation (5.3) using a standard Brownian Dynamics algorithm in an $L \times L$ box with periodic boundary conditions. In all cases, we use $k = \mu = 1$ so the interaction
Figure 5.6: Active cell percentage versus time derived from fluorescent cell tracking of both WT and ΔFrzE cells. Note that ΔFrzE cells are active even in the early starvation stage, while WT cells undergo a “resting” phase in the first 5 hours.

The timescale \( \tau_D = (\mu k)^{-1} \) sets the unit time and we use the particle radius \( a \) as the unit of length (\( a = 1 \)). To prevent particles from passing through each other, we set \( v_0 = (a\mu k)/100 \). We fix the packing fraction \( \phi = N\pi a^2/L^2 \), which sets the total number of particles, \( N \). The rotational diffusion \( D_r \) and the reversal frequency \( f_{\text{rev}} \) are varied to obtain the desired \( \text{Pe}_r^{-1} \).

For each set of parameters, we average over 10–100 runs and use a jackknife method \cite{Amit and Martin-Mayor, 2005, Patch et al., 2017a} to estimate statistical errors. In order to compute the length scale \( L(t) \) and its coarsening exponent (Fig. 5.1D) we used a large system size with \( L = 1000 \), \( \text{Pe}_r^{-1} = 0.01 \) and a packing fraction of \( \phi = 0.5 \) (\( N = 159,154 \) particles), averaging over 100 independent runs. For other quantities we did not need such a large system size. The phase diagram was computed on systems with \( L = 200 \) (10 runs for each set of parameters).

### 5.1.4 Measuring \( L(t) \)

In experiments, local density is estimated as the intensity of measured light level at each pixel, where density has an inverse relationship with light intensity (darker is more dense). Once the experimental \( S(k) \) distribution has been obtained, it is fit with an exponential
Figure 5.7: A single particle has radius $a$ and moves with velocity $v_0 \hat{n}_i$. The direction of self-propulsion is continuously affected by a white noise with variance proportional to $D_r$ and directional reversals at times given by a Poisson process with frequency $f_{\text{rev}}$. These two parameters can be combined into an effective rotational diffusion coefficient: $D_r^{\text{eff}} = D_r + 2f_{\text{rev}}$. In addition, there is a spring-like repulsive interaction force $F_{ij}$ between each pair of particles. High-density clusters nucleate when colliding particles are caged by their surrounding neighbors before being able to reorient.

In simulations, we discretize the system by dividing it into bins with width equal to the particle radius $a$ and assigning a binary 1 or 0 value to each bin depending on whether or not a particle is centered in it. In order to reduce noise, the resulting density distribution is averaged over exponentially increasing temporal bins [Patch et al. 2017a]. The FFT is then computed in 2D to produce the structure factor $S(k,t)$ (Fig. 5.8). The average length scale was calculated from the first moment of $S(k,t)$

$$L(t) = \frac{\int_{2\pi/N_{\text{FFT}}}^{k_{\text{max}}} S(k,t) \, dk}{\int_{2\pi/N_{\text{FFT}}}^{k_{\text{max}}} kS(k,t) \, dk},$$

(5.4)
where \( k_{\text{max}} \) is chosen to exclude noisy, high-frequency modes and sample the mean of the lowest-wavenumber peak in \( S(k) \). We use a jackknife method\cite{Amit2005, Patch2017} to estimate errors.

### 5.1.5 Distinguishing phase behavior

To quantitatively distinguish between nucleation and growth phenomena and spinodal decomposition, we take advantage of the temporarily spontaneous nature of the spinodal decomposition phase transition. We measured the size of aggregates over time (Fig. 5.9A) and then calculated the distribution of times at which aggregates reach one half of their final size. The variance in this distribution, calculated per experiment, is bimodal with a clear lower peak at \( \text{Var}(t) < 1 \text{h} \) (Fig. 5.9B), corresponding to experiments in which spinodal de-
Figure 5.9: (A) Examples of WT FB size evolution at different densities. The top two conditions stimulate nucleation and growth as fruiting bodies form at varying times. The bottom two conditions stimulate spinodal decomposition where the emergence of fruiting bodies is synchronized. (B) Histogram of the variance in the time when fruit bodies reach half-maximal size. When this time variance is smaller than 1h, we conclude that the system is undergoing spinodal decomposition. (C) The fraction of clusters observed over time is shown for 100 simulations at varying packing fractions ($Pe_r^{-1} = 0.01$). The resulting deviation of the nucleation times shows how as density is decreasing from just inside the coexistence region, significantly longer mean times and wider variances in times of nucleation occur. This provides a clear distinction between spinodal decomposition and nucleation and growth, allowing for a quantitative threshold between the two.
composition occurs, and higher-value peaks that correspond to nucleation and growth where aggregates form randomly over time.

We performed a similar calculation using the simulations. Since we are limited by the simulation size, in many cases we only get one cluster at the end. To probe if a system undergoes a spontaneous transition for each set of parameters, we performed 100 simulations and compared the cluster growth across simulations (Fig. 5.9C). For sets of parameters that lead to spinodal decomposition, the distribution of aggregation times has a clear peak, while for nucleation and growth a large tail develops (with an eventually diverging average as we approach the phase boundary).
5.1.6 Producing the simulation phase diagram

Here, we follow a simple technique for mapping the phase space of ABPs [Redner et al. 2013a] by measuring the distribution of local densities in our simulations. In this technique, a unimodal distribution signifies a homogeneous system while a bimodal distribution signifies phase-separated state. Examples of these are shown in Fig. 5.10. We sample local density using square windows of width $L_W = 20a$ (the total size of the system is $L = 200a$). For high density circle-packing, which is at a density above those we generally measure in the dense phase, a window would contain $\approx 115$ particles. We use this to set the density bin width of length $d\phi \approx 0.01$.

Drawing from the last half (steady state) of each run, we average histograms over 10 runs. We determine dominant phase densities by selecting the peaks from each distribution. These densities are then used as coordinates in our phase diagram, paired with the configuration $P_{e_t}$.

5.2 Effective rotational diffusion in the presence of directional reversals

We consider the overdamped dynamics of a single self-propelled particle with directional reversals in two dimensions. The directional unit vector $\hat{n} = (\cos \theta, \sin \theta)$ is set by the angular direction $\theta \in [0, 2\pi)$. This evolves in time as

$$d\theta = \sqrt{2D_r}dW_t, \quad (5.5)$$

where $D_r$ is rotational diffusion and $W_t$ is a Wiener process. Particle direction periodically reverses as a Poisson process where the waiting time distribution between two reversals is exponential. The overdamped equation of motion is expressed as a stochastic process driven

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[^3]: Thanks to Suraj Shankar for calculating the MSD for ABPs with reversals, revealing this very nice and useful result.
by dichotomous Markov noise (DMN)\textsuperscript{Sancho 1984},

\[ \text{d}x(t) = v_0 \text{\textbf{n}}(t) \text{d}\xi_t, \]

(5.6)

where \( \xi_t \in \{-1, 1\} \) is the symmetric DMN with zero mean and exponential correlation \( \mathbb{E}[\xi_t, \xi_{t'}] = \exp(-2f_{\text{rev}}|t-t'|) \), and \( f_{\text{rev}} \) is the mean reversal rate. Particle position \((x(t), y(t))\) evolves as

\[
\begin{align*}
    x(t) &= x_0 + v_0 \int_0^t \text{d}\xi_{t'} \cos \theta(t') \\
y(t) &= y_0 + v_0 \int_0^t \text{d}\xi_{t'} \sin \theta(t'),
\end{align*}
\]

(5.7) (5.8)

where \((x_0, y_0)\) is the initial position. Since \( \theta(t) \) and \( \xi_t \) are independent random variables, we separately average over all realizations of the rotational noise and initial angular conditions and then average over all realizations of the DMN to calculate the mean-square displacement

\[
\langle |x(t) - x(t')|^2 \rangle = \frac{v_0^2}{2D_{\text{eff}}} \left[ t - \frac{1}{D_{\text{eff}}} (1 - e^{-D_{\text{eff}}t}) \right],
\]

(5.9)

where \( D_{\text{eff}} = D_t + 2f_{\text{rev}} \) is the effective rotational diffusion.
Chapter 6

Conclusion

The collective behavior of living systems is a rich subject of study, and active matter is a fascinating, relatively new approach to examining how motility contributes to the formation of these groups. Using minimal active matter models, we have shown that important aspects of seemingly complex organizational processes can be captured using, for instance, by the interplay of motility and crowding. From the unique self-driven nature of active particles, a rich set of open problems has emerged in this field. These have been studied in a combination of analytic, numerical, and experimental work to build a quantitative understanding of self-assembly and emergent structures and patterns.

In this dissertation, I begin in Chapter 1 by giving some general background of active matter, classifying active system in terms of their symmetry and properties and the kinds of macroscopic order and disorder that result from activity and interactions. In Chapters 1 and 2 I provide conceptual background on Brownian motion and the Fluctuation-Dissipation Theorem (FDT), both of which are of critical importance for understanding the nonequilibrium behavior of active particles. The majority of my work focuses on motility-induced phase separation (MIPS), a novel phenomenon in which purely repulsive spherical Active Brownian Particles (ABPs) can spontaneously separate into dense and dilute phases. In the second part of Chapter 2 I have therefore reviewed the main theoretical, numerical, and
experimental approaches that have been used to explore this phenomenon. The remainder of this dissertation covers the three main projects that I have worked on during my graduate studies, all with the goal of studying MIPS and a few of its remarkable features.

In Chapter 3, I describe numerical work aimed at quantifying the pressure of fluids of ABPs. I have examined finite-size effects in the kinetics of phase separation for both closed systems and periodic boundary conditions. We show that in ABPs pressure is generically a nonmonotonic function of density and connect this nonmonotonicity to MIPS. We also show correlations in the time evolution of the mean pressure towards its steady state value and the kinetics of MIPS through the formation and growth of clusters that grow to the system size. For parameter values corresponding to phase-separated steady states, we identified two dynamical regimes: rapid cluster formation, followed by relaxation to the steady state, something analogous to activation energy in kinetic processes. This work is published in Ref. Patch et al. [2017a].

In Chapter 4, I describe more recent work aimed at quantifying the emergent properties of the interface of coexisting motility-induced phases. Previous work shows that the mechanical tension measured along the interface between these phases is negative, which we reproduce generically. While this would mean that the interface is not stable and must break up, we find that these out-of-equilibrium systems display long-time stability and have intrinsically stiff boundaries by generating and utilizing local pressures anisotropies that direct flow along the interface using its fluctuating curvature. We examine this phenomenon in detail using active Brownian particle simulations and a novel local frame of reference that is located and oriented by the algorithmically located interface and oriented along it. The flow along the interface appears to be an out-of-equilibrium, Marangoni-like effect, drawing even deeper connection to equilibrium analogs. Furthermore, we discuss the implications of our observations on phenomenological models of interfacial dynamics, indicating the difficulty in modeling surface tension at the continuum level. This work is can be found in pre print form in Ref. Patch et al. [2018].
Chapter 5 discusses our collaborative work with the experimental groups of Joshua Schae-vitz (Princeton University) and Roy D. Welch (Syracuse University), which aims to at understand self-organization in *M. xanthus*, which we model as a modified ABP whose single-cell parameters are measured experimentally for direct comparison. By combining high-resolution single cell tracking experiments with numerical simulations, we show that starvation-induced fruiting body (FB) formation in *Myxococcus xanthus* is a phase separation driven at the level of individual cells that tune their motility over time. Thus, *M. xanthus* aggregation data can be organized in a phase diagram in terms of cell density and a dimensionless Péclet number that combines the cell its speed and its reversal frequency. This work suggests that *M. xanthus* evolved to take advantage of a self-driven non-equilibrium phase transition that can be controlled at the single cell level, which is a remarkable thing to observe quantitatively. This work is presented in pre-print form in Ref. Liu et al. [2017].

In general, I find the study of soft matter and active matter to be a wonderful opportunity to think about emergent behavior that may otherwise be assumed to be orchestrated by leaders and hierarchy that is perhaps magical or divine or of some other immaterial origin. In many examples of emergent behavior, leaderless groups self-organize, which is itself really quite fascinating. For this reason, while the study of collective behavior holds great potential for applications in fields like biology, medicine, and biomedical engineering, where it is currently focused, it also has implications for areas such as graphic design [1] finance [Sornette 2009], and philosophy [de Landa 2015]. Of even more basic interest to me, soft and active matter represents a great opportunity for both future research and also a more inclusive style of physics education because it is so oriented toward collaboration and several captivating experiments can be done on a table top or viewed under a standard lab microscope. Examples of this from soft matter are abundant, and some highlights include the beautiful granular experiments conducted in Bob Behringer’s lab [Jaeger et al. 1996] and 1The Vicsek model was actually developed following the insight of computer scientist Craig Reynolds who created it in 1987 to model dynamic bird flocks and herds [Reynolds 1987]. His active particles, called “boids”, have been used in popular movies like Batman Returns (1992) and The Lion King (1994).
the grueling studies of paper crumpling by Shmuel Rubinstein [Rubinstein 2016], along with other more classic systems that are less commonly known, like liquid crystals [Lalanne and Hare 1976], which are widely used in liquid crystal displays (LCDs). In active matter, the vibrated granular rods Sriram Ramaswamy [Kumar et al. 2014] are fascinating, and many other table-top active agents exist like the autophoretic colloids [Howse et al. 2007], [Palacci et al. 2010], [Theurkauff et al. 2012], rollers [Bricard et al. 2013], and droplets [Thutupalli et al. 2011] previously mentioned. Additionally, there is a great opportunity to engage with biological subjects like *M. xanthus* \(^2\) or *Escherichia coli*. Moreover, examples of collective behavior, like flocks, schools, and herds, are already objects of fascination in the eyes of the general public and hold a great potential for a subject of interdisciplinary education. Doing outreach sharing my own research has proven fun and engaging even for students who are not interested in science and have not even taken a high school physics course yet. Soft active matter is a unique and fascinating field that diverges from the current physics paradigm that says we must focus on the very large or very small phenomena of our universe—which are all, of course, of great interest as well!

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\(^2\) *M. xanthus* fruiting body formation has been documented extensively with a set of microscopes they ingeniously 3D printed and assembled on site in the Welch Lab at Syracuse University.
Appendices
Appendix A

Locating the interface

We have utilized two techniques to locate the interface and quantify its properties. The first uses a global frame of reference with axes normal and tangential to the mean (temporally and spatially averaged) interface ($x$ and $y$) and yields a height map projected onto the $y$-axis, $h(y)$. The second traces an outline of the boundary of the strip yielding a parameterized curve $h(s)$ that captures fluctuations and overhangs. In both cases, for a given snapshot we quantify the configuration and position of left and right interface, denoted by $h_{L,R}$.

A.1 Height map $h(y)$

To calculate the height map we follow a straightforward procedure:

1. Distribute the cluster particles in $n_{\text{bin}}$ bins according to their $y$-position.

2. Sort particles in each bin according to their $x$-distance from the cluster center.

3. Average this $x$-distance of the left-most or right-most $n_{\text{avg}}$ particles. The resulting quantity is identified with $h_{L/R}(y)$. In general we use $n_{\text{avg}} = 3$.

The width $d_y = L_y/n_{\text{bins}}$ of the bins is chosen as $d_y = 2r_0$. We have also verified that slightly larger bins do not significantly change our measurements.
A.2 Interface contour $\tilde{h}(s)$

We have also quantified the interface not as a height map but as a curve parameterized by its arc length. To do so, we have implemented a classic contour-finding algorithm with square pixel resolution $l$ at the size of particles, $l = 2r_0$. Schematically, the algorithm proceeds as follows:

1. Discretize the system into square pixels of size $l$.

2. Mark all pixels containing cluster particles, call this set $G$.

3. Dilate $G$ by creating a new set $D$ such that $G \subset D$ where all pixels adjacent to members of $G$ are marked in $D$.


5. Connect contours using a depth-first-search algorithm, collating adjacent points into subsets $c$ such that $c \in C$.

The result can be seen in Fig. 4.1. Once the sets $c$ are available, we locate the longest contours that cross the periodic boundaries an odd number of times. These are then sorted in left and right according to their $x$-distance from the strip center of mass $x_{CM}$. The height $\tilde{h}(s)$ is then defined as the distance of these points from $x_{CM}$. Note that $\tilde{h}(s)$ is a multiple-valued function on scales just wider than a single pixel.

A.3 Smoothing

While the above measure of the interface is generally useful for determining the interface length, including the more fractal inlets that exist down to the single-particle level, the noise on our measurements of local curvature is significantly reduced when we smooth the interface contours $\tilde{h}(s)$ to $h(s)$. To do this, we calculate the shortest path along $\tilde{h}(s)$ using
the Dijkstra algorithm\cite{Cormen2009} on this relatively sparse, but connected set. More specifically we

1. Choose any pixel $p \in c$ with only two neighbor pixels $p_-, p_+ \in c$.

2. Run Dijkstra on reduced contour $(c - p)$, starting at $p_+$ and ending at $p_-$.

3. Add $p$ back to the Dijkstra path to connect the set $h(s)$.

The resulting $h(s)$ is now a smooth connected path that winds the periodic box in the $y$-direction.

A.4 Determination of local normal to the interface

Using either the basic contour $\hat{h}(s)$ or the smoothed $h(s)$, we calculate a local normal $\hat{n}$ using the local tangent vectors, as defined by each pixel and two nearest neighbors. We coarse-grain these vectors by averaging the normal vectors of the $n_{cg}$ nearest neighbors contour pixels. We test the result by eye and find that a coarse-graining of $n \approx 10$ neighboring pixels works well at several $\ell_p$. The result is a set of normal vectors $n(s)$ along the interface. For consistency, we use the same number of normals for the unsmoothed and smoothed interfaces, although this number could be reduced in the smoothed case.
Figure A.1: Normal (top) and tangential (middle) pressure broken down into swim, interaction and total parts. The bottom panel shows the various contributions (total, swim and interaction) to the difference $p_n - p_t$ as a function of the distance $x$ from the dense phase center of mass, for $\ell_p = 140$. This difference is the integrand in the expression for the tension, Eq. (4.11).
Appendix B

Local frame

The local frame is defined using $h(s)$ or $\tilde{h}(s)$ and the set of normal vectors $n(s)$, where $s$ is the set of connected points defining the contour. Contours $\tilde{h}(s)$ are multiple-valued functions, so when using this contour we ignore those points that don’t have enough neighbors to properly define a normal. In the case of $h(s)$, this problem is avoided by definition.

Using $h(s)$ and $n(s)$, we define a set of bins contained in a box of width $w_c$ length $L_c$, oriented along $n(s)$ and centered at $h(s)$. The bin width $dn_c = 4r_0$ sets the spatial resolution of our local frame data. A schematic of this frame is shown in Fig. 4.1. With this binning technique, we collect information for all particles that fall inside the box. Given the local normal, it is straightforward to properly transform the various scalar, vector and tensor physical quantities measured in this work from the global ($x$-$y$) frame to the local frame.
Appendix C

Correlation between curvature and tangential currents in the local frame

Strong correlations between the local curvature and spatially-resolved tangential currents are implied by the curvature-dependent interfacial tension shown in Fig. 4.5. Due to the wild fluctuations at the interface, however, it is difficult to directly visualize these curvature-dependent flow profiles (as was done in the case of active particles near a hard boundary [Nikola et al. 2016]).

To obtain sufficient statistics, we instead compare the pressures and currents in our local frame slices (see bottom right frame of Fig. 4.1 for a pictorial definition of the slices) associated with parts of the interface beyond or within the average height of the interface. On average, if $\bar{h}$ is the average position of the interface, then slices associated with $\delta h(s) = h(s) - \bar{h} > 0$ correspond to regions of negative curvature (“mountains”) and slices associated with $\delta h(s) < 0$ on average correspond to regions of positive curvature (“valleys”), with $s$ is the position of the slice along the interface. Although indirect, this simple criterion allows for easy collection and processing of the large amounts of data needed to get a reasonable signal-to-noise ratio.

For ease of notation, in this section we introduce a local tension $\tilde{\gamma}(s)$ calculated from its
Figure C.1: (top) The difference in the integrand in the mechanical definition surface tension for local slices beyond ($\delta h(s) > 0$) or within ($\delta h(s) < 0$) the average position of the interface, as a function of distance from the interface in that slice. The integrand is less negative in the valleys than the mountains, consistent with the results in Fig. 4.5. (bottom) The difference in transverse currents for the same sets of local slices. The excess transverse currents in the “mountains” suggests a Marangoni-like effect connecting local geometry and shear flow along the interface.

We also examine the difference between the tangential current density of cluster particles and we find that there is indeed a greater transverse current in the “mountains” than in the “valleys” (Fig. -bottom). Although indirect, these two observations suggest a clear
connection between gradients in surface tension and local tangential mass transfer. A natural target of future study would be a direct investigation of the curvature - tangential current correlations.
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Education

2012–2018 PhD Physics, Syracuse University, Syracuse, NY.
2007–2009 BA Physics, Boston University, Boston, MA.
2005–2007 coursework in Media, Political Science, American University, Washington, DC.

Research and work

2014–2018 IGERT Fellow, Syracuse University, Syracuse, NY.
— IGERT training in communication, policy, ethics, and physical cell biology
— Research in statistical mechanics of active matter
— Interdisciplinary research studying the collective behavior of M. Xanthus
— Advised by Prof. Cristina Marchetti

2012–2014 Teaching Assistant, Syracuse University, Syracuse, NY.
— Ran recitation and lab in a range of introductory physics courses (see Teaching)
— Lecturer, Introductory Mechanics (Summer 2013)

2010–2012 Post-baccalaureate Researcher, Yale University, New Haven, CT.
— Contributed to a neutrino cross-section measurement
— Developer in the Liquid Argon Software (LArSoft) group
— Aided in multiple stages of MicroBooNE’s DOE Critical Decision process
— Based at Fermilab under the direction of Prof. Bonnie Fleming

2009–2010 Technician, Fermilab, Batavia, IL.
— Monitored MiniBooNE neutrino detector at Fermilab
— Automated routine detector and beamline checks
— Explored use of boosted decision trees for particle identification, led by Prof. Byron Roe.

2008–2009 Undergraduate Research, Boston University, Boston, MA.
— Studied thin-films semi- and super-conductors, and nanoscale resonators
— Learned and conducted full fabrication process for nanoscale devices
— Supervised by Prof. Pritiraj Mohanty

2007 Summer Undergraduate Research, American University, Washington, DC.
— Studied quantum entanglement
— Ran calculations in Mathematica
— Research directed by Prof. Nathan Harshman

Publications

2018 Adam Patch, Daniel Sussman, David Yllanes, and M. Cristina Marchetti Curvature-dependent tension and tangential flows at the interface of motility induced phases [under review] (2018)


2016 M. Cristina Marchetti, Yoauen Fily, Silke Henkes, Adam Patch David Yllanes. *Minimal model of active colloids highlights the role of mechanical interactions controlling the emergent behavior of active matter* COCIS (2016).


### Awards

2016 IGERT Fellowship
2016 3rd Prize, Stevenson Biomaterials Lecture Poster Session
2014 IGERT Fellowship
2013 Henry Levinstein Fellowship
2013 Syracuse University CAS Summer Fellowship
2009 Boston University UROP Student Research Award
2008 Boston University UROP Student Research Award
2005-07 American University Dean’s Scholarship

### Conference and Workshop Attendance

2018 APS March Meeting, Los Angeles, CA, Talk.
2017 APS March Meeting, New Orleans, LA, Talk.
2017 AAAS Annual Meeting, Boston, MA, Policy Workshop.
2016 Active Complex Matter School, IESC, Cargese, France, Poster.
2016 Active and Smart Matter ICAM Workshop, Syracuse University, Poster.
2016 APS March Meeting, Baltimore, MD, Talk.
2015 School on Soft Matter and Complex Fluids, UMass, Amherst, Poster.
2015 113th Statistical Mechanics Conference, Rutgers University.
2014 112th Statistical Mechanics Conference, Rutgers University.
2012 Excellence in Detector and Instrumentation Technology School, Fermilab.

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2011 Fundamental Physics at the Intensity Frontier Workshop, Rockville, MD.
2011 International Neutrino Summer School, Geneva, Switzerland.
2011 Accelerated C++: A short course in programming by example, Fermilab.
2010 Fermilab Users Conference, Fermilab.
2009 Society of Physics Students Chapter Meeting, Boston University.
2009 Undergraduate Women in Physics Conference, Yale University.
2008 Sigma Pi Sigma Quadrennial Congress, Fermilab.
2008 Society of Physics Students Chapter Meeting, University of Connecticut.

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Organizing

2017–2018 Graduate Science Policy Group, Syracuse University, President and Founder.
2017–2018 Syracuse Graduate Employees United, Syracuse University.
2017 Syracuse March For Science, Syracuse, NY, in collaboration with local activists.
2016 Active and Smart Matter ICAM Workshop, Syracuse University.
2016 Conference for Undergraduate Women in Physics, Syracuse University.

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Teaching

Spring 2014 Stars, Galaxies, and the Universe, Syracuse, Lab.
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Computing

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