

Structure and mechanics of active colloids

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Abstract

Minimal models of active Brownian colloids consisting of self-propelled spherical particles with purely repulsive interactions have recently been identified as excellent quantitative testing grounds for theories of active matter and have been the subject of extensive numerical and analytical investigation. These systems do not exhibit aligned or flocking states, but do have a rich phase diagram, forming active gases, liquids and solids with novel mechanical properties. This article reviews recent advances in the understanding of such models, including the description of the active gas and its swim pressure, the motility-induced phase separation and the high-density crystalline and glassy behavior.

Keywords: active matter, phase separation, active glasses, swim pressure

1. Introduction

Living entities, on scales from birds to individual cells, organize in complex patterns with collective behaviors that serve important biological functions. Examples range from the flocking of birds [1] to the sorting and organization of cells in morphogenesis [2]. Work over the last ten years has shown that many aspects of this complex organization can be captured by physical models based on a minimal set of rules or interactions, leading to the emergence of the new field of active matter [3]. This is defined as a distinct category of non-equilibrium matter in which energy uptake, dissipation and movement take place at the level of discrete microscopic constituents. The active matter paradigm has additionally inspired the development of ingenious synthetic chemical and mechanical analogues, such as “active” colloids: micron-size polystyrene spheres partly coated with a catalyst that promotes the decomposition of one of the components of the ambient fluid, resulting in self-propulsion of the colloidal particles [4, 5, 6, 7]. Collections of such active synthetic particles have been shown to spontaneously assemble in coherent mesoscale structures with remarkable life-like properties [8, 9].

Active systems exhibit rich emergent behaviors, where a collection of many interacting entities shows large-scale spatial or temporal organization in states with novel macroscopic properties. For instance, a dense swarm of bacteria can behave collectively as a living fluid with novel rheology [10, 11], self-organize in complex regular patterns [12], exhibit turbulent motion [13], or ‘freeze’ into a solid-like biofilm [14]. This type of

behavior is of course well known in inert inanimate matter that exhibits transitions between different phases upon the tuning of an external parameter, such as temperature, or the application of external forces that perturb the system at its boundaries (e.g., shear stresses) or globally (e.g., an electric or magnetic field). It acquires, however, a new unexplored richness in active systems that are tuned out of equilibrium by energy generated internally by each unit. The active matter paradigm aims at describing and classifying the behavior of this new class of non-equilibrium systems. It does so by drawing on our understanding of familiar states of matter and of the transitions between them as controlled by interactions between atoms and molecules. New states of matter arise when we put together many units that are individually driven or motile. How are these new states formed? Are they controlled solely by local interactions among the active particles or is chemical signaling required to understand the emergence of these new states? Can we classify and describe them and control the transitions between such states as we know how to do with familiar inert matter?

Colloids have played a key role in condensed matter physics as model systems for atomic materials where pair interaction can be customized and equilibrium phase transitions and glassy behavior can be investigated with optical microscopy [16]. Although many active entities, from bacteria to birds, are elongated in shape and order in states with local or global liquid crystalline order leading to collective flocking, a growing body of work on synthetic active systems, such as autophoretic colloids [5, 6, 17], rollers [18], and droplets [19], has shown that even spherical active particles can exhibit novel behaviors arising from the irreversible dynamics of each constituent, providing an excellent system for the quantitative testing of active matter theories. This has led to extensive theoretical and numerical studies of minimal models of active Brownian colloids con-

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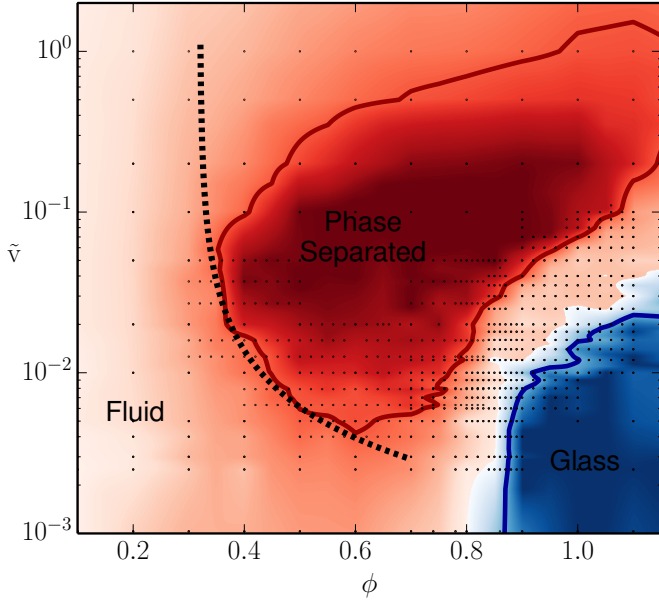


Figure 1: Numerical phase diagram of a polydisperse active suspension with soft repulsion obtained by integrating Eqs (1–2) for $D_r/(\mu k) = 5 \times 10^{-4}$ and $D_t = 0$, reproduced from Ref. [15] with permission from the Royal Society of Chemistry. The red region corresponds to a phase separated system. The blue region corresponds to a glass as characterized by the behavior of the MSD. The glass would be replaced by a crystalline state in a monodisperse suspension. The dotted line is the mean-field spinodal line given by $\rho_c^-(\text{Pe}_t)$ from Eq. (15) for $D_t = 0$.

sisting of self-propelled spherical particles with purely repulsive interactions [20, 21]. These systems do not exhibit aligned or flocking states, but form active gases, liquids and solids, as summarized in the phase diagram shown in Fig. 1. The interplay between motility and steric effects is responsible for intriguing new phenomena, including motility-induced phase separation in the absence of any attractive interactions [22, 20], Casimir-type forces [23], and ratchet effects [24]. These models have provided an excellent playground for addressing fundamental questions about the non-equilibrium statistical mechanics of active systems and whether equilibrium-like notions, such as effective temperature or equations of state, may be useful to describe them.

In this article we review recent advances on the theoretical description of collections of active Brownian particles (ABP) defined as spherical self-propelled particles with purely repulsive interactions by organizing their behaviors in terms of the new active gases, liquids and solid phases formed by these systems.

The rest of this paper is organized as follows. In Section 2 the minimal model of ABP is presented, emphasizing its parameters, limiting cases, and critical values. Next, in Section 3, we consider the properties of active gases in the context of recent work characterizing their mechanical properties and defining a pressure equation of state. From the dilute ideal active gas limit, tuning the rotational Péclet number and increasing density beyond a critical value results in phase separation, where signifi-

cant groups of active particles find their self-propulsion velocity caged due to increased interactions, yielding an active liquid coexisting with the active gas. This has been characterized by equations of mean velocity and critical density, which are presented and supported by simulation. This motility-induced phase separation (MIPS) is analyzed in Section 4 using continuum equations. The high-density limit, discussed in Section 5, is one in which solids and glasses of active colloids are formed, and this is distinguished from the glasses or crystalline states that results from high density collections of passive colloids. Finally, a discussion and outlook is provided in Section 6, raising current questions regarding the interplay of noise and damping in the current models of active systems.

2. A minimal model of active colloids

The rich behavior of active colloids has been studied using a minimal model of self-propelled particles (SPPs) that allows for both analytical and numerical progress. In this model hydrodynamic interactions are neglected and the ambient fluid is assumed to only provide friction, rendering the dynamics overdamped. Each colloid is modeled as a spherical particle of radius a_i , with an orientation defined by the axis of self-propulsion. In the following we will discuss both monodisperse systems, where all disks have the same radius a , and polydisperse systems, where a will denote the mean radius and the radii are uniformly distributed with 20% polydispersity. While most of the work on this model known in the literature as ABP has been carried out in two dimensions (2d), and we will restrict ourselves to this case here, many of the results described also hold in three dimensions [25]. Each particle is characterized by the position $\mathbf{r}_i$ of its center and its orientation $\mathbf{e}_i = (\cos \theta_i, \sin \theta_i)$ which in 2d corresponds to a single angle θ_i . The dynamics is then described by coupled Langevin equations

$$\dot{\mathbf{r}}_i = v_0 \mathbf{e}_i + \mu \sum_j \mathbf{f}_{ij} + \boldsymbol{\eta}_i(t), \quad (1)$$

$$\dot{\theta}_i = \eta_i^r(t), \quad (2)$$

where v_0 is the active (self-propulsion) speed and μ the mobility. The particles interact via short-range radial repulsive forces $\mathbf{f}_{ij} = -\frac{\partial U(|\mathbf{r}_{ij}|)}{\partial \mathbf{r}_i}$, where $U(r)$ is a pair interaction that depends on the separation $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ of the centers of the two particles. Both soft and hard repulsive forces have been used in the literature, and the qualitative behavior of the system is not affected by the details of the pair repulsion, provided it is pairwise, radial, and decoupled from the angular dynamics. Here we assume soft repulsive forces $\mathbf{f}_{ij} = f_{ij} \hat{\mathbf{r}}_{ij}$, with $\hat{\mathbf{r}}_{ij} = (\mathbf{r}_i - \mathbf{r}_j)/r_{ij}$, $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$, and $f_{ij} = k(a_i + a_j - r_{ij})$ if $r_{ij} < a_i + a_j$ and $f_{ij} = 0$ otherwise. We consider N disks in an area $A = L^2$, with $\rho = N/A$ the number density and $\phi = \sum_i \pi a_i^2 / A$ the packing fraction.

The random force in Eq. (1) describes thermal noise, with zero mean and correlations $\langle \eta_{i\alpha}(t) \eta_{j\beta}(t') \rangle = 2D_t \delta_{\alpha\beta} \delta_{ij} \delta(t - t')$, where $D_t = k_B T \mu$ is the translational diffusion coefficient and $k_B T$ the thermal energy. The direction of self propulsion fluctuates due to non-thermal processes. This is described by the

random torque η_i^r which is also chosen with zero mean and correlations $\langle \eta_i^r(t) \eta_j^r(t') \rangle = 2D_r \delta_{ij} \delta(t - t')$, with D_r the rotational diffusion rate, which is treated as an independent parameter because in many realizations, including bacterial suspensions [26] and active colloids [8], the rotational noise is athermal.

It is instructive to note that Eqs. (1) and (2) can be combined into a single equation of the form [22]

$$\partial_t \mathbf{r}_i = \boldsymbol{\xi}_i + \mu \sum_j \mathbf{f}_{ij} + \boldsymbol{\eta}_i(t), \quad (3)$$

where $\boldsymbol{\xi}_i(t) = v_0 \mathbf{e}_i(t)$ is a non-Markovian noise with zero mean and correlations

$$\langle \xi_{i\alpha}(t) \xi_{j\beta}(t') \rangle = D_a \delta_{\alpha\beta} \delta_{ij} \frac{e^{-|t-t'|/\tau_r}}{\tau_r} \quad (4)$$

with $D_a = v_0^2 \tau_r / 2$ the diffusion constant of the persistent motion. In other words the active forcing on the right-hand side of Eq. (1) is equivalent to non-Markovian noise with a persistence time $\tau_r = 1/D_r$. This key property of ABP arises because in this minimal model of active colloids the single-particle angular dynamics is decoupled from interactions and from the angular dynamics of other particles. Note that Eqs. 1 and 4 are distinct from Gaussian colored noise due to the bounded speed distribution here. In the limit $\tau_r \rightarrow 0$, for fixed D_a , the exponential decay can be approximated by a delta function, $(e^{-|t-t'|/\tau_r})/\tau_r \rightarrow \delta(t - t')$. In this limit, a single active Brownian particle behaves like a thermal Brownian particle with an effective temperature $k_B T_{\text{eff}} = \frac{v_0^2}{2\mu D_r}$ [22].

In interacting ABP the thermal limit is only realized, however, if the orientational correlation time τ_r is much smaller than all time scales present in the system. In particular, τ_r must be small compared to the mean free time between collisions, $\tau_f \simeq (2av_0\rho)^{-1}$, and to the interaction time $\tau_k = (\mu k)^{-1}$. The dimensionless ratio $\zeta = \tau_r/\tau_f$ is especially relevant for phase separation, whose onset has been interpreted in terms of a critical value of ζ [27]. A recent study has also pointed out the crucial role played by ζ in active suspensions, where hydrodynamic interactions are important [28].

We will see below that ABP can be tuned through active gas, liquid and solid phases by tuning activity, as measured by a dimensionless rotational Péclet number $\text{Pe}_r = v_0 \tau_r / a$ and the packing fraction ϕ ¹.

3. Active gases

It is useful to first examine the behavior of an ideal gas of SPPs. In the absence of interactions, each particle performs a persistent random walk. The dynamics is characterized by the persistence time τ_r of the path and the associated persistence length $\ell_p = v_0 \tau_r$. The rotational Péclet number also represents the ratio of the persistence length to the size of the particles,

$\text{Pe}_r = \ell_p / a$ and provides a measure of persistence or activity. The mean square displacement (MSD) of a single SPP can be calculated exactly and it is given by

$$\langle [\Delta \mathbf{r}(t)]^2 \rangle = 4D_r t + 2v_0^2 \tau_r \left[t - \tau_r (1 - e^{-t/\tau_r}) \right], \quad (5)$$

with $\Delta \mathbf{r}(t) = \mathbf{r}(t) - \mathbf{r}(0)$. The dynamics is ballistic for $t \ll \tau_r$, with $\langle [\Delta \mathbf{r}(t)]^2 \rangle \sim v_0^2 t^2$, and diffusive for $t \gg \tau_r$, with $\langle [\Delta \mathbf{r}(t)]^2 \rangle \sim 4[D_r + D_a]t$, and $D_a = v_0^2/(2D_r) = \ell_p^2/(2\tau_r)$. The same expression is obtained for run-and-tumble bacteria, with the replacement $D_r \rightarrow \alpha$ and α the tumble rate, indicating that the coarse-grained dynamics is insensitive to whether changes in direction of self-propulsion are continuous or discrete. A detailed comparison of rotational and run-and-tumble dynamics can be found in Ref. [32]. For both bacteria and active colloids the thermal diffusion coefficient D_t is typically two orders of magnitudes smaller than the active diffusion D_a . This justifies neglecting thermal noise in Eq. (1).

Recently there has been substantial theoretical effort towards characterizing the mechanical properties of an active gas and understanding whether this non-equilibrium system can be described by a pressure equation of state [33, 34, 29, 35, 36]. In equilibrium, the pressure p of a fluid can be defined in three equivalent ways: (i) as the mechanical force per unit area on the walls of the container, (ii) from thermodynamics as the derivative of a free energy, and (iii) as the trace of the hydrodynamic stress tensor of the fluid, which in turn represents the momentum flux in the system. In equilibrium all three definitions give the same expression and the pressure is a state function. Recent work by us and others has shown that the pressure is also a state function for an active fluid of spherical SPPs with purely repulsive interactions [34, 29, 35], but becomes dependent on the nature of the walls as soon as the active particles exert torques on each other or on the boundaries, as would for instance be the case for non-spherical SPPs [36].

The pressure p_D of a passive fluid with pairwise forces $\mathbf{f}_{ij}$ can be written as the trace of the stress tensor, using the familiar Irving-Kirkwood formula [37],

$$p_D = \left\langle \frac{1}{2A} \sum_{i < j} \mathbf{f}_{ij} \cdot \mathbf{r}_{ij} \right\rangle, \quad (6)$$

where A is the area of the system and the brackets denote an average over the noise. This expression highlights the meaning of pressure as describing moment flux across a unit line (or a unit plane in three dimensions). For passive gases in thermal equilibrium to this direct contribution from interaction one must add the momentum carried by free streaming particles, which gives the ideal gas pressure $p_0 = \rho k_B T$, with $\rho = N/A$ the number density. In a fluid of SPPs there is an additional contribution to the pressure that describes the flux of self-propulsive force across a unit line. This contribution, unique to active systems, was first identified in Refs. [33, 34, 29] and was dubbed ‘active’ or ‘swim’ pressure. We will use the latter term here that has now become accepted in the literature. In Ref. [34] it was shown that the swim pressure can be written in a virial-type form as

$$p_s = \left\langle \frac{1}{2A} \sum_i \mathbf{F}_i^a \cdot \mathbf{r}_i \right\rangle, \quad (7)$$

¹We stress that Refs. [29, 30] characterize activity in terms of a translational Péclet number defined in terms of the diffusivity D_a , hence proportional to $1/v_0$. Here we prefer to use the rotational Péclet number, which is commonly used in the active matter literature and is a direct measure of persistence.

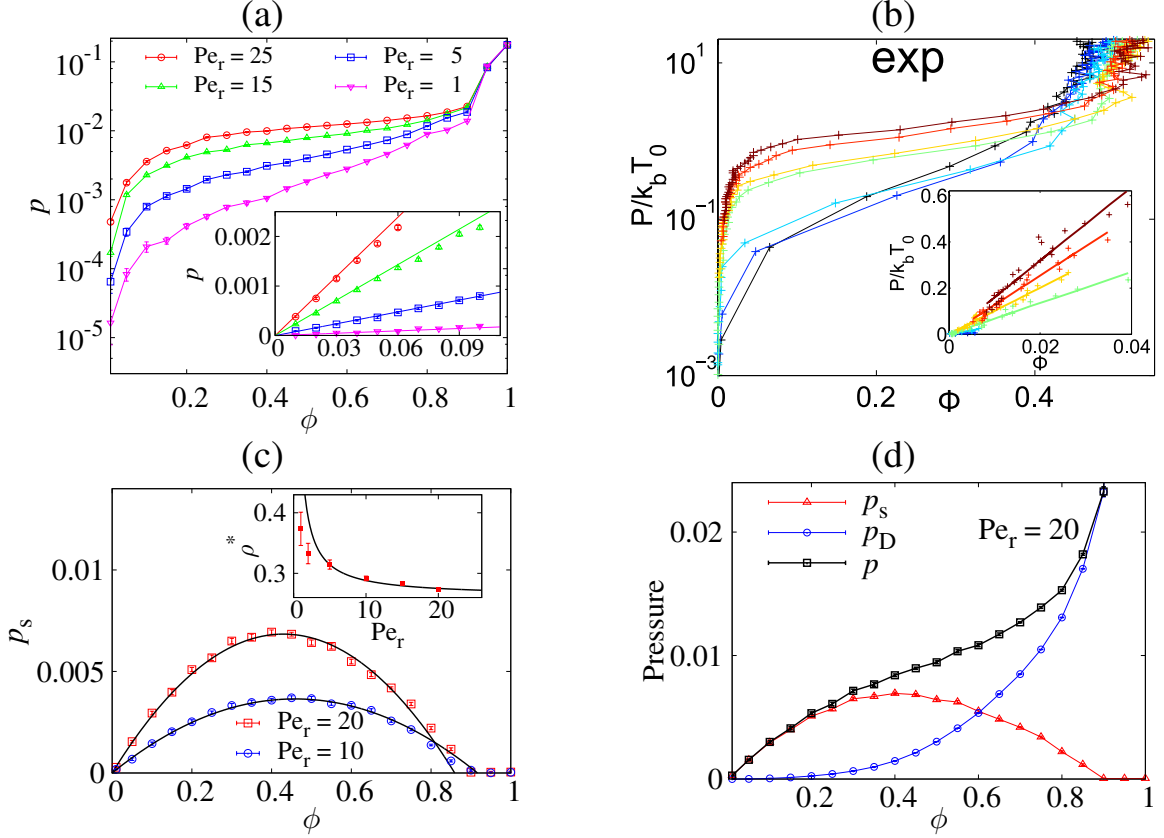


Figure 2: Pressure as a function of packing fraction ϕ from (a) our simulations of self-propelled particles with soft repulsive interactions for increasing values of $Pe_r = \ell_p/a$, obtained by decreasing D_r at fixed v_0 , and (b) sedimentation experiments of active Janus colloids by Ginot *et al.* [31] (reproduced with permission from Ref. [31]), with $T_0 = 300K$ the ambient temperature and the various curves corresponding to increasing concentration of H_2O_2 resulting in increasing self-propulsion speed (from bottom to top). 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 where the notion of effective temperature holds. In the inset of (a) the lines are the ideal gas swim pressure given in Eq. (8), with no adjustable parameters. Frame (c) shows the swim pressure vs ϕ for $Pe_r = 10, 20$. The solid lines are fits to Eq. (9) using $v(\rho) = v_0(1 - \rho/\rho_*)$ (for each Pe_r ρ_* is the only fit parameter). The inset shows $\rho_*(Pe_r)$ and a fit to (10). Frame (d) displays the various contributions to the pressure for $Pe_r = 20$. 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 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.

where $\mathbf{F}_i^a = v_0 \mathbf{e}_i / \mu$ is the propulsive force on each particle. The total pressure of an active gas is then $p = p_0 + p_D + p_a$. In the remainder of this section we will consider active systems at $T = 0$ and ignore the thermal ideal gas contribution. It was shown in Ref. [34] that for repulsive self-propelled disks the pressure calculated as the sum of Eqs. (6) and (7) is indeed identical to the force per unit area on the walls of the container, demonstrating that in this system the pressure is a state function. When the active units exchange torques among themselves and/or with the walls of the container, as is the case for non-spherical particles or in the presence of orientation-dependent interactions, the force on a wall depends on the properties of the wall and the definition of a mechanical state function that plays the role of pressure does not seem possible [36].

In the absence of interactions $p_D = 0$ and one can readily calculate the pressure p_s^0 of an active ideal gas using Eq. (7) and assuming that the initial position of the particle is not correlated with the noise, with the result $p_s^0(t) = \frac{\rho v_0^2}{2\mu D_r} (1 - e^{-D_r t})$. In the limit of large systems, one can let $t \rightarrow \infty$ in this expression to obtain the pressure of an ideal active gas in the thermodynamic

limit,

$$p_s^0 = \rho \frac{v_0^2}{2\mu D_r} = \rho k_B T_{\text{eff}}. \quad (8)$$

In other words the swim pressure of an ideal active gas is simply the pressure of an ideal gas at the temperature T_{eff} . Although Eq. (8) replaces the kinetic contribution to the pressure that one would have in a thermal passive gas, we will see below that, unlike the ideal gas pressure of thermal systems, this contribution is strongly renormalized by interactions. Additionally, active systems are much more sensitive to finite size effects and care is required when taking the thermodynamic limit. If the persistence length ℓ_p is comparable to or exceeds the linear size L of the container, then particles travel ballistically and one can estimate the pressure as $p_a^0(t \sim L/v_0) = \frac{\rho v_0^2}{2\mu D_r} (1 - e^{-L/\ell_p}) \simeq \frac{\rho v_0 a L}{2\mu}$ where the last approximate equality holds when $\ell_p \gg L$. For high activity, the active gas behaves like a highly diluted Knudsen gas and the pressure depends on the size of the container. It has also been shown recently that boundary curvature can strongly affect the density and force distribution in ABP [38]. Care must therefore be taken in numerical simu-

lations as the mapping of a dilute active gas of purely repulsive spherical colloids onto a thermal gas at an effective temperature $T_{\text{eff}} = v_0^2/(2\mu D_r)$ only holds when $\ell_p \ll L$. Strong finite-size effects are observed when ℓ_p is a fraction of L . In this case active gases behave quite differently from thermal ones: they accumulate at the walls of the container [34] or around fixed impurities, exert Casimir-type forces [23], and spontaneously sort when different in size or activity [34]. Péclet numbers in excess of 100, corresponding to persistence lengths of order 100 particle sizes, are easily reached in experiments in active colloids [39] and finite size effects should be observable in microfluidic devices.

At higher density, interactions become important. It is instructive to note that the kinetic contribution to the pressure of thermal Brownian particles can also be calculated through a virial-type expression akin to the one defining the swim pressure of ABP, given by $p_0 = \lim_{t \rightarrow \infty} \frac{1}{2A} \langle \sum_i \eta_i \cdot \mathbf{r}_i \rangle$, with η_i the thermal noise. Assuming, as customary, that the initial position of the particles is not correlated with the noise, and making use of the fact that the noise is uncorrelated in time, this expression gives exactly $p_0 = \rho k_B T$ at all densities ρ . In contrast, the stochastic propulsive force entering the definition of the swim pressure, Eq. (7), is correlated in time (see Eq. (4)), leading to the dependence of the swim pressure on interparticle forces. The total pressure of a monodisperse system of repulsive SPPs is shown in Fig. 2(a) as a function of packing fraction for several values of the persistence length. At low density the system behaves like a gas of ‘hot’ repulsive colloids with effective temperature T_{eff} , as shown in the inset. At intermediate density the swim pressure is strongly suppressed due to collisional slowing down (Fig. 2(c)) and the total pressure remains almost constant over a range of packing fractions. This is the region where the system spontaneously phase separates, as discussed below, and a fluid of repulsive SPPs behaves qualitatively like a passive fluid of attractive particles. At high density the swim pressure is negligible and repulsive interactions dominate, yielding the sharp increase of the pressure associated with the transition to a glassy or solid state, as in passive systems. The pressure of active Janus colloids was recently measured in sedimentation experiments [31], confirming the prediction of the simulations (see Fig. 2(b)). These experiments also confirmed that dilute active gases behave like ‘hot’ passive colloids with an effective temperature enhanced by activity and that this mapping holds even in the presence of interactions up to moderate densities, but below the region of spontaneous phase separation.

It was shown in Ref. [35] that in the thermodynamic limit the swim pressure given in Eq. (7) can be recast in an intuitive form, given by

$$p_s = \rho \frac{v_0 v(\rho)}{2\mu D_r}, \quad (9)$$

where $v(\rho)$ is the mean velocity of the particles along the direction of self-propulsion, given by $v(\rho) = v_0 + \mu \langle \mathbf{e}_i \cdot \sum_{j \neq i} \mathbf{f}_{ij} \rangle$. We note that this expression only holds if $\ell_p \ll L$ and in fact reduces to the bulk pressure of an active ideal gas, $p_s^0 = \rho v_0^2/(2\mu D_r)$, in the absence of interactions. The suppression of the swim pressure due to collisional slowing down can then be

modeled phenomenologically at moderate densities as a linear decay of the propulsion speed, with $v(\rho) = v_0(1 - \rho/\rho_*)$ for $\rho < \rho_*$ and $v(\rho) = 0$ for $\rho > \rho_*$. In general ρ_* will depend on Pe_r . This linear decay of $v(\rho)$ with ρ has been seen in simulations [22] and justified by relating the linear decay rate to the pair correlation function [40] and by estimating it via a kinetic argument [41, 15] as arising from the fact that at finite density particles can be stalled for the duration τ_c of collisions with other particles. At low density, where the mean free time between collisions $\tau_f = (v_0 2a\rho)^{-1}$ exceeds τ_c , the mean speed can then be written as $v(\rho) = v_0(1 - \tau_c/\tau_f)$. The collision time τ_c was treated as a constant fitting parameter in Ref. [41], but will in general depend on Péclet number. It can be estimated as controlled by two delay mechanisms: the time a/v_0 it takes two interacting particles to move around each other and the reorientation time D_r^{-1} . Since the collision time τ_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

$$\rho_*(\text{Pe}_r) = \frac{c}{a^2} \left(1 + \frac{1}{\text{Pe}_r} \right), \quad (10)$$

where c is a number of order unity. In our simulations (see Fig. 2), we find that this expression works well for $\text{Pe}_r > 2$, with $c \approx 0.26$. The swim pressure for moderately dense gases can then be written as $p_s = \rho \frac{v_0^2}{2\mu D_r} \left[1 - \frac{c\phi}{\pi} (1 + 1/\text{Pe}_r)^{-1} \right]$. At low density, the direct part of the pressure can be estimated as $p_D = \rho(v_0/\mu)\rho a^3$, where v_0/μ is the force scale and ρa^3 is the typical displacement of a particle between collisions [30]. This expression holds as long as the particles do not overlap.

4. Active fluids and motility-induced phase separation

The most striking phenomenon exhibited by our minimal model of active colloids is motility-induced phase separation (MIPS), where an active fluid of particles with purely repulsive interactions spontaneously phase separates into a dense liquid surrounded by an active gas [22]. This remarkable phenomenon arises from the collisional slowing down of the active particles’ speed $v(\rho)$ that yields the suppression of the swim pressure shown in Fig. 2(c), combined with the fact that the lack of detailed balance allows self-propelled particles to accumulate in the regions where $v(\rho)$ is small [42, 43]. While experiments often see strong clustering, but not complete phase separation [17, 8], MIPS has now been seen in numerical simulations for a variety of repulsive interaction, ranging from soft harmonic potential to WCA and hard spheres, in both two and three dimensions [21, 44, 25].

The phase behavior of the system, as obtained from simulations of the polydisperse limit of the model described in Section 2, is summarized in Fig. 1. Three regimes can be identified: a homogeneous fluid phase, a solid or frozen state at high packing fraction and relatively low activity, and a regime where the system shows macroscopic phase separation into two bulk phases. The phase separated regime exists in an intermediate range of packing fraction and activity, corresponding to the region where the pressure remains approximately constant

with increasing packing fraction. For soft repulsion, it changes continuously from a high density liquid cluster surrounded by a gas at lower packing fractions, to a hole of gas phase inside a densely packed liquid at higher packing fractions [15]. The existence of a homogeneous liquid phase intermediate between the solid and the phase separated state seems to be a generic feature, but is not fully understood. In contrast, the suppression of phase separation at high self-propulsion speed is specific to the soft nature of the repulsive potential used in our simulations, where for $v_0/a\mu k > 1$ particles can just pass through each other.

The threshold $\rho_c(\text{Pe}_r)$ for phase separation has been estimated in various ways in the literature. A naive estimate is obtained by assuming that phase separation occurs when $\tau_f < \tau_r$, corresponding to the condition that a particle experiences many collisions before changing direction, i.e., it gets trapped by other particles, with the result $\rho_c(\text{Pe}_r) \sim (a^2\text{Pe}_r)^{-1}$. The same estimate was obtained by Redner *et al.* [27] via a kinetic argument that equates the flux $\rho_{\text{gas}}v_0$ of particles that enter the cluster to the flux out of the cluster, $\kappa D_r/a$, with κ a fitting parameter. The dependence on D_r arises because particles must turn around to point outward before they can leave the cluster. 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 ρ and 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 for arbitrarily large Pe_r phase separation will occur no matter how low the density. This behavior is not, however, born out by simulations that find a minimum threshold of density below which there is no phase separation.

Another approach is to use continuum equations for an active fluid where motility suppression is incorporated through the density-dependent propulsion speed $v(\rho)$ that controls the swim pressure [43, 22, 45, 41, 40], and then employ linear stability analysis to locate the spinodal. The dynamics is governed by equations for the density $\rho(\mathbf{r}, t)$ and the polarization density $\mathbf{p}(\mathbf{r}, t)$ that describes the local orientation of the particles' axis of self-propulsion, given by [22]

$$\partial_t \rho = -\nabla \cdot [v(\rho)\mathbf{p} - D_r \nabla \rho], \quad (11)$$

$$\partial_t \mathbf{p} = -D_r \nabla^2 \mathbf{p} - \nabla \pi(\rho) + K \nabla^2 \mathbf{p}, \quad (12)$$

where $\pi(\rho) = v(\rho)\rho/2 = p(\rho)\mu D_r/v_0$, with $p(\rho)$ the pressure. For times $t \gg D_r^{-1}$, one can neglect the time derivative of the polarization in Eq. (12) relative to the damping term and eliminate $\mathbf{p}$ to obtain a nonlinear diffusion equation for the density, given by

$$\partial_t \rho = \nabla \cdot [\mathcal{D}(\rho) \nabla \rho], \quad (13)$$

with effective diffusivity

$$\mathcal{D}(\rho) = D_r + \frac{v^2(\rho)}{2D_r} \left(1 + \frac{d \ln v}{d \ln \rho} \right). \quad (14)$$

The linear stability of a homogeneous state of constant density $\bar{\rho}$ can be analyzed by examining the dynamics of density fluctuations, $\delta \rho = \rho - \bar{\rho}$. The decay of fluctuations is controlled by a diffusive mode with diffusion constant $\mathcal{D}(\bar{\rho})$ that can become negative, signaling the instability of the uniform state.

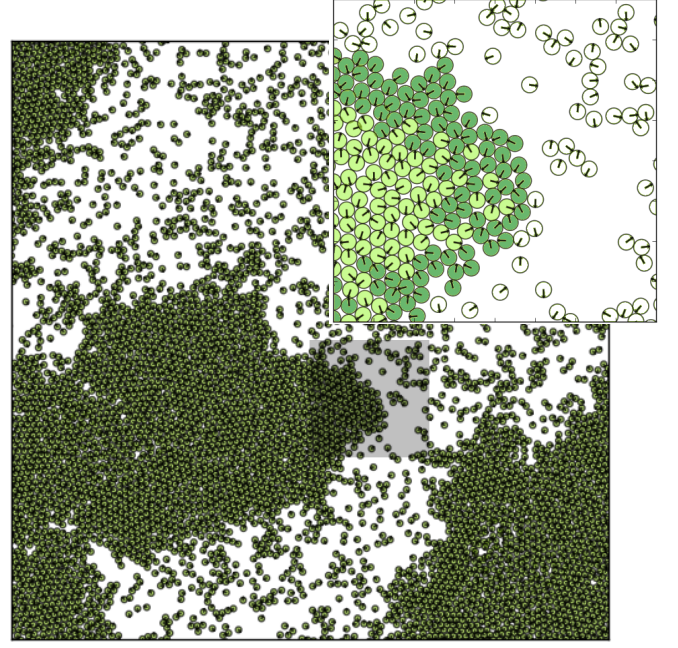


Figure 3: Snapshot of an ABP simulation with inset zooming in on the shaded region of the background image. The inset shows particles in the dense liquid phase tagged with green (dark green for inward-pointing boundary particles, light green for particles in the bulk of the dense liquid), while gas particles 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 $\text{Pe}_r = 50$.

The onset of the instability corresponds to $\mathcal{D}(\bar{\rho}) = 0$, which determines the spinodal line. When $D_t = 0$ the threshold for phase separation in $\rho_c(\text{Pe}_r) = \rho_*(\text{Pe}_r)/2 = (2a)^{-2} (1 + \text{Pe}_r^{-1})$. This expression does yield a minimum density below which there is no phase separation, as seen in simulations [15], but it fails to account for the observed lower bound for the value of Pe_r required for phase separation. A lower bound is obtained when $D_t \neq 0$. In this case one finds

$$\rho_c^\pm(\text{Pe}_r) = \frac{\rho_*(\text{Pe}_r)}{4} \left(3 \pm \sqrt{1 - \left(\frac{\text{Pe}_r^c}{\text{Pe}_r} \right)^2} \right), \quad (15)$$

where $\text{Pe}_r^c = 4 \sqrt{D_t/(a^2 D_r)}$ represents the minimum value of Pe_r required for phase separation. Thermal diffusion suppresses phase separation and drives this critical Pe_r to larger values. The condition $\text{Pe}_r > \text{Pe}_r^c$ for phase separation can also be written as $D_t < v^2/a^2 D_r$, indicating that the observed spinodal decomposition requires a sufficiently small D_r or long persistence time. The expression given in Eq. (15) with the choice of minus sign corresponds to the dotted line in Fig. 1. It is, however, unclear whether thermal diffusion is really the mechanism responsible for the lower bound on Pe_r , given the simulations leading to Fig. 1 were carried out with $D_t = 0$ and do show a finite threshold. Although a finite value of D_t can be generated by interactions even if $D_t = 0$ at the single particle level in the numerical model, this discrepancy suggests that other mechanisms not yet

understood may be at play here.

For finite D_t one must in general retain the dynamics of the polarization. The analysis of the modes in this case can be found in Ref. [15] and yields a growth rate with the characteristic behavior expected for a spinodal, with a maximum growth rate at wavevector $\sim \sqrt{-\mathcal{D}/2KD_t}$.

By including noise in the continuum model, one can examine the spectrum of fluctuations and evaluate the structure factor $S(q) = \langle \delta\rho_q \delta\rho_{-q} \rangle$. At small wavevector one finds $S(q) \sim 1/(q^2 + \xi^{-2})$, with $\xi = \sqrt{KD_t/(D_t\mathcal{D})} \sim [\rho - \rho_c(\text{Pe}_r)]^{-1/2}$ a correlation length that diverges on the spinodal line [22]. Both the divergence of $S(q=0, \rho)$ at $\rho = \rho_c$ and the scaling $S(q, \rho_c) \sim q^{-2}$ are consistent with the behavior at an equilibrium gas-liquid spinodal line. Although the numerics can be fitted to this behavior [22], the divergence of ξ is generally masked by nucleation processes that further support the liquid-gas analogy [27]. A true divergence is expected at the critical point at Pe_r^c , but it has not yet been possible to extract numerical values for the critical exponents. It has also been shown numerically that the coarsening dynamics closely resembles that of an equilibrium spinodal decomposition and that activity weakly affects on the growth in time of the domains size $L(t)$ that is found to be very close to the Cahn-Hilliard form, $L(t) \sim t^{1/3}$, describing the coarsening of a passive system with no momentum conservation² although differences exist when gradient terms accounting for interfacial phenomena are included in the theory [27, 41, 46]. In contrast, experiments in bacteria and active colloids generally do not see complete separation in two bulk phases, but rather observe the assembly of finite-size clusters that persist for very long times. In bacterial suspensions birth/death events that eliminate density conservation have been shown to arrest the phase separation yielding the formation of finite-size clusters [47, 34]. Attractive interactions have also been suggested as a possible mechanism for arresting the phase separation [8], but the mechanism that control the size of finite clusters in active colloids remain to be understood.

The q^{-2} divergence of the structure factor as $q \rightarrow 0$ is also the signature of the giant number fluctuations (GNF) predicted and observed ubiquitously in the ordered state of flocking models [48, 49, 50, 3]. MIPS and GNF are, however, two distinct phenomena. In repulsive SPPs the large density fluctuations occur in a disordered state and are associated with a critical point and phase separation. The large density fluctuations that occur in the ordered state of active fluids with both polar and nematic order are the consequence of the coupling of a conserved field - the density - to a non-conserved noisy polarization field. The q^{-2} behavior of the correlation is in fact a generic feature of systems where the deterministic dynamics of a conserved field couples to non-conserved noise, resulting in power-law decay of correlations [51], and is also ubiquitous in driven granular materials with inelastic interactions [52].

The onset of MIPS suggests that at moderate density a system of repulsive active colloids behaves like a thermal system

with attractive interactions. This correspondence was recently demonstrated by correlating the pressure measured in sedimentation experiments of active colloids with numerical simulations [31]. It has additionally been explored in models of Brownian particles with Gaussian colored noise correlated over a time τ_r , where the colored nature of the noise can be incorporated as an effective interaction in a many-body Fokker-Planck equation [53, 54]. Using this approach it has been shown recently that for weakly persistent motion (small τ_r) activity can be recast in the form of an effective pair potential with an attractive part of strength that increases with increasing Pe_r [55]. On the other hand, when the pair interparticle potential actually has an attractive component, activity can yield a weak repulsive contribution to the effective potential, hence competing with attraction by promoting particle escape from the potential well. In other words, self-propulsion can actually drive a system that has undergone bulk phase separation due to attractive interactions back into a uniform state. It should be noted, however, that the ABP model and the model with Gaussian colored noise are characterized by different speed distributions, which is Gaussian in the latter, but bounded by v_0 in ABP and in run-and-tumble models. Simulations have additionally demonstrated that the competition of attraction and activity yields an intermediate regime where complete phase separation is arrested and particles assemble in a gel-like structure that has been seen in simulations in both two and three dimensions [56, 55, 57].

Finally, it has been shown that hydrodynamic interactions can suppress phase separation by aligning the particles and therefore suppressing the self-trapping responsible for cluster formation [58, 59]. On the other hand, strong clustering is often observed in active systems even in the presence of aligning rules [3], indicating that more work needs to be done to understand the effect of aligning torques on the MIPS of repulsive active particles.

5. Active crystals and active glasses

At large packing fraction ABP have been shown to freeze in solid states. While this may not seem surprising since purely repulsive systems are known to form crystalline and glassy states in equilibrium, it is not a priori obvious that such states can survive in the presence of continuous energy input at the microscopic scale. Key to the formation of active solid states is the fact that steric repulsion strongly suppresses motility at high density, as discussed in Section 4. This results in particle caging and in what was first described by Henkes and collaborators as ‘active jamming’ [60], although that work was for a model of repulsive SPPs with alignment interactions, where confinement was required to impede collective flocking.

In equilibrium freezing in two dimensions is a continuous, two-step process, where the system goes from a liquid to a hexatic liquid state with quasi-long range bond orientational order, and then to a crystalline state with long-range orientational order and quasi-long range translational order. Crystallization can be quantified using both structural and dynamical criteria. The onset of translational order identified via structural probes generally coincides with criteria for dynamical ar-

²Values of the coarsening exponent slightly smaller than $1/3$ have been reported in simulations [27, 41], but it is not clear whether these simulations had indeed reached the asymptotic regime.

rest, such as the vanishing of the long-time diffusion coefficient $D = \lim_{t \rightarrow \infty} \frac{1}{4t} \langle \Delta r(t)^2 \rangle$. Crystalline order has been seen in phase separating active systems both in experiments and simulations [40, 8, 61, 62]. In simulations of monodisperse systems the dense phase in the phase separated regime exhibits substantial structural order resembling a colloidal crystal near the hexatic-crystal transition, but with unusual super-diffusive behavior [27]. Crystalline clusters have also been seen in the experiments, such as the ‘living crystals’ formed by light activated colloids engineered by Palacci *et al.* [8]. Numerical simulations have also examined the onset of bond-orientational order in ABP [27] and its correlation with structural arrest, concluding that the onset of order remains continuous, but is shifted to higher density than in equilibrium systems [63]. Additionally, Bialke *et al.* [63] reported the existence of an intermediate region between the liquid and the solid states characterized by large structural fluctuations. It is tempting to identify this phase as an active hexatic phase similar to but distinct from the equilibrium hexatic that has been observed for instance in superparamagnetic colloidal particles confined to an air-water interface [64]. Finally, in models with aligning interactions the interplay of activity, alignment and structural order can lead to partially ordered liquid crystalline resting and traveling states that are beyond the scope of the present article [65, 66].

Our own work has focused on polydisperse systems, where even in equilibrium crystalline order is bypassed in favor of glassy or jammed states. In this case the characterization of glassy behavior relies on criteria for dynamical arrest, such as the strong suppression of diffusion at intermediate times, when particles are persistently caged. Fig. 4 shows the evolution of the mean square displacement of a polydisperse assembly of repulsive SPPs over a range of packing fractions. At small and intermediate packing fraction the long-time dynamics remains diffusive, but at high packing fraction the MSD is bounded over the time of the simulation, indicating a jammed or frozen state. The fluctuations in the actively jammed state are intimately connected to the low frequency or ‘soft’ modes of a solid that are associated with the jamming transition of passive thermal systems [60]. At small v_0 active systems form solid phases that can be analyzed in terms of a complete set of normal modes with normal frequencies ω , determined by the inter-particle interactions [60]. As in the familiar description of lattice vibrations in terms of phonons, the lowest frequencies correspond to large scale collective structural rearrangements of the system. Work on passive granular matter has shown that such low frequency modes have much lower energy barriers than high frequency modes [67]. The jammed state is then a steady state with structural rearrangements involving a handful of low frequency modes that also provide easy paths for unjamming as v_0 is increased. This hints at a generic connection between the dynamics of jammed active systems at low D_r and the kinetic arrest of passive glassy systems, as explored in Refs. [68, 69, 55]. Note that the prominence of low frequency modes in the active jamming dynamics is strongly linked to the polarization-velocity alignment mechanism, and more work is necessary to disentangle the interplay of activity, alignment and caging.

One interesting feature of the onset of glassy behavior in re-

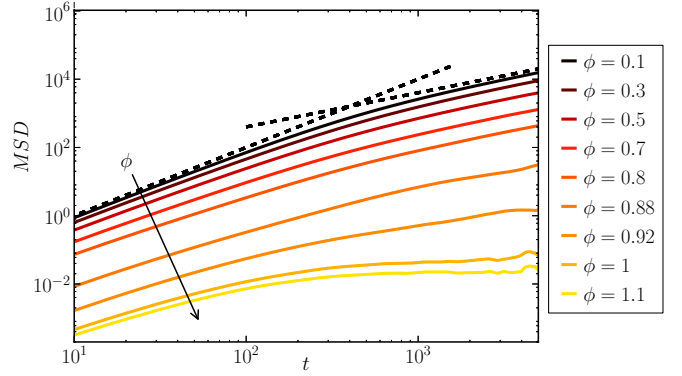


Figure 4: MSD versus time for various packing fractions ϕ from our simulations of polydisperse ABP with soft repulsive interactions, Eqs. (1–2), for $Pe_r = 20$ showing ballistic growth at small times and the crossover from diffusive to bounded behavior with increasing packing fraction at long time. The dashed lines have slopes 2 and 1.

pulsive SPPs is the shift of the glass transition to packing fraction higher than the value $\phi_G \approx 0.8$ obtained for thermal systems in the limit of vanishing motility, $v_0 \rightarrow 0$ [70, 71, 15]. This shift increases with increasing persistence time τ_r and has been reported in both simulations of SPPs with hard repulsions where activity pushes the glass transition towards the random close packed limit of $\phi_{RCP} = 0.842$ and in our simulations of soft disks (see phase diagram of Fig. 1), where we find $\phi_G(v_0 \rightarrow 0) \approx \phi_{RCP}$ for $D_r/\mu k = 5 \times 10^{-4}$.

Finally, Ni *et al.* have shown numerically that doping a hard sphere glass with a small number of active particles promotes crystallization bypassing the glass transition [72]. This suggests that a small concentration of active particles may be used as a means for facilitating the formation of large crystalline states that would be otherwise hindered by defects and grain boundaries.

6. Discussion and Outlook

Collections of repulsive self-propelled particles, also known as active Brownian particles, provide a minimal realization of an active system with rich non-equilibrium behavior. They exhibit active gas, liquid and solid phases with novel mechanical properties. At low density they behave as ‘hot’ colloids with an effective temperature controlled by activity and a novel swim pressure unique to active systems. At intermediate density they resemble attractive colloids and exhibit a motility-induced phase separation. At high density they form solid and glassy phases that have provided new insights in the physics of glassy and jammed solids.

The key property that has allowed much theoretical progress in the description of this minimal active system is that the noisy single-particle angular dynamics is decoupled from interactions. In other words active Brownian colloids do not exert torques on each other nor on the walls of a container. As a consequence the effect of activity can be mapped onto a non-Markovian noise of strength controlled by the self-propulsion speed and with time correlation given by the persistence time

τ_r , as shown in Eqs. (3–4). This has led to renewed interest in stochastic models of Brownian particles with Gaussian colored noise, for which exact results exist in the literature [53, 54, 73, 74]. Although these models are not precisely equivalent to ABP due to the different speed distributions, useful insight has been obtained from these studies [55, 69]. On the other hand, torques are likely to be present in most experimental realizations, as they can arise from the non-spherical shape of the particles, from lateral friction, from hydrodynamic interactions or explicit aligning rules, and will generally modify the collective behavior of the systems. It has already been shown, for instance, that the pressure is a state function only for the case of repulsive spherical active Brownian particles, independent of the nature of the interaction of the system with the walls. The presence of torques may also arrest the bulk phase separation, leading instead to clustering or micro-phase separation, as ubiquitously seen in experiments. Work remains to be done to fully understand how alignment, induced either by anisotropic steric effects or by aligning interactions, modifies the phase behavior of active colloids and to make quantitative contact with experiments.

In the limit of small persistence time ($\tau_r \rightarrow 0$), active Brownian colloids are equivalent to Brownian colloids. The opposite limit of $\tau_r \rightarrow \infty$ is intriguing and has not been fully explored. In this limit the active drive becomes correlated in time and resembles quenched disorder. At high density the fluctuations patterns tend to exhibit both spatially and temporally long-ranged correlations [75] and resemble those seen in sheared passive granular matter [76, 77], suggesting an intriguing connection that needs to be further investigated.

Finally, quite unexplored is the effect of noise in the continuum dynamics as described for instance by Eqs. (11) and (12). Most work using phenomenological continuum models to describe active matter has focused on deterministic equations or has included white Gaussian noise in the linear theory to evaluate correlation functions [22]. On the other hand, the noise in active systems is in general multiplicative in the density [78] and may, for instance, provide a mechanisms for selecting non-linear dynamical states. The minimal model of active Brownian colloids may provide an excellent playground for beginning the exploration of the role of multiplicative noise in mesoscopic theories of active matter.

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