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Surface tension and interfacial phenomena in active matter

by

Luke Anthony Langford

A dissertation submitted in partial satisfaction of the

requirements for the degree of

Doctor of Philosophy

in

Engineering - Materials Science and Engineering

in the

Graduate Division

of the

University of California, Berkeley

Committee in charge:

Professor Ahmad K. Omar, Chair

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Spring 2026

Surface tension and interfacial phenomena in active matter

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Luke Anthony Langford

Abstract

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Doctor of Philosophy in Engineering - Materials Science and Engineering

University of California, Berkeley

Professor Ahmad K. Omar, Chair

Active matter, composed of agents that consume energy at the microscopic scale, exhibits a wealth of collective behaviors that transcend traditional equilibrium thermodynamics. An example of this behavior is motility-induced phase separation (MIPS), where self-propelled particles assemble into dense and dilute phases in the absence of attractive interactions. While the bulk phase behavior of MIPS is becoming increasingly well-understood, the interfaces separating these active phases remains a subject of significant debate. Surprising observations, such as the measurement of negative surface tensions and the emergence of unique "bubbly" phase separation in two dimensions, challenge our traditional understanding of interfaces. This thesis is devoted to resolving these outstanding questions surrounding active interfaces. We first develop a generalized capillary-wave theory for active systems by deriving an equation of motion for active interfaces and extracting out a non-equilibrium surface tension informed by the microscopic particle dynamics. We are able to extend this generalized capillary-wave theory to chiral systems, predicting (and verifying) the presence of odd-surface flows when spatial parity symmetry is broken. We then investigate nucleation and coarsening in active matter, providing a route to calculating finite-size corrections to binodals

and extending classical coarsening theories to non-equilibrium systems. Finally, we examine the origins of bubbly phase separation in two-dimensional MIPS, revealing that appreciable formation of bubbles depends on the interplay between hexatic order and translational noise. Collectively, this work aims to establish a more systematic theoretical foundation for understanding the structure, stability, and dynamics of interfaces in active matter.

This work is dedicated to my parents

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Publications by the author used in this thesis

L. Langford and A. K. Omar, *Phys. Rev. E* 110, 054604 (2024)

L. Langford and A. K. Omar, *J. Chem. Phys.* 163, 064901 (2025)

L. Langford and A. K. Omar, *Phys. Rev. Lett.* 134, 068301 (2025)

L. Langford and A. K. Omar, arXiv:2603.17320

Chapter 1

Introduction

1.1 What is active matter?

Much of the phenomena observed in the *natural* world, from the flocking of birds [1] to the healing of wounds [2], are processes made possible by the consumption of fuel at the smallest relevant lengthscales. This microscopic injection of energy into a system by its *living* components results in a breakdown of thermodynamic equilibrium. This breakdown of thermodynamic equilibrium is qualitatively distinct from non-equilibrium systems which are microscopically passive, such as metastable systems which eventually relax to equilibrium (e.g. a diamond that eventually transforms to graphite) or boundary-driven systems (e.g. a thermally conductive metal rod held at different temperatures on each end). Indeed, when systems remain passive at microscopic length scales, theoretical tools built on top of traditional thermodynamics such as Cahn-Hilliard dynamics [3], Onsager's regression hypothesis [4], or linear response [5, 6] remain meaningful. Active systems, on the other hand, display microscopic dynamics that are no longer variational and thus break free from the constraints of Boltzmann statistics. This results in the remarkable variety of behaviors observed in the living world from the organization of membrane-less organelles [7, 8] to the dangers of collective oscillations in human crowd surges [9].

It has recently become clear that the phenomenology of active matter is not limited to the living world. *Artificial* systems of colloidal particles which microscopically consume energy to produce mechanical work have been synthesized in a variety of ways. Examples of such systems are diverse: ranging from catalytic beads immersed in hydrogen peroxide [10], oil droplets converting chemical gradients to Marangoni flows [11], to Janus particles driven by electromagnetic fields [12]. These active colloids have potential for applications ranging from microscopic patterning beyond the scope of lithography [13] to soft programmable microrobotics [14]. However, the departure from equilibrium results in a lack of theoretical tools for understanding their behavior. This has motivated theoretical work towards rebuilding thermodynamic-like tools which may be applied to active systems. One of the classic thermodynamic tools is that of phase equilibrium, and because active systems ubiquitously display phase separation [15], it would

behoove us to extend the thermodynamics of phase behavior to active matter.

1.2 Motility-Induced Phase Separation and Interfaces

From spatial segregation of colonies of bacteria [16] to suspensions of self-catalytic colloids [10], swaths of active matter systems display phase separation induced by the propulsion of their individual agents (such as bacterium or colloidal particles) – even when no such phase behavior exists in the absence of the propulsion. This phase behavior is thus referred to as *motility-induced phase separation*, or MIPS.

Qualitatively, MIPS can be understood on relatively simple terms. Collisions between self-propelled particles results in a reduction of speed with increases in density. This reduction in speed implies that clusters of particles resulting from random fluctuations are slow to dissolve. At high enough density or active speed, this can result in a positive feedback loop resulting in spontaneous growth of clusters and eventually phase separation [15]. A crucial step towards quantifying MIPS was taken by Takatori *et al.*, who found that the active force exerted by self-propelled particles results in the emergence of a pressure-like quantity [17, 18]. This “active pressure” is proportional to the active force and density of particles, but at sufficiently high densities was measured to decrease due to the aforementioned reduction in effective speed. Thus, even if the pressure due to particle interactions (which governs phase behavior at equilibrium) monotonically increases with density, the combination of the active pressure and interaction pressure may be non-monotonic and therefore contain a spinodal. Multiple works [19–21] were able to apply the concept of an active pressure (or, in some cases, chemical potential) towards quantification of the binodal densities by generalizing the Maxwell equal-area construction¹. The phase diagram of a model system of active matter and snapshot of a simulation of the corresponding system is presented in Fig. 1.1.

¹Note that a naïve application of the Maxwell construction does not accurately predict the binodals outside of equilibrium, as illustrated in Fig. 1.1. Careful work must be done to identify system-specific integration variables to perform the construction with respect to. See Ref. [21] for further detail.

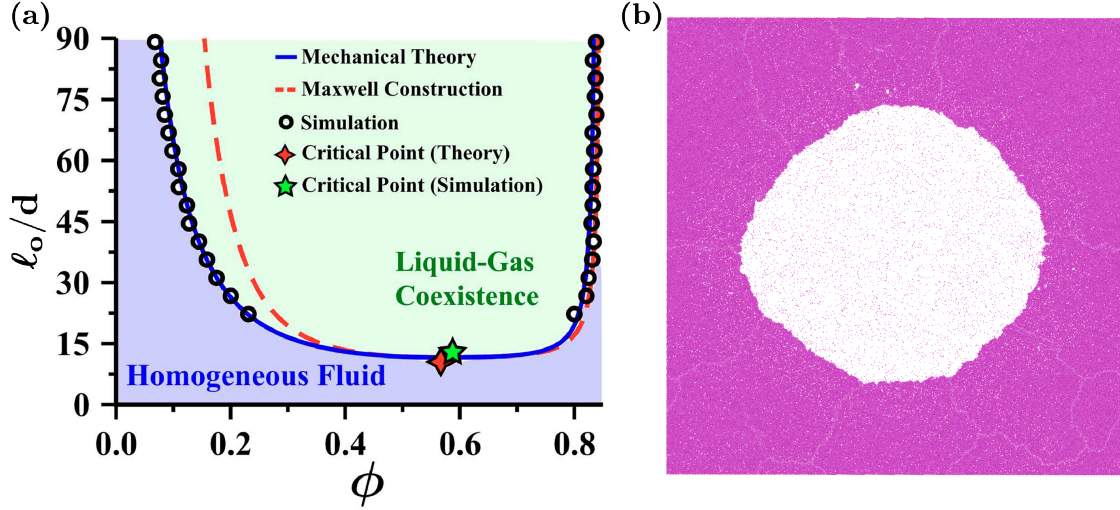


Figure 1.1: (a) Phase diagram of two dimensional active Brownian particles. Here ϕ is the area fraction and ℓ_o/d is the active persistence length normalized by the particle diameter, which serves as a measure of activity. Dashed red lines show the phase diagram theoretically predicted from a naïve application of the Maxwell construction, while solid blue lines show the phase diagram theoretically predicted phase diagram using the generalized construction presented by Omar *et al* [21]. Phase diagram is reproduced with permission from Ref. [21]. (b) Brownian dynamics simulation snapshot of $N = 3 \times 10^5$ two dimensional active Brownian particles with activity $\ell_o/d = 56$ and an overall area fraction $\phi_o = 0.5$.

The phase diagram is qualitatively similar to that of an equilibrium liquid-gas phase separation: A critical point and lever rule-respecting binodals are observed, with the motile persistence length ℓ_o normalized by the particle diameter d serving a similar role to the inverse of temperature. Further details on this simulated model system will be thoroughly discussed in the body of this thesis.

Clearly, the *bulk* phase behavior of active systems is becoming increasingly well-understood. The *interface* separating active phases, however, remains a topic of contention. These interfaces have been the subject of numerous surprising findings, including propagating waves [22] and odd-surface flows [23]. Most controversially, however, has been the measurement of negative surface tension despite manifestly stable interfaces [24, 25]. These measurements, which are the result of applying the formula found by Kirkwood and Buff [26] based on the stress tensor, have sparked significant debate with most arguments coming in the form of propos-

ing *new* definitions of surface tension through various physical motivations [27–30]. Despite these puzzling measurements, active interfaces are remarkably similar to those at equilibrium. The spectra of active capillary waves [25] scales with the same characteristic k^{-2} dependency (where k is the wave vector magnitude) that equilibrium interfaces scale with [31]. The nucleation of active phases proceeds with kinetics that are described remarkably well by Becker-Döring theory [32] – a perspective that requires detailed balance to be satisfied – and subsequently coarsen with the same $t^{1/3}$ rate predicted by equilibrium Lifshitz-Sylozov-Wagner theory [33].

Further unique properties of MIPS potentially implicate the interface. For example, in two dimensions – but not three – the liquid phase of MIPS has been found to be decorated with bubbles [34–36]. The presence of these bubbles has led some to label two dimensional MIPS as a new type of phase separation entirely called “bubbly phase separation” [37]. Various explanations for the origins of bubbly phase separation have been proposed ranging from self-organized criticality [38] to topological defects [35]. One proposed explanation [39], grounded in the modified Cahn-Hilliard framework active model B+ (AMB+), derives a non-equilibrium analog to surface tension called a “pseudotension” and finds that negative pseudotensions can result in dense phases decorated with bubbles. This explanation suggestively points to MIPS as an example of negative pseudotension, and we later discuss the applicability of that theory towards MIPS in great detail.

This thesis is entirely concerned with the resolution of the above puzzles regarding active interfaces. We first attempt to directly understand the stability of active interfaces by deriving a stochastic differential equation for active interfaces, effectively generalizing capillary-wave theory to active systems. We find that these results can be readily applied to active systems that display *chirality*, and that in the presence of chirality the derived interfacial dynamics assume complex terms that imply the presence of odd-surface flows. We also investigate nucleation, finding the finite-size corrections to the binodal predictions of Ref. [21] as well as a route towards calculating the effective nucleation barrier in active systems. These results on nucleation can then be extended to coarsening, finding routes to generalizing Lifshitz-Sylozov-Wagner theory to active systems. Lastly, we investigate the bubbly

phases of two dimensional active matter, finding that the formation of appreciable bubbles requires both hexatic order and the presence of finite (even vanishingly small) translational noise.

1.3 Structure of this thesis

The remainder of this thesis is divided into four chapters based on four separate publications, as well as a final chapter reserved for conclusions.

Chapter 2: Active Capillary Waves

The stability of active interfaces is investigated. A method for deriving fluctuating hydrodynamics starting from microscopic equations of motion is presented. From the fluctuating hydrodynamics, an equation of motion for the interface (to linear order) is extracted. These interfacial dynamics contain a term with the *units* of surface tension that only reduces to the mechanical Kirkwood-Buff definition of surface tension at equilibrium. The equation of motion implies a capillary-wave spectrum scaling as k^{-2} , identical to that of an equilibrium system. Large scale Brownian dynamics simulations are then employed to verify the validity of the derived theory. This Chapter thus generalizes capillary-wave theory to active systems and offers an explanation for how mechanical surface tension can be negative while interfaces remain stable and surface-area minimizing.

Chapter 3: Chiral Active Interfaces

The role of chirality in active interfaces is investigated. The methods of Chapter 2 are extended to account for systems with microscopic equations of motion that violate spatial parity, resulting in chiral dynamics. Chirality is found to raise the critical point of the phase separation, and the corresponding interfacial equation of motion is found to have imaginary terms only when chirality is present. These imaginary terms are found to result in the odd-surface flows – net fluxes confined to

the interfaces that point in a single direction determined by chirality. The existence of these flows are then verified through Brownian dynamics simulations of chiral active particles.

Chapter 4: Active Nucleation

The nucleation and coarsening of active phases is investigated. The bulk coexistence theory of Ref [21] is extended to account for finite size (i.e. interfacial curvature). Corrections to the bulk binodals due to finite size are derived, generalizing the Gibbs-Thomson effect to active systems. The fluctuating hydrodynamics of Chapter 2 are then applied to nucleation, and the size dynamics of a nucleus are extracted allowing for the identification of a nucleation barrier. These results are then applied to solve for the coarsening dynamics, finding both a non-equilibrium analog to Lifshitz-Sylozov-Wagner theory and also generalizing the numerical algorithm for coarsening developed by Voorhees and Glicksman [40, 41] to active systems.

Chapter 4: Thermal Noise, Hexatic Order, and Bubbles

The bubbly phases of two dimensional active matter are investigated through Brownian dynamics simulation. Dependence of the bubble population and distribution on hexatic effects are systematically probed by introducing polydispersity. Dependence of the bubbles on translational noise is also investigated. Shockingly, it is found that appreciable bubble formation only occurs for monodisperse systems with *any finite* (even vanishingly small) magnitude of translational noise.

Chapter 6: Conclusions and Outlook

The contents of the preceeding chapters are summarized. Additionally, a perspective synthesizing the results of this thesis with other work in the literature is offered to guide future work.

Chapter 2

Active Capillary Waves

Part of this chapter has been published in or included in:

L. Langford and A. K. Omar, *Phys. Rev. E* 110, 054604 (2024) [[42](#)]

2.1 Introduction

The interface associated with motility-induced phase separation (MIPS) – the phenomenon in which purely repulsive particles phase separate at sufficient activity – has been crucial in attempts to understand nonequilibrium interfaces. Efforts to understand active interfaces have focused on the surface tension, γ , the central material property for describing *equilibrium* fluid interfaces. Indeed, the equilibrium surface tension controls capillary fluctuations [31, 43], the Laplace pressure and shape of finite-sized fluid droplets [44], as well as the nucleation barrier of droplets [45]. Kirkwood and Buff [26] provided the mechanical definition of surface tension using reversible work arguments – a stress-based definition which may appear to be applicable to systems out of equilibrium. However, when applying this definition to an active interface, Bialké *et al.* [24] measured a strongly *negative* surface tension. Subsequent attempts to alternatively define the surface tension were found to reduce the magnitude of the measured tension with the sign remaining negative [29] or relied on equilibrium relations (such as the Gibbs-Duhem relation) that are strictly inapplicable to active systems [28].

Despite the ambiguity in defining the surface tension, the MIPS interface displays scaling that is remarkably similar to equilibrium capillary-wave theory (CWT) [24], with the interfacial height fluctuations of phase-separated active Brownian particles (ABPs) scaling as $\langle |h(k)|^2 \rangle \sim k^{-2}$ at low wavevector magnitudes k [25]. In equilibrium, these height fluctuations are proportional to the inverse interfacial stiffness, $\gamma/k_B T$: thermal energy acts to excite interfacial fluctuations and surface tension acts as a restoring force [31]. The observed consistency of active interfaces [24, 25, 46] with CWT raises a number of intriguing questions: what is the theoretical foundation for this k^{-2} scaling?; what plays the role of thermal energy in exciting an athermal active interface?; and, crucially, what is the definition and nature of the surface tension that attenuates these fluctuations?

Recent progress has been made towards the theory of nonequilibrium interfacial fluctuations. Lee [27] proposed an interfacial equation-of-motion using kinetic theory but neglected the effects of mass conservation. Nardini and colleagues [39, 47, 48], building on the work of Bray [49], made this approach more concrete

by linking the stochastic density field dynamics of the continuum active model B+ (AMB+) to those of the interfacial height field [48]. The stationary height fluctuation spectra that emerged from their equation-of-motion was in agreement with CWT. This perspective for deriving interfacial height field dynamics *begins* from the stochastic dynamics of a coarse-grained density field (models B [50] and AMB+ in Refs. [49] and [48], respectively) and knowledge of the averaged density profile of the macroscopically phase separated state [3, 39, 51]. Importantly, while obtaining closed-form expressions for the fluctuating hydrodynamics of microscopic models of active matter remains an outstanding challenging, the theoretical description of the phase-separated state of active Brownian spheres, including the average density profile, was recently presented in Ref. [21]. We are now poised to begin developing a physical understanding of how *microscopic* forces contribute to nonequilibrium interfacial dynamics.

In this Chapter, we present a fluctuating hydrodynamic description of interacting active Brownian particles (ABPs) that was derived beginning from microscopic considerations. This derivation is summarized in Appendix A.1. The fluctuating hydrodynamics are then used to solve for the dynamics of the interfacial height field, allowing us to identify the *capillary-wave tension* (γ_{cw}) of MIPS and the *entirely athermal* energy scale ($k_B T^{\text{act}}$) that excites the interface. Crucially, we show that this surface tension *is distinct* from the mechanical surface tension and includes effects beyond the interparticle forces that determine equilibrium interfacial tensions, including nematic surface flows that act to stabilize the interface against active fluctuations. These contributions are reminiscent of the physical picture proposed by Edwards and Wilkinson [52], in which a surface tension emerges from preferential accumulation of granular particles at concave surfaces over convex surfaces. Our derived height field dynamics are found to recover area-minimizing Boltzmann statistics, i.e., $P[h(x)] \sim \exp \left[-(\gamma_{cw}/k_B T^{\text{act}}) \int d\mathbf{x} |\nabla h|^2 \right]$ in the limit of long wavelengths despite the microscopic dynamics remaining far from equilibrium. It is precisely in this limit of area-minimizing dynamics that CWT is recovered.

2.2 Results and discussion

Fluctuating Hydrodynamics of ABPs

Our aim is to systematically derive the interfacial dynamics of active phase separations beginning from microscopic considerations. We consider a system of N interacting ABPs in which the time variation of the position $\mathbf{r}_i$ and orientation $\mathbf{q}_i$ of the i th particle follow overdamped Langevin equations:

$$\dot{\mathbf{r}}_i = U_o \mathbf{q}_i + \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij}, \quad (2.1a)$$

$$\dot{\mathbf{q}}_i = \mathbf{q}_i \times \boldsymbol{\Omega}_i, \quad (2.1b)$$

where $\mathbf{F}_{ij}$ is the (pairwise) interparticle force, U_o is the intrinsic active speed, ζ is the translational drag coefficient, and $\boldsymbol{\Omega}_i$ is a stochastic angular velocity with zero mean and variance $\langle \boldsymbol{\Omega}_i(t) \boldsymbol{\Omega}_j(t') \rangle = 2D_R \delta_{ij} \delta(t - t') \mathbf{I}$ (where $\mathbf{I}$ is the identity tensor). Here, D_R is the rotational diffusivity, which may be athermal in origin, and is inversely related to the orientational relaxation time $\tau_R \equiv D_R^{-1}$. One can then define an intrinsic run length, $\ell_o = U_o \tau_R$, the typical distance an ideal particle travels before reorienting.

We seek to describe the dynamics of an instantaneous interface that is defined by the coarse-grained density field, $\rho(\mathbf{r}, t) = \sum_{i=1}^N \Delta(\mathbf{r} - \mathbf{r}_i(t))$ where Δ is a kernel of finite spatial width such that ρ is spatially continuous. Beginning from Eqs. (2.1a) and (2.1b), we derive a fluctuating hydrodynamic [53] description of the coarse-grained density of ABPs, as detailed in Appendix A.1. The evolution equation for the density satisfies the continuity equation $\partial \rho / \partial t = -\nabla \cdot \mathbf{J}$ where the density flux, $\mathbf{J} = (\nabla \cdot \boldsymbol{\sigma}^C + \zeta U_o \mathbf{m}) / \zeta$, contains a diffusive contribution from the interparticle conservative stress, $\boldsymbol{\sigma}^C$, and a term proportional to the polar order density, $\mathbf{m}(\mathbf{r}, t) = \sum_{i=1}^N \mathbf{q}_i(t) \Delta(\mathbf{r} - \mathbf{r}_i(t))$.

An evolution equation for $\mathbf{m}(\mathbf{r}, t)$ and all other one-body orientational fields is required for a full spatial and temporal description of the density field. However, the inclusion of higher order orientational fields simply acts to increase the temporal

and spatial resolution [54]. To describe MIPS, inclusion of the traceless nematic order density $\mathbf{Q}' = \mathbf{Q} - \rho/d\mathbf{I}$ [where $\mathbf{Q}(\mathbf{r}, t) = \sum_{i=1}^N \mathbf{q}_i(t)\mathbf{q}_i(t)\Delta(\mathbf{r} - \mathbf{r}_i(t))$], is necessary [21]. We therefore close the hierarchy of equations by assuming $\mathbf{B}(\mathbf{r}, t) = \sum_{i=1}^N \mathbf{q}_i(t)\mathbf{q}_i(t)\mathbf{q}_i(t)\Delta(\mathbf{r} - \mathbf{r}_i(t))$ is isotropic [21]. We further note that the polar and nematic order relax on faster timescales than the density field, allowing us to neglect their time variation and assume quasi-stationary density relaxation. Under these closures, the *deterministic* component of the polarization force density takes the form $\frac{\xi\ell_o}{d-1}\nabla \cdot (U\mathbf{Q}) \equiv \nabla \cdot \boldsymbol{\sigma}^{\text{act}}$, where U is the effective speed of the particles (reduced from U_o due to interparticle interactions) and $\boldsymbol{\sigma}^{\text{act}}$ is an active *effective* stress [17, 29, 55, 56]. The nematic field can be directly related to the density field, allowing for a closed set of equations solely in terms of the density with:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \mathbf{J}, \quad (2.2a)$$

$$\mathbf{J} = \frac{1}{\xi} \nabla \cdot (\boldsymbol{\sigma}^C + \boldsymbol{\sigma}^{\text{act}}) + \boldsymbol{\eta}^{\text{act}}, \quad (2.2b)$$

$$\begin{aligned} \boldsymbol{\sigma}^C = & \left[-p_C(\rho) + \kappa_1(\rho)\nabla^2\rho + \kappa_2(\rho)|\nabla\rho|^2 \right] \mathbf{I} \\ & + \kappa_3(\rho)\nabla\rho\nabla\rho + \kappa_4(\rho)\nabla\nabla\rho, \end{aligned} \quad (2.2c)$$

$$\boldsymbol{\sigma}^{\text{act}} = \left[-\frac{\ell_o U_o \xi \bar{U} \rho}{d(d-1)} + a(\rho)\nabla^2\rho \right] \mathbf{I} + b(\rho)\nabla\rho\nabla\rho, \quad (2.2d)$$

$$a(\rho) = \frac{3\ell_o^2}{2d(d-1)(d+2)} \bar{U}^2 \frac{dp_C}{d\rho}, \quad (2.2e)$$

$$b(\rho) = \frac{3\ell_o^2 \bar{U}}{2d(d-1)(d+2)} \frac{d}{d\rho} \left[\bar{U} \frac{dp_C}{d\rho} \right], \quad (2.2f)$$

where $p_C(\rho)$ is the conservative interaction pressure [21], $\bar{U}(\rho) \equiv U(\rho)/U_o$ is the dimensionless effective speed [21], $d(> 1)$ is the system dimensionality. Here $\{\kappa_i(\rho)\}$ are expansion coefficients of the conservative stress¹ in the reversible limit as first derived by Korteweg [43, 44, 57]. We note that the derivation of Eq. (2.2)

¹In equilibrium, $\kappa_1(\rho) = \rho\kappa(\rho)$, $\kappa_2(\rho) = (\kappa(\rho) + \rho\kappa'(\rho)/2)$, $\kappa_3(\rho) = -\kappa(\rho)$, $\kappa_4(\rho) = 0$, where $\kappa(\rho)$ is related to a moment of the direct correlation function and pairwise interaction force [43].

made use of a quasi-stationary approximation (i.e., $|\mathbf{J}|$ is small) which is valid for dynamics near the flux-free states (e.g., stationary phase coexistence) under consideration in this work.

Equation (2.2) introduces an entirely athermal stochastic contribution to the flux, $\boldsymbol{\eta}^{\text{act}}$, with zero mean and a variance of:

$$\langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \boldsymbol{\eta}^{\text{act}}(\mathbf{r}', t') \rangle = 2 \frac{k_B T^{\text{act}}}{\zeta} \left(\rho \mathbf{I} - \frac{d}{d-1} \mathbf{Q}' \right) \delta(t - t') \delta(\mathbf{r} - \mathbf{r}') , \quad (2.3)$$

where $k_B T^{\text{act}} \equiv \ell_o \zeta U_o / d(d-1)$ is the active energy scale. We reemphasize that, within our closure, the traceless nematic field $\mathbf{Q}'$ can be expressed fully in terms of the density field. To arrive at these noise statistics we *approximate* the coarse-graining kernel as $\Delta(\mathbf{r} - \mathbf{r}_i(t)) \approx \delta(\mathbf{r} - \mathbf{r}_i(t))$ [39, 58–60], allowing us to make use of the properties of delta functions [53] and express Eq. (2.3) solely in terms of one-body fields, as shown in Appendix A.1. We emphasize that this approximation was only necessary for finding the noise variance. These statistics are similar to those found by Cugliandolo *et al.* [61] in the context of passive dipolar particles². Unlike the fluctuating hydrodynamic description of passive systems, the noise derived here is anisotropic due to the presence of the nematic order. In a system with no spontaneous nematic order (i.e., $\mathbf{Q}' = \mathbf{0}$), the noise reduces to a form identical to that of a passive system [53] with $k_B T^{\text{act}}$ playing the same role as thermal energy. However, even in this scenario where the fluctuation-dissipation theorem is satisfied, the dynamics described in Eq. 2.2 remain distinctly active as the nematic flows encapsulated in $\boldsymbol{\sigma}^{\text{act}}$ cannot be derived from a variational principle. As expected, *deterministic* terms in Eq. (2.2) are identical to those of Ref. [21]. However, the route to deriving an expression for the density evolution which preserves the *stochastic* fluctuations is distinct from the route used in Ref. [21] which averaged over all noise and focused on stationary states.

²We are not aware of the presence of Eq. (2.3) in the *active* literature, likely due to the emphasis on 2d systems with orientations restricted to be in the plane [62].

System Details and Molecular Dynamics Simulations

The above fluctuating hydrodynamics may be applied to any system with particles obeying Eq. (2.1) – pairwise interactions of different forms will simply alter the constitutive relations present in Eq. (2.2). Here, we focus on three dimensional (3d) active Brownian hard spheres as: (i) the order parameter distinguishing the two coexisting phases is simply the volume fraction; (ii) the phase diagram is well characterized [63] and entirely described by two dimensionless geometric parameters, the run length ℓ_o/d_{hs} (where d_{hs} is the hard-sphere diameter) and volume fraction; and (iii) expressions for the required constitutive relations have been recently developed [21]. Furthermore, for active hard spheres, MIPS occurs at an appreciable level of activity [$\ell_o/d_{\text{hs}} \geq O(10)$] resulting in $\{\kappa_i(\rho)\} \ll a(\rho), b(\rho)$ [21] and allowing us to neglect the interaction interfacial stresses present in Eq. (2.2c). This approximation has recently been justified numerically through multiple approaches Refs. [29, 64]. We conduct particle-based Brownian dynamics simulations [65] at activities and volume fractions within the MIPS binodal, with simulation details in Appendix A.4. The chosen simulation geometry results in a planar interface with its normal (on average) in the z -direction [63]. The remaining coordinates are tangent to the interface and are denoted by the vector $\mathbf{x}$, i.e., $\mathbf{r} \rightarrow (\mathbf{x}, z)$. The height field $h(\mathbf{x})$ associated with the instantaneous interface [66] is defined as the location in space where ρ is halfway between the binodal densities [see Fig. 2.1(a)], i.e., $\rho(\mathbf{x}, z = h) = (\rho^{\text{liq}} + \rho^{\text{gas}})/2 = \rho^{\text{surf}}$. The discrete Fourier transform ($\mathbf{x} \rightarrow \mathbf{k}$) of $h(\mathbf{x})$ is denoted by $\tilde{h}(\mathbf{k})$.

The computational cost for observing the relaxation of capillary-waves in molecular dynamics associated with k scales proportionally to the number of particles in the system ($1/k^d$) multiplied by the timescale for the relaxation of the capillary wave (which we will later demonstrate is $1/k^3$), limiting the numerical accessibility of the mesoscopic limit where CWT might apply. We therefore conduct, to our knowledge, the largest scale simulation of 3d ABPs currently in the literature, with 631444 particles for a duration of $89000 d_{\text{hs}}/U_o$ [65]. The calculated fluctuation spectrum is shown in Fig. 2.1(b). Power law fits (i.e., $\langle |h(\mathbf{k})|^2 \rangle = K_s k^\omega$) to the first decade of wavevectors reveal that the height fluctuations approach a k^{-2}

scaling in the low- k limit [see Fig. 2.1(c)], consistent with the 2d data of Patch *et al.* [25] and corroborating the apparent applicability of CWT to these driven interfaces. These fits also demonstrate the interfacial stiffness, K_s^{-1} [see Fig. 2.1(c)], scales linearly with run length, again consistent with 2d simulations [25].

Fluctuations of MIPS interfaces

In order to connect the dynamics of the density field to those of an interfacial height field we introduce the ansatz proposed by Bray *et al.* [49], which states that the instantaneous stochastic density field is, within a displacement of the interfacial height field h in the z -dimension, equal to the noise-averaged stationary density field, φ :

$$\rho(\mathbf{r}, t) = \varphi [z - h(\mathbf{x}, t)] . \quad (2.4)$$

This ansatz implies that $|\partial h / \partial \mathbf{x}|^2 \ll 1$ [49]; it is equivalent to stating the normal vector to contours of ρ negligibly deviate from the z -direction³.

As recently demonstrated by Fausti *et al.* [48], substitution of the ansatz [Eq. (2.4)] into the continuity equation [Eq. (2.2)] results in nonlinear terms which coincide with those found in the quasi-1D flux balance used to derive the generalized Maxwell construction pseudovvariable needed to calculate the binodals [20, 21, 71]. The use of the pseudovvariable allows us to incorporate the effects of nonlinear terms in the fluctuating hydrodynamics (physically interpreted as nematic flows, as discussed in Ref. [42]) to the interfacial tension (linear term in the height field dynamics). This pseudovvariable, $\mathcal{E}$, satisfies the differential equation $a \partial^2 \mathcal{E} / \partial \rho^2 = (2b - \partial a / \partial \rho) \partial \mathcal{E} / \partial \rho$ and is found to be $\mathcal{E} \sim p_C$ for the present system [21].

The ansatz [Eq. (2.4)] coupled with our fluctuating hydrodynamic theory allow for the determination of a *linear* equation-of-motion for $h(k)$, the continuous

³Note that the stochastic density evolution equation has been derived using Itô's discretization scheme. However, we proceed as if it could be interpreted in the Stratonovich sense with no modification. In applying Bray's ansatz, we switch to a Stratonovich scheme and neglect the spurious drift term, which has been shown to vanish when considering conserved order parameters with a stochastic flux [68–70].

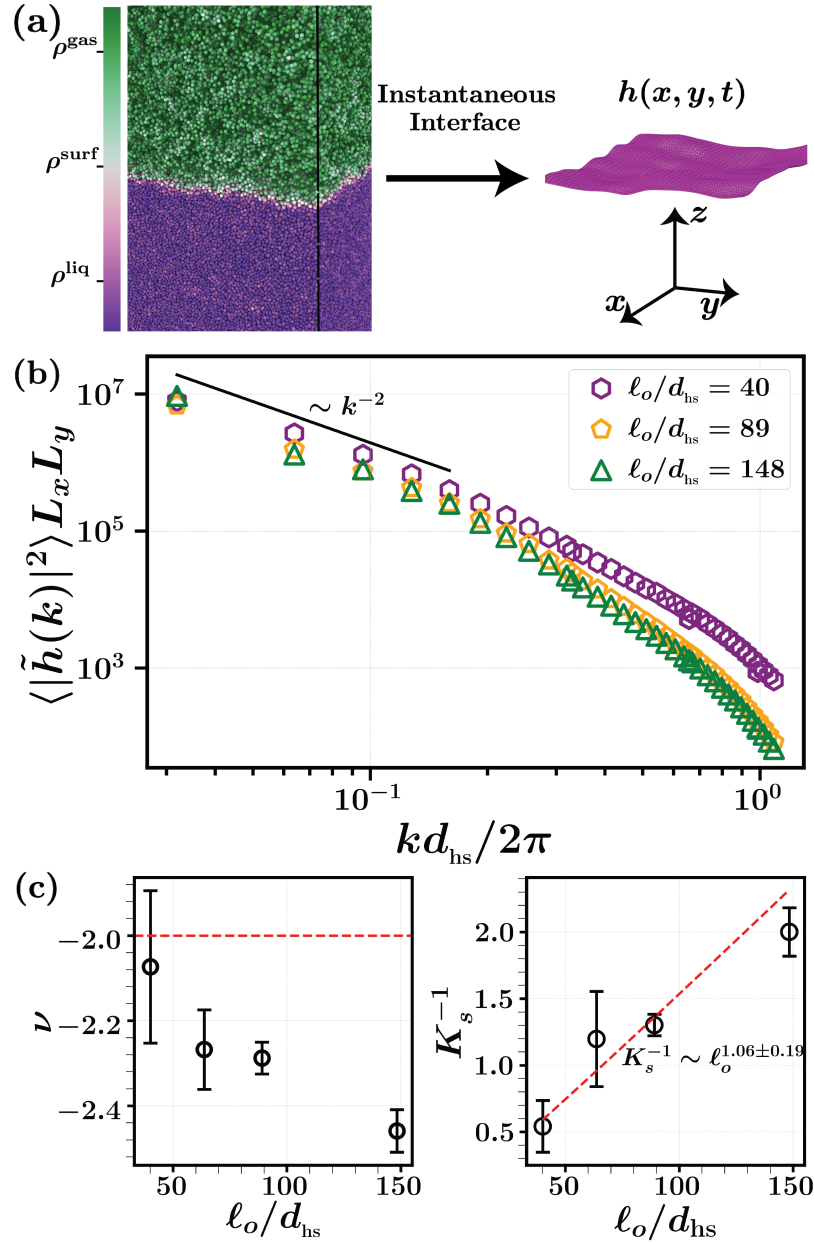


Figure 2.1: (a) Visualization [67] of instantaneous interface construction obtained through Brownian dynamics. (b) Average interfacial fluctuation spectrum (units of d_{hs}^4). (c) Values of ω (dimensionless) and K_s^{-1} (units of d_{hs}^{-2}) obtained from power law fits $\langle |\tilde{h}(\mathbf{k})|^2 \rangle = K_s k^\omega$.

Fourier transform of $h(\mathbf{x})$. The resulting Langevin equation (see Appendix A.2 for a derivation) for $h(k)$ is:

$$\zeta_{\text{eff}} \frac{\partial h}{\partial t} = -k^3 \gamma_{\text{cw}} h + \mathcal{X}^{\text{iso}} + \mathcal{X}^{\text{aniso}}, \quad (2.5a)$$

$$\zeta_{\text{eff}}(k) = \frac{\zeta A^2(k)}{2B(k)\rho^{\text{surf}}}, \quad (2.5b)$$

$$\gamma_{\text{cw}}(k) = \frac{A(k)}{\rho^{\text{surf}} B(k)} \left[\int_{-\infty}^{\infty} du \frac{\partial \mathcal{E}}{\partial u} \frac{\partial \varphi}{\partial u} a(\varphi) + \int_{-\infty}^{\infty} du \frac{\partial \mathcal{E}}{\partial u} \int_{-\infty}^{\infty} dz' \text{sgn}(u - z') e^{-k|u-z'|} b(\varphi) \left(\frac{\partial \varphi}{\partial z'} \right)^2 \right], \quad (2.5c)$$

where the integration coordinate is defined as $u = z - h$. γ_{cw} is the capillary-wave tension and has physical dimensions of energy per unit area (length) in 3d (2d). ζ_{eff} is an effective drag coefficient, and $\mathcal{X}^{\text{iso}}(\mathbf{k}, t)$ and $\mathcal{X}^{\text{aniso}}(\mathbf{k}, t)$ are independent stochastic forces with zero mean that, respectively, originate from the isotropic and anisotropic contributions to $\boldsymbol{\eta}^{\text{act}}$. The variance of $\mathcal{X}^{\text{iso}}$ is:

$$\langle \mathcal{X}^{\text{iso}}(\mathbf{k}, t) \mathcal{X}^{\text{iso}}(\mathbf{k}', t') \rangle = 2k(k_B T^{\text{act}}) \zeta_{\text{eff}} \delta(t - t') \delta(\mathbf{k} + \mathbf{k}') (2\pi)^{(d-1)}, \quad (2.6)$$

while the variance of $\mathcal{X}^{\text{aniso}}$ and integral expressions for $A(k)$, $B(k)$ (which solely depend on φ and $\mathcal{E}$) are provided in Appendix A.2.

The derived capillary-wave tension $\gamma_{\text{cw}}(k)$ is distinct from the mechanical surface tension, as shown in Appendix A.2. For active hard spheres, the active force and diameter must set the scale of the surface tension with $\gamma_{\text{cw}} \sim \zeta U_o / d_{\text{hs}}$ while the run length dependence is nontrivial. Defining a reduced run length, $\lambda \equiv \ell_o / \ell_o^{\text{crit}} - 1$ (where $\lambda > 0$ results in the phase-separated states of interest), the surface tension has the form $\gamma_{\text{cw}} d_{\text{hs}} / \zeta U_o = g(\lambda, k)$ where $g(\lambda, k)$ is a dimensionless function that must approach zero as the critical point is approached (i.e., $\lambda \rightarrow 0$). In this limit of vanishing capillary tension, the characteristic timescale that naturally emerges from Eq. (2.5a), $\tau(k) \equiv \zeta_{\text{eff}} / \gamma_{\text{cw}} k^3 \rightarrow \infty$. These increasingly long timescales with decreasing λ are consistent with the increased statistical uncertainty numerically observed at low activities in Fig. 2.1. We note that the reduction of γ_{cw} with

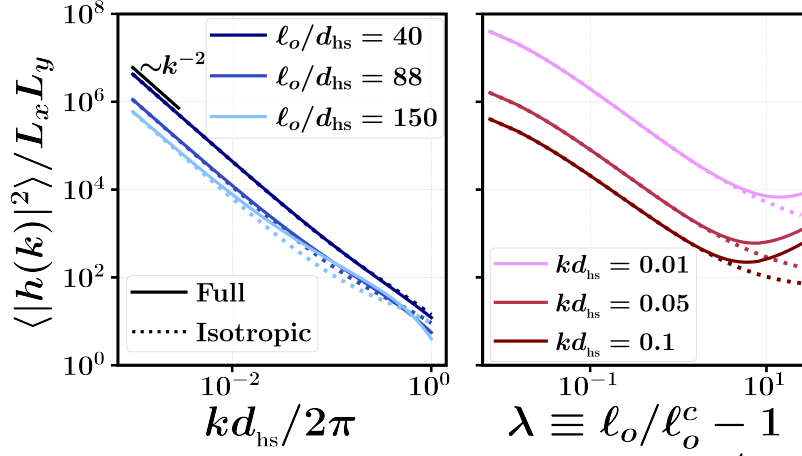


Figure 2.2: Stationary fluctuations of the interfacial height field (units of d_{hs}^4) as given by Eq. (2.7) (solid lines) and Eq. (2.8) (dotted lines). The critical activity is $\ell_o^c/d_{hs} \approx 16.2$. The anisotropic terms of Eq. (2.7) result in a non-monotonic dependence of the stationary fluctuations with activity.

decreasing λ implies a vanishing stabilizing force (with $\gamma_{cw} > 0$) in Eq. (2.5a) as the critical point is approached – in this regime, the assumptions underlying the ansatz [Eq. (2.4)] no longer hold.

To determine the natural energy scale of γ_{cw} we use Eq. (2.5a) to determine the stationary fluctuations of the height field with:

$$\langle |h(k)|^2 \rangle = \frac{L^{(d-1)}}{\gamma_{cw}} \left[\frac{k_B T^{\text{act}}}{k^2} + \frac{C(k) + D(k)}{2k^3(d-1)\rho^{\text{surf}} B(k)} \right], \quad (2.7)$$

where $L^{(d-1)}$ is the projected area (length) of the interface for 3d (2d) systems. The first term in Eq. (2.7) arises entirely due to $\mathcal{X}^{\text{iso}}$ and is consistent with CWT while the second term emerges from $\mathcal{X}^{\text{aniso}}$ ($C(k)$ and $D(k)$ are provided in Appendix A.2). In the low- k limit, the scaling of the second term of Eq. (2.7) with k can be identified as $1/k$ by inspecting the forms of $C(k)$ and $D(k)$ provided in Appendix A.2. Using the equations-of-state of 3d ABPs [21] and solving for φ , we calculate the fluctuations with and without anisotropic contributions, as shown in Fig. 2.2. While the anisotropic contributions can result in a non-monotonic dependence of the height fluctuations with activity at finite wavelengths, these contributions are negligible at wavelengths much larger than the run length,

i.e., $\ell_o k \ll 1$, and at low activities. As a result, CWT is precisely recovered at low k with:

$$\langle |h(k)|^2 \rangle = \frac{L^{(d-1)} k_B T^{\text{act}}}{\gamma_{\text{cw}} k^2} + \mathcal{O}\left(\frac{1}{k}\right). \quad (2.8)$$

While in this large wavelength limit, the variance of the height fluctuations are consistent with CWT, it can be shown that *all statistical moments* of $h(k)$ will have the same form of those of an equilibrium interface. We demonstrate this by solving the steady-state Fokker-Planck equation associated with Eq. (2.5a) in Appendix A.3, finding a Boltzmann distribution of interfacial shapes $P[h(\mathbf{x})] \propto \exp\left[-(\gamma_{\text{cw}}/k_B T^{\text{act}}) \int d\mathbf{x} |\nabla h|^2\right]$. This distribution implies that at long wavelengths our interfacial dynamics can be exactly mapped onto that of an equilibrium system [49] with an interfacial energy given by the product of γ_{cw} with the surface area and a temperature of $k_B T^{\text{act}}$. Thus, when the anisotropic fluctuations can be ignored, *an area-minimizing principle is recovered for the MIPS interface*, despite the nonequilibrium origins of the interfacial tension and athermal excitations.

From Eq. (2.8) it is clear that interfacial fluctuations are excited by the athermal active energy scale and suppressed by the capillary-wave tension: the active interfacial stiffness is $\gamma_{\text{cw}}/k_B T^{\text{act}}$. We numerically evaluate this stiffness, shown in Fig. 2.3. The capillary-wave tension is positive for all activities and increases towards a constant in the macroscopic (i.e., $k \rightarrow 0$) limit. In the limit of large activity, we find that the interfacial stiffness scales linearly with run length, consistent with the observed stiffness scaling reported by Patch *et al.* [25]. Our theory now allows us to determine the origin of this scaling by identifying the precise form of γ_{cw} [Eq. (2.5c)] and the interfacial stiffness. While the active energy scale has a trivial scaling with reduced activity, $k_B T^{\text{act}} \sim \lambda$, the scaling of the capillary-wave tension with activity requires examining the λ dependence of the density profile (ϕ), interfacial coefficients (a and b), and the pseudovisible ($\mathcal{E}$). For the present system, $\mathcal{E}$ is independent of activity while the density profile is also largely independent of activity at large λ . The latter can be appreciated by considering the relatively constant binodal densities of the MIPS phase diagram at large λ [21, 63].

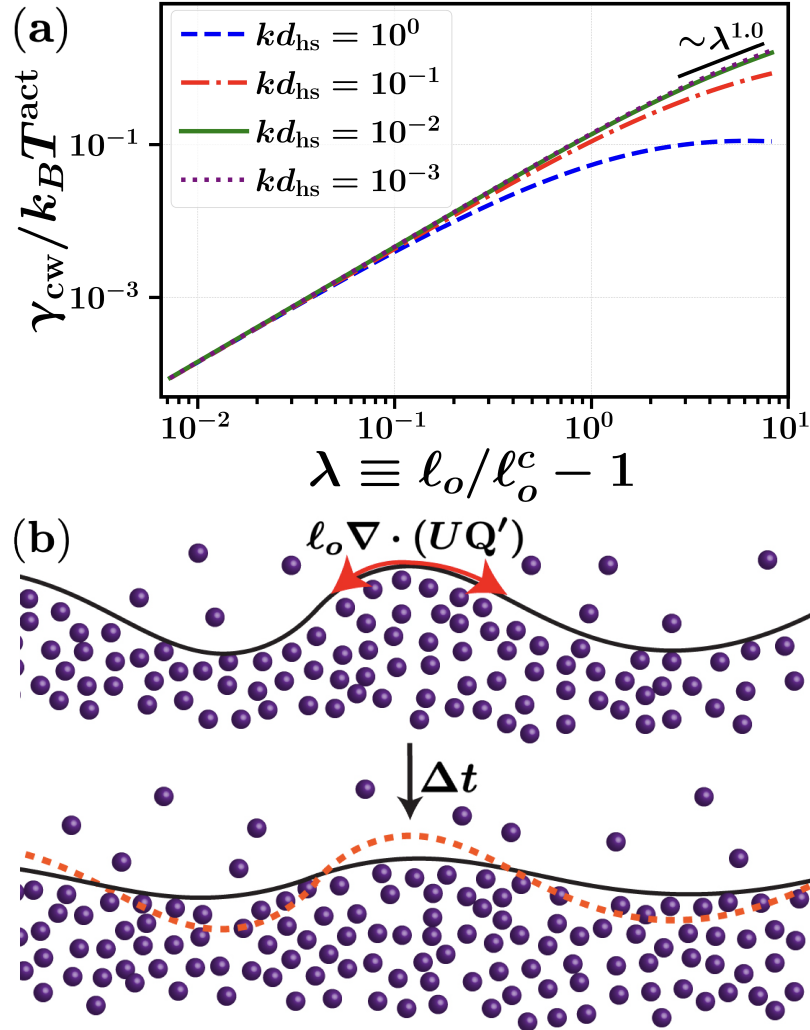


Figure 2.3: (a) Interfacial stiffness (units of d_{hs}^{-2}) as a function of the reduced activity, λ , and k , calculated using Eq. (2.5c). (b) Schematic demonstrating the impact of tangential nematic flows that are encapsulated in γ_{cw} . Following time evolution $\Delta t \sim \tau(k) \equiv \zeta_{\text{eff}}/k^3 \gamma_{\text{cw}}$, nematic flows lead to particle escape from convex hills and accumulation at convex troughs.

The asymptotic activity dependence of γ_{cw} thus closely follows that of the active interfacial stress coefficients, a and b , which generate nematic flows in response to density gradients and scale as λ^2 . The physical origin for the linear dependence of the stiffness on activity is therefore a result of stabilizing nematic flows competing with the active energy scale. Fig. 2.3(b) provides a schematic of nematic flows which result in particles escaping from hills and accumulating in troughs.

While we have focused on the stationary properties of the interface, our theory also makes predictions for dynamic properties. Starting from Eq. (2.5a), we solve for the interfacial power spectra $\langle |h(k, \omega)|^2 \rangle$:

$$\langle |h(k, \omega)|^2 \rangle = \frac{2kL^{d-1}\Gamma k_B T^{\text{act}}}{\zeta_{\text{eff}}(\omega^2 + \tau(k)^{-2})}, \quad (2.9)$$

where Γ is the total simulation time. We note that Eq. (2.9) assumes values of k such that anisotropic fluctuations are negligible (i.e., $\ell_o k \ll 1$). At these low values of k and in the low frequency limit (i.e., $\omega \rightarrow 0$), we predict the power spectra to scale as k^{-5} . Figure 2.4 displays the power spectra measured from Brownian dynamics simulation. We note that despite the extensive size and duration of our computer simulations, our analysis is limited to less than a single decade of k values, and thus a robust power-law fit cannot be established. Nevertheless, it is encouraging that the fit obtained approaches the theoretically predicted scaling of k^{-5} with increasing activity, agreeing well with this prediction at the highest activity. A potential source of error for the low run length dynamics are nonlinear corrections [72, 73] to Eq. (2.5a), neglected in the present study through use of Eq. (2.4). These nonlinear corrections are anticipated to be more important closer to the critical point due to the larger magnitude of interfacial fluctuations [see Fig. 2.1(c)]. At higher activities, error may also come from the significance of anisotropic noise as the identified parameter $k\ell_o$ grows. These competing dependencies of our theory's error may offer an explanation for the non-monotonic behavior of the inset of Fig. 2.4. We hope that this finding will encourage additional numerical studies of the dynamics of active interfaces.

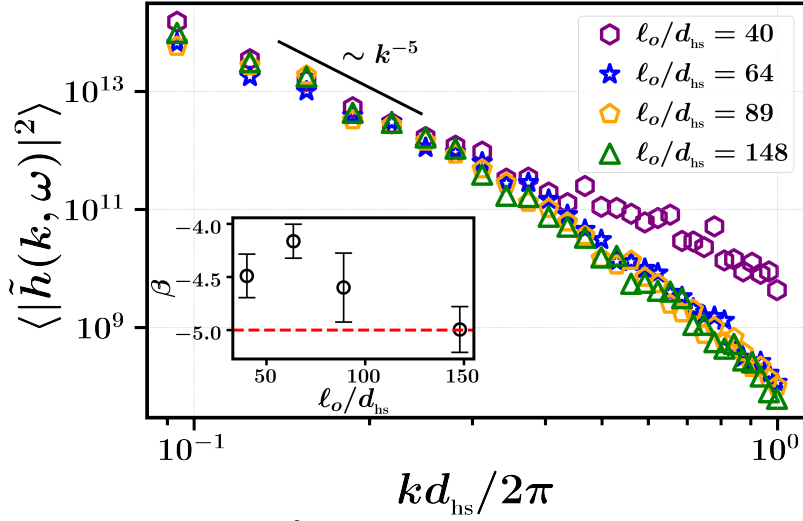


Figure 2.4: Power spectra $\langle |h(k, \omega)|^2 \rangle$ at each simulated activity as a function of k with $\omega d_{hs}/2\pi U_o = 2.82 \times 10^{-4}$ fixed. (Inset) Power law scaling obtained from logarithmic fits to $\langle |h(k, \omega)|^2 \rangle \sim k^\beta$ for low values of k (i.e., $k d_{hs}/2\pi < 0.5$).

2.3 Conclusions

Starting from microscopic particle dynamics, we systematically derived a mesoscopic Langevin equation for the interface of MIPS in this Chapter. From this equation-of-motion, we can identify the capillary-wave tension governing the dynamics of the interface and identify the athermal source of interfacial excitations. Interestingly, the derived energy scale for interfacial excitations is consistent with predictions made utilizing stochastic thermodynamics [24, 74]. Our capillary-wave tension encapsulates the effects of active nematic flows on the height field dynamics and is rooted in mass transport generated by nonconservative forces. The origins of this tension are similar to the physical picture proposed by Edwards and Wilkinson [52], in which surface tension emerged from deposited particles preferentially settling in local minima, and perhaps points to a general phenomena of dynamically-stabilized interfaces. Our interfacial Langevin equation recovers a surface-area minimizing Boltzmann distribution at low activities and low wavevectors, despite the microscopic particle dynamics breaking time-reversal symmetry. In this limit, an active analog to CWT is recovered with an interfacial stiffness that

scales linearly with run length.

The fluctuating hydrodynamics derived in this Chapter may be applied to understand other nonequilibrium phenomena requiring a stochastic description, including active nucleation [75–78], which we cover in Chapter 4. By deriving interfacial dynamics beginning from microscopic equations of motion, the procedure outline here can also provide a way forward for examining the interfacial dynamics of chiral active matter, which can exhibit odd surface flows [23]. This path for future work is presented in Chapter 3. This work might also be extended to include multiple order parameters, shedding light on phenomena such as active crystallization [63] or multicomponent driven systems [46, 79–82], such as biomolecular condensates [7, 8, 83]. Finally, while our derived interfacial dynamics are linear, the inclusion of nonlinear contributions could allow for the determination of the universality class of the MIPS interface [72, 73, 84].

Chapter 3

Chiral Active Interfaces

Part of this chapter has been published in or included in:
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3.1 Introduction

Active phase separation, a phenomenon displayed by a broad array of systems ranging from the internals of a living cell [7] to self-catalytic colloidal surfers [10] and motile bacteria [16], has been a subject of considerable recent interest in statistical physics, biology, and soft matter. Both the phase behavior and interface dividing the coexisting phases has been subject of many intriguing findings ranging from odd surface flows [23] to propagating waves [22]. In particular, the interface dividing motility induced phase separations (MIPS) [15] has been associated with a *negative* mechanical surface tension [24], which has sparked much controversy [28, 29, 86]. Considerable recent progress has been made towards the derivation of physically relevant surface tensions in phase separated active systems [25, 27, 42, 48]. Despite this progress, much of the theoretical insights have tacitly *assumed dynamics respecting spatial parity* – a symmetry that many active systems violate. Whether arising from externally applied magnetic fields [23], core biological behavior [87], or a helical nanomotor geometry [88], these systems are inherently *chiral*. While the theory surrounding *odd transport* phenomena that generically arises from chiral active systems is increasingly well-established [89–96], the *phase behavior* and *interfacial* properties of such systems has remained relatively unexplored.

In this chapter, we systematically derive the interfacial dynamics of a chiral active interface beginning from microscopic dynamics that violates *both* time reversal symmetry and spatial parity. En route to doing so, we develop a nonequilibrium mechanical description [21] of chiral MIPS, finding that chirality raises the critical motility for the onset of phase separation. Building on a number of frameworks [42, 48, 49, 97], we then derive an approximate fluctuating hydrodynamic description of interacting chiral active particles and ultimately the dynamics of a phase-separated chiral active interface. These dynamics reveal the microscopic definition for the capillary tension γ_{cw} , which governs the stability of the interface, as well as the odd flow coefficient ν_{odd} which generates travelling waves at the surface. Surprisingly, we find that the stationary statistics of the interface are controlled *exclusively* by γ_{cw} while ν_{odd} only affects dynamical correlations. Brownian dynamics simula-

tions of chiral active matter allow us to confirm these theoretical predictions and the precise activity and chirality dependencies of the observed phenomena.

3.2 Results and discussion

Model System

We consider a *two-dimensional* (2D) system of N interacting chiral active Brownian particles (cABPs) [98–101] in which the time variation of the position $\mathbf{r}_i$ and (2D) orientation $\mathbf{q}_i$ of the i th particle follow overdamped Langevin equations:

$$\dot{\mathbf{r}}_i = U_o \mathbf{q}_i + \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij}, \quad (3.1a)$$

$$\dot{\mathbf{q}}_i = \boldsymbol{\omega}_o \times \mathbf{q}_i + \boldsymbol{\Omega}_i \times \mathbf{q}_i, \quad (3.1b)$$

where $\mathbf{F}_{ij}$ is the (pairwise) interparticle force; $U_o, \boldsymbol{\omega}_o$ are the intrinsic active speed and angular velocity, respectively; ζ is the translational drag coefficient; and $\boldsymbol{\Omega}_i$ is a stochastic angular velocity. In two dimensions, the only component of $\boldsymbol{\omega}_o$ and $\boldsymbol{\Omega}_i$ relevant to the dynamics of $\mathbf{q}_i$ will be that of the out-of-plane direction, which we describe with unit vector $\mathbf{e}_z$. We can express $\boldsymbol{\omega}_o = \omega_o \mathbf{e}_z$, with the sign of ω_o determining the handedness of the chiral dynamics. We express $\boldsymbol{\Omega}_i$ as $\Omega_i \mathbf{e}_z$, where Ω_i has zero mean and variance $\langle \Omega_i(t) \Omega_j(t') \rangle = 2D_R \delta_{ij} \delta(t - t')$. Here, D_R is the rotational diffusivity, which may be athermal in origin, and is inversely related to the orientational relaxation time $\tau_R \equiv D_R^{-1}$. Translational noise is neglected as our focus is to isolate the role of chirality and activity¹. When the pairwise forces $\mathbf{F}_{ij}$ are chosen such that particles effectively exclude area with diameter d , the system is fully characterized by three dimensionless quantities: the overall area

¹The addition of translational noise, characterized by a translational diffusivity of D_T , would result in an additional dimensionless parameter which can be defined as $\mathcal{T} = D_T / (D_R d^2)$. As $\mathcal{T} \rightarrow \infty$, translational diffusivity dominates and MIPS (and other activity-generated phenomena) is anticipated to vanish. As the primary axes we aim to explore are chirality and activity, we choose to fix $\mathcal{T}$ and work in the “athermal” limit of $\mathcal{T} = 0$.

fraction $\phi \equiv \rho\pi d^2/4$ (where ρ is the number density); the dimensionless intrinsic run length, $\ell_o/d = U_o\tau_R/d$; and the ratio of chiral and rotary diffusion timescales, $\chi \equiv \omega_o\tau_R$. We note that this dimensionless measure of the chirality also has a clear geometric interpretation. The intrinsic run length ℓ_o is a measure of the persistence length of an ideal and achiral active particle trajectory while $U_o/|\omega_o|$ is the radius of the trajectory of an ideal particle in the absence of rotational diffusion. The ratio of these two length scales is simply $\ell_o|\omega_o|/U_o \equiv |\chi|$. We recover the achiral ABP model in the limit of $\chi \rightarrow 0$.

Phase Coexistence of Chiral Active Matter

Understanding the bulk phase coexistence of a system is prerequisite to understanding its interface. Here, we formulate a nonequilibrium theory for the phase diagram of cABPs following a recently proposed mechanical framework [21]. As explicitly detailed in Appendix B.1, we derive the dynamics of the density field, the relevant order parameter for fluid-fluid coexistence, beginning from the N -body distribution of microstates and Eq. (3.1). Just as for ABPs, the microscopically derived mechanical balance for cABPs consists of the conservative stress (σ^C) generated by particle interactions and an active stress (σ_{act}) induced by the active force with $\mathbf{0} = \nabla \cdot (\sigma^C + \sigma_{\text{act}})$. For repulsive achiral ABPs, the active stress is the origin of the mechanical instability that underlies MIPS [18, 102]. Chiral dynamics directly alter the active stress by generating *antisymmetric* active stresses (much like chiral active dumbbells [103]) for finite χ while also diminishing the symmetric components.

While the total dynamic stress tensor $\Sigma \equiv \sigma_{\text{act}} + \sigma^C$ is no longer symmetric for finite χ (see Appendix B.1 for our derived expressions for Σ) this proves to be inconsequential for macroscopic phase coexistence. We consider two macroscopic coexisting phases with an interface which (on average) has a normal direction parallel to $\mathbf{e}_x$ and translational invariance in the y -direction. In this scenario, only the symmetric contribution to the stress appears in the static mechanical balance which reduces to $0 = d\Sigma_{xx}/dx$. We can then invoke the recently derived mechanical

coexistence criteria [21]:

$$\mathcal{P}(\rho^{\text{liq}}) = \mathcal{P}(\rho^{\text{gas}}) \equiv \mathcal{P}^{\text{coexist}}, \quad (3.2a)$$

$$\int_{\rho^{\text{liq}}}^{\rho^{\text{gas}}} [\mathcal{P} - \mathcal{P}^{\text{coexist}}] E(\rho) d\rho = 0, \quad (3.2b)$$

where $\mathcal{P}(\rho)$ is the bulk contribution to the dynamic pressure, and $E(\rho)$ is determined from the interfacial contributions to the dynamic pressure. Equation (3.2b) takes the form of a “weighted-area” Maxwell construction [20, 21, 71].

With a precise form for $\mathcal{P}(\rho)$ and $E(\rho)$, we can use Eq. (3.2) to determine the coexisting densities, ρ^{liq} and ρ^{gas} . For cABPs we find (see Appendix B.1):

$$\mathcal{P}(\rho) = p_C(\rho) + p_{\text{act}}(\rho), \quad (3.2c)$$

$$p_{\text{act}}(\rho) = \frac{\zeta U_o \ell_o \rho \bar{U}(\rho)}{2(1 + \chi^2)}, \quad (3.2d)$$

$$E(\rho) \sim [\bar{U}(\rho)]^{-\frac{\chi^2}{4}} \left(\frac{\partial p_C}{\partial \rho} \right)^{1 - \frac{\chi^2}{4}}, \quad (3.2e)$$

where $p_C(\rho)$ and $p_{\text{act}}(\rho)$ are the bulk contributions to the conservative and active stresses, respectively. $\bar{U}(\rho)$ is the dimensionless average active speed which ranges from 0 (interactions completely arrest particle motion) to 1 (particles move unimpeded with speed U_o) which depends on the particle interactions and density. Both the dynamic pressure and weighted-area construction variable reduce to those recently found for ABPs [21] in the limit $\chi \rightarrow 0$, as anticipated. To determine the qualitative impact of chirality, we adopt forms of the equations of state that have either been previously proposed (p_C) or are similar to established functions ($\bar{U}$) for repulsive ABPs (the precise forms are provided in Appendix B.1).

The phase diagram for cABPs is shown in Fig. 2.1(a) as a function of activity for several values of χ . Increasing chirality monotonically raises the critical activity indicating that *chirality acts to stabilize active fluids*. This can be immediately understood by examining the form for the bulk dynamic pressure. For achiral

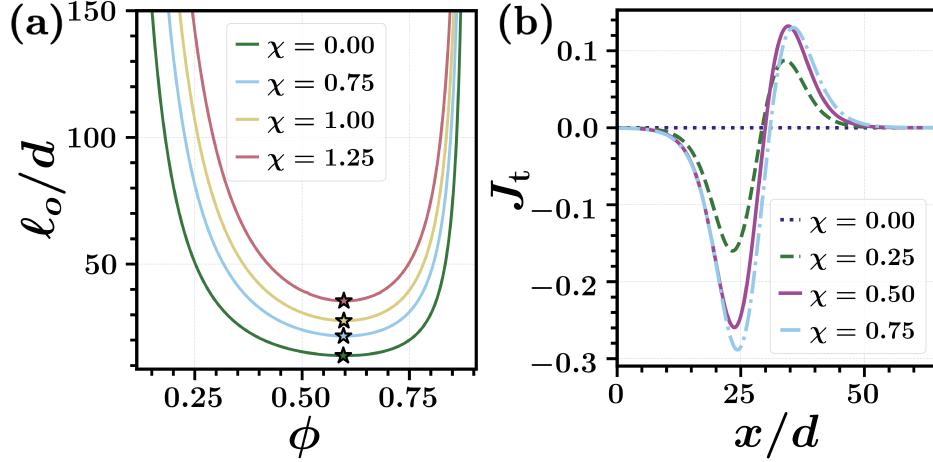


Figure 3.1: (a) Theoretical phase diagram of chiral ABPs for selected values of χ . Stars indicate critical points. (b) Theoretical steady-state $J_t(x)$ (units of U_o/a_p where $a_p = \pi d^2/4$ is the particle area) for $\ell_o/d = 125$. $J_t(x) \rightarrow 0$ as $\chi \rightarrow 0$.

ABPs, the nonmonotonic dependence of the active pressure is what gives rise to MIPS. Chirality acts to reduce the impact of the active pressure through the $(1 + \chi^2)$ factor present in Eq. (3.2d). Physically, the active pressure can be thought of as a measure of the *effective persistence length* of the particle trajectories with $p_{\text{act}} \propto \rho \zeta U_o \ell^{\text{eff}}$ where $\ell^{\text{eff}} \equiv U_o \bar{U} \tau_R / (1 + \chi^2)$. While the reduction in the effective run length arising from steric interactions has a significant density dependence (crucial for MIPS and reflected in $\bar{U}$), increasing chirality *uniformly* reduces ℓ^{eff} *for all densities*. Increasing χ causes particles to depart from their persistent trajectories and instead move in increasingly tight orbits, diminishing the active pressure. This quadratic rescaling of the effective run length with chirality is consistent with the analytical model of cABPs proposed in Ref. [100]. Finally, we note that the persistence motion of ABPs is what gives rise to many of their intriguing nonequilibrium behavior. In addition to MIPS, these behaviors include accumulation at boundaries [54] and inducing *repulsive* depletion forces [104] between passive particles. The reduction in effective run length with chirality may be a useful perspective towards understanding why these behaviors can be altered through chirality [101].

Intriguingly, while the average density flux normal to the interface must vanish

in this stationary coexistence scenario the *tangential flux* can remain finite. This stands in contrast to passive systems and active systems with achiral dynamics. While the flux associated with this density profile is zero in the direction normal to the interface, the flux *tangential* to the interface (i.e., parallel to $\mathbf{e}_y$) may be nonzero when χ is finite. We can compute the spatial tangential number density flux $J_t(x) \equiv \mathbf{J} \cdot \mathbf{e}_y$ by first numerically determining the complete spatial density profile, $\varphi(x)$ [42]. We indeed find *odd tangential flows* in the vicinity of the interface [see Fig. 3.1(b)] and we thus expect such flows to affect the interfacial dynamics, which existing theories of active interfaces [42, 48] have yet to account for. It is important to note that these finite tangential flux are permitted in our theory, but in an experiment which enforces no flux boundary conditions in all directions, the translational invariance of the density profile will be lost in the tangential direction, and details of the experimental geometry may become crucial for understanding phase behavior.

Fluctuating Hydrodynamics of Chiral Active Matter

To construct a theory for chiral interfacial dynamics, we first build a fluctuating hydrodynamic [53, 61] description of cABPs [see Appendix B.2 for further detail]:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \mathbf{J}, \quad (3.3a)$$

$$\mathbf{J} = \frac{1}{\zeta} \nabla \cdot \Sigma + \boldsymbol{\eta}^{\text{act}}, \quad (3.3b)$$

where $\boldsymbol{\eta}^{\text{act}}$ is a stochastic component of the flux with zero mean and correlations:

$$\begin{aligned} \langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \boldsymbol{\eta}^{\text{act}}(\mathbf{r}', t') \rangle = & 2 \frac{k_B T^{\text{act}}}{\zeta} (\rho \mathbf{I} - 2\mathbf{Q}) \\ & \times \delta(\mathbf{r}' - \mathbf{r}) \delta(t - t'). \end{aligned} \quad (3.4)$$

We note that the derivation of these dynamics has assumed the magnitude of the flux $|\mathbf{J}|$ to be small. Here, $k_B T^{\text{act}} \equiv \zeta U_o \ell_o / 2$ is the athermal active energy

scale and $\mathbf{Q}$ is the traceless nematic order tensor, which is a closed function of ρ within our approximations. Full expressions for the stress tensor contributions are provided in Appendix B.2. It is worth noting that the antisymmetric stress will affect the flux but cannot affect the density field dynamics as two divergences of any antisymmetric tensor must vanish. However, spatial parity violating terms (e.g., terms proportional to $\partial_x \rho \partial_y \rho$) also arise in the symmetric stress which *does* impact the density field dynamics.

Capillary Fluctuations and Odd Surface Flows

Now that we have described the flux-free phase separated states *and* weak fluctuations of the stochastic density field, we are now well-poised to probe dynamics of the interface. In order to connect the fluctuations of the density field to that of the interface, we introduce the linearizing ansatz proposed by Bray *et al.* [49, 97]:

$$\rho(\mathbf{r}, t) = \varphi[x - h(y)], \quad (3.5)$$

which assumes $|\partial_y h| \ll 1$.

As demonstrated by Ref. [48] and applied to microscopic systems in Chapter 2, substitution of Eq. (3.5) into Eqs. (3.3) and integration after multiplication by the appropriate Green's function as well as the appropriate pseudovector² results in a linearized Langevin equation for the interfacial dynamics. This Fourier transformed Langevin equation with k corresponding to a wavevector parallel to $\mathbf{e}_y$ (see Appendix B.2 and Chapter 2 for details) is given by:

$$\zeta_{\text{eff}} \frac{\partial h}{\partial t} = -k^2 |k| \gamma_{\text{cw}} h - i k |k| \nu_{\text{odd}} h + \xi^{\text{iso}} + \xi^{\text{aniso}} + \mathcal{O}(h^2 k^3), \quad (3.6)$$

where ζ_{eff} is an effective drag, γ_{cw} is the capillary tension [42, 48], and we have introduced ν_{odd} , the *odd flow coefficient*. ξ^{iso} is a noise originating from the

²While in achiral systems the pseudovector for interfacial dynamics is identical to the Maxwell construction pseudovector, they are generally different in chiral systems. This is further discussed in Appendix B.2

isotropic components of $\boldsymbol{\eta}^{\text{act}}$ with zero average and correlations:

$$\langle \xi^{\text{iso}}(k, t) \xi^{\text{iso}}(k', t') \rangle = 4\pi |k| (k_B T^{\text{act}}) \zeta_{\text{eff}} \delta(k + k') \delta(t - t'). \quad (3.7)$$

Full expressions for γ_{cw} , ν_{odd} , ζ_{eff} , (which are dependent on k), and the correlations of ξ^{aniso} are provided in Appendix B.2, where also we include plots of the k dependence of γ_{cw} , ν_{odd} , and ζ_{eff} .

Equation (3.6) differs from those derived by Refs. [42, 48] due to the complex term proportional to ν_{odd} , which couples real and imaginary parts of $h(k)$, thus generating flows determined by the sign of χ . However, the criteria for a stable interface remains $\gamma_{\text{cw}} > 0$. In this long wavelength limit (where ξ^{aniso} may be safely discarded [42]), the stationary fluctuations of $h(k)$ follow the familiar scaling of capillary-wave theory with:

$$\langle |h(k)|^2 \rangle = \frac{k_B T^{\text{act}} L}{\gamma_{\text{cw}} k^2}, \quad (3.8)$$

where L is the length of the system in the y -dimension. Additionally, in this long wavelength limit, the distribution of capillary waves may be determined from the steady-state Fokker-Planck equation [6] corresponding to Eq. (3.6):

$$P[h(k)] \sim \exp \left[-\frac{k^2 \gamma_{\text{cw}} \left(\text{Re}(h(k))^2 + \text{Im}(h(k))^2 \right)}{2L k_B T^{\text{act}}} \right]. \quad (3.9)$$

Surprisingly, only the capillary tension γ_{cw} enters the stationary statistics of $h(k)$. From Eqs. (3.8) and (3.9), one can see that interfacial fluctuations of chiral active interfaces are well-described by a surface-area minimizing Boltzmann distribution proportional to the interfacial stiffness $\gamma_{\text{cw}}/k_B T^{\text{act}}$ just as their achiral counterparts [42, 48]. The odd flow coefficient ν_{odd} , while crucial for describing the travelling-wave *dynamics* of $h(k)$, is irrelevant to the steady-state distribution governing the *structure* of $h(k)$. However, the impact of ν_{odd} is evident in the power spectrum which (for low k) takes the form:

$$\langle |h(k, \omega)|^2 \rangle = \frac{2|k| k_B T^{\text{act}} \zeta_{\text{eff}} L \Gamma}{|k|^6 \gamma_{\text{cw}}^2 + (\omega \zeta_{\text{eff}} + k|k| \nu_{\text{odd}})^2}, \quad (3.10)$$

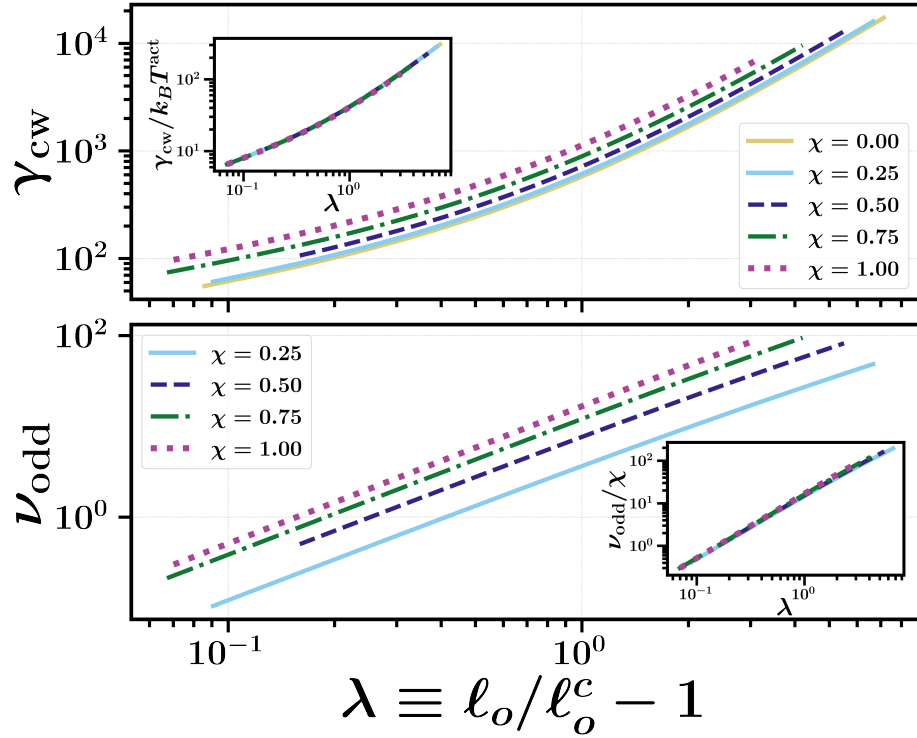


Figure 3.2: $k \rightarrow 0$ limit of γ_{cw} (units of $\zeta U_o/d$) and ν_{odd} (units of $\zeta U_o/d^2$) as a function of reduced run length. Insets demonstrate collapse of curves from normalization of γ_{cw} and ν_{odd} by $k_B T^{\text{act}}$ and χ , respectively.

where Γ is the total time duration of the observation window.

Inspecting the full expression for γ_{cw} [Appendix B.2], we find that its form is similar to that of parity-symmetric active systems and therefore should obey similar scaling. The primary influence of χ on the interfacial stiffness is through its impact on the critical point. We therefore compute $\gamma_{\text{cw}}(k \rightarrow 0)$ as a function of a *reduced* run length $\lambda \equiv \ell_o/\ell_o^c - 1$ [see Fig. 3.2], where the critical activity ℓ_o^c now increases with χ . Guided by our theory, a complete curve collapse is achieved upon normalization by $k_B T^{\text{act}}$ as shown in the Fig. 2.3 inset. Similarly, inspecting the form of ν_{odd} [Appendix B.2] we find that it is linearly proportional to χ . Evaluating $\nu_{\text{odd}}(k \rightarrow 0)$ as a function of λ [included in Fig. 3.2] also reveals a complete collapse of the odd flow coefficient upon normalization by χ .

Brownian Dynamics Data

Our theory predicts that the breaking of spatial-parity symmetry ($\chi \neq 0$) results in odd surface flows at the MIPS interface and that these odd surface flows play *no role* in the stationary statistics of $h(k)$, which is entirely controlled by the interfacial stiffness $\gamma_{\text{cw}}/k_B T^{\text{act}}$. Further, we predict that chirality acts to stabilize the homogeneous fluid, raising the critical point and narrowing the binodals for increasing χ at fixed ℓ_o . To test these predictions, we conduct large-scale Brownian dynamics (BD) simulations [65] of macroscopically phase separated cABPs at an array of values of χ and ℓ_o/d , with simulation details provided in Appendix B.3.

The density of the liquid and gas phase was measured for all simulated combinations of (ℓ_o, χ) and are tabulated in Appendix B.1. The measured densities follow the qualitative trend of narrowing binodals with increasing χ , but quantitative discrepancies between theory and simulation are prominent for the gas phase density. In order to measure the interfacial dynamics, the instantaneous shape of the interface $h(y)$ is extracted directly from Brownian dynamics [25]. The stationary height fluctuations $\langle |\tilde{h}(k)|^2 \rangle$ are then computed from $\tilde{h}(k)$, the *discrete* Fourier transform of $h(y)$. Figure 3.2 reveals both a collapse in the interfacial stiffness when plotted as a function of λ *and* a linear scaling with λ at high run length length. This theoretical prediction, combined with Eq. (3.8), implies that the product of $\langle |\tilde{h}(k)|^2 \rangle$ and λ should collapse across all values of ℓ_o and χ at low values of k where we expect our theory to hold. We find this collapse reflected in the fluctuation spectra measured from particle based simulation, [see Fig. 3.3(a)], demonstrating that chirality only impacts the stationary statistics by altering the MIPS critical point. Further, the predicted k^{-2} is also robustly measured with statistical uncertainty reported in Appendix B.2. Thus, the *qualitative* behavior of cABP interfaces is captured by our theory.

The odd flow coefficient ν_{odd} may be inferred from the power spectrum [see Eq. (3.10)], but in practice the measurement is challenging statistically. We therefore directly measure the odd surface flows by recording the tangential flux of particles, $J_t(x)$, as theoretically predicted in Fig. 3.1(b). ν_{odd} is finite when $\chi \neq 0$ and Eq. (3.6) implies that a net velocity of particles will develop in the vicinity

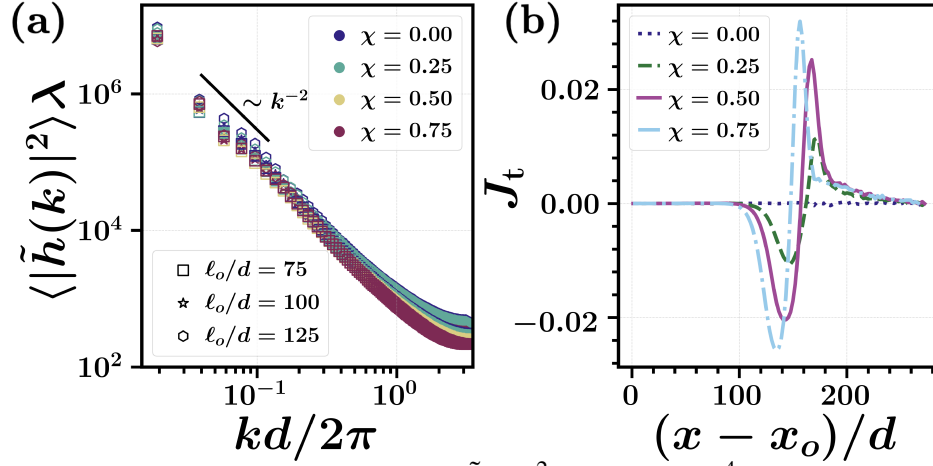


Figure 3.3: (a) Stationary fluctuation spectra $\langle |\tilde{h}(k)|^2 \rangle \lambda$ (units of d^4) for twelve combinations of ℓ_o/d and χ from BD simulations. k^{-2} scaling is measured across over a decade of k values, with error bars reported in Appendix B.2. Predicted collapse from multiplication by λ is also observed. (b) Steady-state $J_t(x)$ (units of U_o/a_p) from BD simulations for $\ell_o/d = 125$.

of the interface. Our simulations corroborate this while also revealing that the spatial profile of $J_t(x)$ [see Fig. 3.3(b)] is qualitatively similar to that predicted by our theory [see Fig. 3.1(b)]. The absence of quantitative agreement likely stems from a combination of factors such as the employed equations of state [see Appendix B.1] and the broadening of the interface due to capillary-waves [31, 43] not accounted for in the theoretically predicted profiles. The equations of state may also be responsible for quantitative discrepancies between the measured and theoretically calculated phase diagram (see Appendix B.1) and fluctuation spectra (see Appendix B.2). Nonetheless, our particle-based simulations verify all qualitative trends predicted by our theory.

3.3 Conclusions

In this Chapter we derive a nonequilibrium theory of phase coexistence, interfacial fluctuations, and odd surface flows for a model system of chiral active matter. We find that chirality both reduces the effective motility and induces non-zero currents tangential to the interface at steady-state, the latter of which emerges in the form

of an imaginary term proportional to an odd flow coefficient ν_{odd} in the interfacial dynamics. Despite the intriguing dynamics arising from these odd flows, the stability of interfacial dynamics and recovery of a surface-area minimizing principle remains controlled by γ_{cw} . Large-scale Brownian dynamics simulations corroborate our qualitative theoretical predictions. By extending the nonequilibrium theory of phase coexistence [19–21, 71, 105] and (linear) interfacial dynamics [42, 48, 49] to parity-breaking systems, it is our hope that these perspectives are now closer experimentally realizable active matter systems. A key remaining challenge towards this goal is understanding behavior at solid boundaries in chiral active matter [95, 106, 107]. Finally, we note that Ma *et al.* [99] recently demonstrated that cABPs can exhibit a diffusive instability that results in “dynamic clusters” which can preclude the observation of MIPS. A complete stability/phase diagram for chiral active matter, which includes both MIPS and the dynamic clustering instability, remains an outstanding challenge that is the subject of future work.

Chapter 4

Active Nucleation

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4.1 Introduction

In recent years it has become clear that many of the intriguing patterns and phases formed by active systems do not occur spontaneously but require a rate-determining rare event [75, 76, 78, 109]. The rare events needed to initiate a variety of active phase transitions often appear remarkably similar to those of the corresponding process in passive systems. Phase separation and crystallization of homogeneous active fluids, under certain conditions, require the formation of a *spherical critical nucleus* [63, 76]. Upon forming, the nuclei can continue to grow and coarsen through the familiar coarsening mechanisms of coalescence and Ostwald ripening [12, 16, 110]. These observations suggest that *classical nucleation theory* (CNT) and established coarsening theories may be applicable to active systems [32, 33, 45, 111–114]. CNT has shaped and framed our understanding of the transition rate of a variety of activated processes in passive systems including the rupture of biological membranes [115], the formation of dust clouds on exoplanets [116], and the motion of ferroelectric domain walls [117]. In its canonical form, CNT implicitly leverages microscopic time-reversal symmetry (TRS) to posit a *free energy barrier* for the rare event. In the case of a metastable fluid that will ultimately crystallize or phase separate, the rare event required to initiate the phase transition is the formation of a spherical finite-sized nucleus. Straightforward thermodynamic arguments weighing the competing effects of the volumetric driving force for forming a nucleus and the associated interfacial penalty allow for the determination of the barrier height and the critical nucleus' density and radius using bulk equations of state (e.g., the chemical potential) and the surface energy.

The naive application of the above ideas to active systems results in immediate issues: concepts such as chemical potential [18, 20] and surface energy [24, 27, 28, 39, 42, 48] are not uniquely defined in active systems. Nevertheless, a number of simulations and numerical studies in the last decade have made it clear that CNT and post-nucleation coarsening theories may be applicable far from equilibrium. Zakine *et al.* developed [118] and applied [119] action minimizing methods to stochastic field dynamics described by Active Model B [51] and found that the transition from homogeneous to inhomogeneous states is well described by the

rate-limiting formation of a radially symmetric nucleus. Particle-based simulations of metastable active fluids undergoing motility-induced phase separation (MIPS) conducted by Redner *et al.* [76] were consistent with the cluster rate formation statistics posited by Becker-Döring theory [32, 45, 112]. These observations suggest that one may be able to construct a theory for the nucleation of active phases with similar physical assumptions as CNT – but without invoking a thermodynamic variational principle.

Significant progress towards generalizing CNT to nonequilibrium fluids has been recently made by Cates, Nardini, and co-workers [39, 78]. Introducing the minimal stochastic field theory, Active Model B+ (AMB+), they demonstrated that the size and density of the critical nucleus can be solely determined from dynamics [39] and elucidated the conditions at which the critical nucleus represents an unstable fixed point. Furthermore, they derived the radial growth dynamics of active nuclei, finding theoretical justification for an effective free energy landscape that controls the size distribution of pre-critical nuclei [78]. Although these efforts have yielded crucial insights towards the nucleation of active phases, explicitly mapping microscopic models of active systems to the phenomenological coefficients of AMB+ may not always be possible. Moreover, the absence of a unique mechanical interpretation of active field theories makes it difficult to understand the physical significance of the *negative mechanical surface tension* [24] (which *does* control the Laplace pressure difference of active bubbles and droplets [19]) of active particles in nucleation.

In this chapter, we seek to answer the following questions beginning from a microscopic description and without appealing to equilibrium notions: Provided the saturation conditions, how might one determine the size (R_C) and density (ρ^{in}) of the critical nucleus?; what is the precise role of the various active surface tensions in the nucleation process?; and how might one describe the post-nucleation growth dynamics of coarsening active fluids? To address these questions, we begin by deriving the conditions for determining the density and radius of a critical nucleus from *purely mechanical considerations*. We apply this framework to fluid nucleation of active Brownian particles (ABPs) and determine the critical nucleus properties as a function of activity and the saturation density. We then combine

the recently developed perspective of Cates and Nardini [78] with our fluctuating hydrodynamics of ABPs [42] to describe the time evolution of the critical nucleus size, finding that droplets of ABPs are associated with a far higher nucleation barrier than bubbles. This nucleation barrier follows the familiar equilibrium intuition of an energetic *penalty* scaling with the surface area of the nucleus competing with an energetic *driving force* scaling with the volume of the nucleus. Building off our active CNT, we are able to extend classic perspectives on coarsening, including those of Lifhitz, Sylozov, and Wagner [33] as well as Voorhees and Glicksman [40, 41], to systems far from equilibrium. We are thus able to numerically investigate large-scale, late-stage coarsening dynamics that would otherwise be prohibitive for particle-based simulations.

Surprisingly, we find that the properties and dynamics of the critical nucleus of ABPs are dependent on *three* different surface tensions. These tensions include the negative mechanical surface tension [24], which controls the pressure difference between the nucleus and parent phase, the capillary-wave tension [42, 48], which drives interfaces to minimize surface area and results in a spherical nucleus geometry, and the Ostwald tension [39], which plays a central role in both the nucleation barrier and coarsening dynamics. At equilibrium, all three of these tensions must be identical and are defined by the energetic cost of increasing interfacial area, but far from equilibrium they are free to take on disparate values. Despite this nonequilibrium peculiarity, we demonstrate the recovery of phenomena typically associated with equilibrium, such as the Gibbs-Thomson effect and nucleation barriers, from dynamics that microscopically violate time reversal symmetry.

4.2 Results and discussion

Equilibrium Nucleation Theory

We briefly recapitulate the thermodynamic arguments of CNT applied to bubble/droplet nucleation, the focus of this work. The complete derivation can be found in Appendix C.5. We consider a macroscopic homogeneous single-component

system prepared with a uniform number density ρ^{sat} within a fixed system volume V (the total number of particles is thus also fixed at $N = V\rho^{\text{sat}}$). We focus on saturation densities ρ^{sat} within the liquid-gas binodal of the phase diagram but far from the spinodal boundaries, limiting our analysis to be far from the critical point. This limitation is necessary in order to ensure that the pathway of the phase transition is unambiguously nucleation and growth, rather than spinodal decomposition. In this region of the phase diagram, a system prepared with a spatially uniform density of ρ^{sat} will remain linearly stable against density fluctuations until a critical nucleus with density ρ^{in} and volume V^{nuc} is formed. We assume that the formation of the critical nucleus is a sufficiently rare event that we may consider a single nucleation event in isolation and a nucleus volume, V^{nuc} , that is much smaller than the system volume. These assumptions provide additional limits on the range of ρ^{sat} in which CNT is applicable.

The extensive Helmholtz free energy for the system is simply $\mathcal{F} = V^{\text{nuc}} f(\rho^{\text{in}}) + (V - V^{\text{nuc}}) f(\rho^{\text{out}}) + \partial V^{\text{nuc}} \gamma$ where $f(\rho)$ is the free energy density of a uniform fluid with density ρ , the density of the parent phase surrounding the nucleus is ρ^{out} , and $\gamma(> 0)$ is the surface energy. The interfacial penalty ensures that the mean-field nucleus shape will be spherical in order to reduce surface area, which we have labeled as ∂V^{nuc} . Conservation of the total particle number results in a constraint of $\rho^{\text{in}} V^{\text{nuc}} + \rho^{\text{out}} (V - V^{\text{nuc}}) = \rho^{\text{sat}} V$. Note that while the spatial density profile must smoothly vary from the nucleus density ρ^{in} to the parent phase density ρ^{out} this interfacial width is much smaller than the nucleus size and its contribution to the particle conservation constraint is negligible. This allows us to focus on the coexisting interior and exterior densities rather than the complete spatial profile. We then extremize the free energy (subject to the particle conservation constraint) with respect to the coexisting densities and nucleus size and find the following conditions:

$$\mu(\rho^{\text{in}}) = \mu(\rho^{\text{out}}), \quad (4.1a)$$

$$p(\rho^{\text{in}}) = p(\rho^{\text{out}}) + \frac{2\gamma}{R_C}, \quad (4.1b)$$

where we have defined the chemical potential $\mu(\rho) \equiv \partial f / \partial \rho$ and pressure $p(\rho) \equiv$

$-f(\rho) + \rho\mu(\rho)$. Here, R_C is the critical radius of the spherical nucleus. These criteria are the expected conditions of chemical potential equality and the Young-Laplace pressure difference which, due to the positive surface energy, results in a higher nucleus pressure relative to the parent phase. Together with the particle conservation constraint, Eqs. (4.1) can be used to solve for ρ^{in} , ρ^{out} and R_C .

We can make use of the condition that the droplet volume is much smaller than the system volume such that $\rho^{\text{out}} \approx \rho^{\text{sat}}$. In this limit, we have effectively changed our thermodynamic ensemble as the nucleus is now in contact with a particle reservoir with fixed chemical potential $\mu(\rho^{\text{sat}}) \equiv \mu^{\text{sat}}$ and thus ρ^{in} can be found directly from $\mu(\rho^{\text{in}}) = \mu^{\text{sat}}$. The critical radius is then simply $R_C = 2\gamma/(\mu^{\text{sat}}\Delta\rho - \Delta f)$ where the difference between an arbitrary property, a , in the interior and exterior of the nucleus is defined as $\Delta a \equiv a^{\text{in}} - a^{\text{sat}}$. The free energy barrier associated with the critical nucleus can be expressed as the sum of the volumetric and interfacial contributions $F^\dagger = 4\pi R_C^2\gamma - \frac{4}{3}\pi R_C^3(\mu^{\text{sat}}\Delta\rho - \Delta f)$ or equivalently, $F^\dagger = \frac{4}{3}\pi\gamma R_C^2$. The probability of observing such a critical nucleus is thus expected to be $P(R_C) \sim \exp[F^\dagger/k_B T]$.

It is clear that in passive systems, *thermodynamics alone* provides a guide for determining the density, size, and statistics of the critical nucleus. Given a saturation density, Eqs. (4.1) are the two equations that allow for the determination of the two unknowns: ρ^{in} and R_C . In the next part of this chapter, we derive analogous equations capable of determining ρ^{in} and R_C for systems *arbitrarily far from equilibrium* by adopting a mechanical perspective.

Mechanics of a Critical Nucleus

The pathway for transitioning from a homogeneous supersaturated active fluid to a state of two-fluid coexistence appears eerily reminiscent to those of passive systems [76, 119]. In both scenarios, the initial activated process is the formation of a *spherical nucleus* with a specific critical radius R_C and internal density ρ^{in} . In the absence of a free energy, the traditional arguments previously outlined used to establish passive classical nucleation theory are no longer applicable. We

therefore take a generalized *mechanical approach* with the aim of extending CNT to systems arbitrarily far from equilibrium. We again limit our analysis to saturation densities near the liquid and gas binodal densities such that the density of the fluid surrounding a critical nucleus is approximately ρ^{sat} and we may focus on a *single* nucleus coexisting with this saturated fluid.

Let us consider the dynamical definition of a critical nucleus. When the critical nucleus is an unstable fixed point ¹, nuclei smaller (larger) than R_C will be associated with a particle flux normal to the interface such that the nuclei shrink (grow). R_C thus represents a critical size at which the direction of this flux switches, so the system must have zero flux normal to the interface precisely when a nucleus has a radius of R_C . For a single realization of an active fluid, the stochastic noise plays a crucial role in the density flux, which we will discuss further later in this chapter. For the purposes of determining the properties of the critical nucleus, we can consider an *ensemble* of supersaturated states. We choose a spherical coordinate system with an origin coinciding with that of the nucleus. The ensemble-averaged nucleus shape will be spherical and the ensemble-averaged density flux will no longer be stochastic. We will determine the critical nucleus size by searching for states of zero radial flux.

Two flux-free states are well-established: the macroscopically phase separated state that the system will eventually reach and the initial metastable homogeneous state. The critical nucleus will correspond to an additional state in which a *finite-size* minority phase with zero radial flux coexists with a parent phase. The condition for determining a stationary state of a density profile is given by the linear momentum balance (see Appendix C.1 and Ref. [21]) which governs the dynamics of the density flux. On timescales where inertia can be neglected (or for all times for overdamped particle dynamics), the radial-component of the linear momentum balance is simply $(\nabla \cdot \sigma + \mathbf{b}) \cdot \mathbf{e}_r = 0$, where σ is the stress tensor, $\mathbf{b}$ are the body forces, and $\mathbf{e}_r$ is the unit vector in the radial dimension. We note that exact microscopic expressions for both σ and $\mathbf{b}$ can be determined through an

¹Tjhung *et al.* [39] demonstrated in some active systems the critical nucleus may be a stable fixed point, in such a case nuclei smaller (larger) than R_C will grow (shrink).

Irving-Kirkwood-Noll procedure [120–122], an approach that we will later adopt in applying this theory to active Brownian particles. In these formulations, terms often appear to be non-local, requiring integrals over all space to, for instance, compute the stress at a particular point in space arising from particle interactions. Nevertheless, these terms are often well-approximated (e.g., when interactions are short-ranged and partial spatial correlations decay quickly) by a series of local terms, the coefficients of which can be systematically determined [123]. We thus focus on systems in which $\boldsymbol{\sigma}$ and $\mathbf{b}$ can be fully expressed as local density gradient expansions. Such a focus may become limiting for application to systems with truly long-ranged forces that prevent a local expansion of stresses/body forces.

Up to a divergence-free term, we may absorb the body force into a stress-like contribution such that $\nabla \cdot \boldsymbol{\sigma} + \mathbf{b} = \nabla \cdot [\boldsymbol{\sigma} + \mathbf{F}] = \nabla \cdot \boldsymbol{\Sigma}$, where $\mathbf{F}$ is the Coulomb integral of $\mathbf{b}$ and $\boldsymbol{\Sigma} \equiv \boldsymbol{\sigma} + \mathbf{F}$ is the dynamic stress tensor. While $\boldsymbol{\sigma}$ and $\mathbf{b}$ are fully local, the Coulomb integral may result in non-local contributions to $\mathbf{F}$. The momentum balance ($\nabla \cdot \boldsymbol{\Sigma} = \mathbf{0}$) can be used to solve for the density profile upon identifying the precise form of the dynamic stress tensor. Under spherical symmetry, $\boldsymbol{\Sigma}$ has only two independent components and can be expressed as $\boldsymbol{\Sigma} = \Sigma_{rr} \mathbf{e}_r \mathbf{e}_r + \Sigma_{tt} (\mathbf{I} - \mathbf{e}_r \mathbf{e}_r)$ where $\mathbf{I}$ is the identity tensor. We can make analytical progress by expanding the dynamic stress tensor to second order in density gradients and assuming spatial isotropy in the expansion coefficients:

$$\begin{aligned} \Sigma_{rr} = & -\mathcal{P}(\rho) + \kappa_1(\rho) \left(\frac{\partial}{\partial r} + \frac{2}{r} \right) \frac{\partial \rho}{\partial r} + (\kappa_2(\rho) + \kappa_3(\rho)) \left(\frac{\partial \rho}{\partial r} \right)^2 \\ & + \kappa_4(\rho) \frac{\partial^2 \rho}{\partial r^2} + \mathcal{K}, \end{aligned} \quad (4.2a)$$

$$\Sigma_{tt} = -\mathcal{P}(\rho) + \kappa_1(\rho) \left(\frac{\partial}{\partial r} + \frac{2}{r} \right) \frac{\partial \rho}{\partial r} + \kappa_2(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{K}, \quad (4.2b)$$

where $-\mathcal{P}(\rho)$ is the bulk isotropic contribution to the dynamic stress tensor, $\mathcal{K}$ is the non-local term originating from the Coulomb integral of $\mathbf{b}$, and $\{\kappa_i\}$ are interfacial coefficients. These interfacial coefficients can be found by formally

carrying out the expansion of $\boldsymbol{\sigma}$ and $\mathbf{b}$ beginning from the microscopic definition of stress *and* expanding the local terms that arise from the Coulomb integral of $\mathbf{b}$. The procedure for determining the precise forms of $\{\kappa_i\}$ and $\mathcal{K}$ from the expansion coefficients of $\boldsymbol{\sigma}$ and $\mathbf{b}$ is outlined in Appendix C.1. In particular, a form for the nonlocal $\mathcal{K}$ term is found. We note that this form is free of any terms with explicit dependence on $1/r$. In a passive system $\mathcal{P}(\rho)$ is simply the bulk pressure and $\{\kappa_i\}$ are given by the Korteweg expansion [57] while non-local terms can be well-approximated by local terms for single-component systems with short-ranged interactions. More generally, the bulk pressure and interfacial coefficients can contain dynamical contributions otherwise absent in passive systems [42]. We also note that under the spherically symmetric configuration and stresses assumed here, setting the radial flux to zero also implies zero flux in the angular directions. In two dimensions, it is possible for a critical nucleus with radial symmetry to develop a finite angular flux if the underlying microscopic dynamics violate spatial parity. Describing the critical nucleus in chiral active matter [23, 99], would require a different expansion of the stress tensor such as those presented in Refs. [85, 103].

Given the above physical picture, we now outline our process to find the zero radial flux states associated with a finite-sized minority phase. Our spherically symmetric geometry results in the linear momentum balance simplifying to:

$$0 = \frac{\partial \Sigma_{rr}}{\partial r} - \frac{2}{r} \mathcal{G}(\rho), \quad (4.3)$$

where we have defined $\mathcal{G}(\rho) = \Sigma_{tt} - \Sigma_{rr}$ as the “excess stress” in the non-radial directions. Deep within the both the nucleus and parent phase, where density gradients vanish, the dynamic stress tensor must be isotropic, implying $\mathcal{G}(\rho)$ is *zero everywhere except within the interfacial region*. If we assume that the width of the interfacial region, w , is small compared to the radius of the critical nucleus, integration of Eq. (4.3) from $r = 0$ to $r = \infty$ results in:

$$\Delta \mathcal{P} = \mathcal{K}|_{r=0} + \frac{2\gamma_{\text{mech}}}{R_C} + \mathcal{O}\left(\frac{1}{R_C^2}\right). \quad (4.4a)$$

Here, γ_{mech} is the *mechanical surface tension* as defined by Kirkwood and Buff [26]:

$$\begin{aligned}\gamma_{\text{mech}} &= \int_0^\infty [\Sigma_{tt} - \Sigma_{rr}] \, dr, \\ &= - \int_0^\infty \left[\kappa_3(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 + \kappa_4(\rho) \frac{\partial^2 \rho}{\partial r^2} \right] \, dr,\end{aligned}\tag{4.4b}$$

where the second equality utilizes our specific form of the dynamic stress [see Eqs. (4.2)]. Equation (4.4a) is a generalized Young-Laplace equation – with the equilibrium surface energy and pressure more generally replaced by the mechanical tension and dynamic pressure, respectively, and an additional contribution arising from the non-local terms. Equation (4.4b) carries the familiar mechanical intuition of surface tension: it measures the excess stress parallel to the interface. This mechanical tension was determined to be *strongly negative* for phase-separated ABPs [24] and has been the subject of some controversy regarding its physical relevance. Solon *et al.* [19] correctly identified that the mechanical surface tension of Kirkwood and Buff controls the Laplace pressure difference of droplets consistent with a fully local (dynamic) stress. More generally, the nonlocal term $\mathcal{K}$ may also contribute to the Laplace pressure difference. Note that Eq. (4.4a) ignores terms higher order in $1/R_C$. This approximation becomes increasingly accurate as the saturation is reduced [see Fig. 4.2].

While the generalized Young-Laplace equation provides a mechanical condition remarkably similar to its equilibrium counterpart, we still require a second condition to allow us to determine our two unknowns, ρ^{in} and R_C . For passive fluids, this condition is equality of chemical potential between the nucleus and the surrounding phase. The search for chemical potential-like quantities for nonequilibrium systems has been the subject of much interest in the last decade in the context of determining macroscopic binodals [18, 20, 21, 56]. These works have resulted in generalized Maxwell constructions [20, 21] that are derived from performing the appropriate integration of the mechanical criteria of coexistence [21, 71].

We follow similar arguments used in constructing the nonequilibrium coexis-

tence criteria of macroscopic binodals [21, 71]. Beginning from the mechanical balance [Eq. (4.3)] we perform an indefinite integral to find an integro-differential equation for Σ_{rr} with the Laplace pressure used as a boundary condition. We then introduce the generalized Maxwell construction pseudovariable [20, 21, 71] $\mathcal{E}$ defined by the differential equation $(\kappa_1 + \kappa_4)\partial^2\mathcal{E}/\partial\rho^2 = (2(\kappa_2 + \kappa_3) - \partial(\kappa_1 + \kappa_4)/\partial\rho)\partial\mathcal{E}/\partial\rho$, which is found to be $\mathcal{E} \sim 1/\rho$ for a passive system. This pseudovariable allows us to eliminate nonlinear terms (just as it does in the case of macroscopic coexistence [20, 21, 71]) by multiplying the antiderivative of Eq. (4.3) by $d\mathcal{E}/dr$ and integrating from $r = 0$ to $r = \infty$ to arrive at:

$$\begin{aligned} \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} d\mathcal{E} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] + \int_0^\infty (\mathcal{E}^{\text{in}} - \mathcal{E}) \frac{\partial\mathcal{K}}{\partial r} dr \\ = \frac{2\gamma_{\text{ost}} \rho^{\text{sat}} \Delta\mathcal{E}}{R_C \Delta\rho} + \mathcal{O}\left(\frac{1}{R_C^2}\right), \end{aligned} \quad (4.5a)$$

where, analogously to Tjhung *et al.* [39], we have defined the ‘‘Ostwald’’ tension as

$$\gamma_{\text{ost}} = \frac{\Delta\rho}{\rho^{\text{sat}} \Delta\mathcal{E}} \left[\int_0^\infty \left(\kappa_1(\rho) \frac{\partial\mathcal{E}}{\partial\rho} \left(\frac{\partial\rho}{\partial r} \right)^2 + \mathcal{G}(\mathcal{E} - \mathcal{E}^{\text{in}}) \right) dr \right]. \quad (4.5b)$$

Note that γ_{ost} has been defined such that it has the same units as mechanical surface tension and becomes identical to γ_{mech} in a passive system. γ_{ost} is named for its importance in understanding the evaporation-condensation mechanism of active coarsening, which we will discuss in final section of this chapter. As ρ^{sat} approaches a binodal of the phase diagram, the right hand side of Eq. (4.5a) vanishes and we are left with one of the macroscopic coexistence criteria. Interestingly, a term depending on gradients (and therefore the interface) remains. At equilibrium, the details of the interface do not modify the coexistence criteria as interfacial effects are generally sub-extensive in the free energy. However, outside of equilibrium no such guarantee exists. This interfacial term vanishes as the derivative of $\mathcal{K}$ approaches zero, and one may refer to the expansion of $\mathcal{K}$ in Appendix C.1 for a precise expression of this limit.

The connection between Eqs. (4.4a) and (4.5a) to the thermodynamic criteria shown in Eqs. (4.1) can be made explicit. As described by Solon *et al.*, the generalized Maxwell constructions on the pressure can also be expressed as a difference in chemical potential-like quantities between coexisting phases [19]. It was subsequently shown that this potential satisfies a generalized Gibbs-Duhem relation [64], which, for the single-component system under consideration, is expressed as $dg = \mathcal{E}d\mathcal{P}$ where g is the generalized chemical potential. The generalized Gibbs-Duhem relation allows Eq. (4.5a) to be expressed as a difference of chemical potentials with the nonequilibrium critical nucleus criteria now taking the form:

$$\Delta g = \mathcal{E}^{\text{in}} \mathcal{K}|_{r=0} + \int_0^\infty \left(2\mathcal{E}^{\text{in}} - \mathcal{E} \right) \frac{\partial \mathcal{K}}{\partial r} dr + \frac{2}{R_C} \left[\mathcal{E}^{\text{in}} \gamma_{\text{mech}} + \frac{\rho^{\text{sat}} \Delta \mathcal{E}}{\Delta \rho} \gamma_{\text{ost}} \right], \quad (4.6a)$$

$$\Delta \mathcal{P} = \mathcal{K}|_{r=0} + \frac{2\gamma_{\text{mech}}}{R_C}. \quad (4.6b)$$

Additionally, by defining a pseudopotential $\Phi \equiv \int \mathcal{P} d\mathcal{E}$, Eq. (4.5a) can be rearranged to express the critical radius as $R_C = 2\gamma_{\text{ost}} \rho^{\text{sat}} \Delta \mathcal{E} / \Delta \rho (\Delta \mathcal{E} \mathcal{P}^{\text{sat}} - \Delta \Phi)$. For passive systems without non-local terms, $\gamma_{\text{mech}} = \gamma_{\text{ost}}$ and $\mathcal{E} \sim 1/\rho$. One can use these relations to readily verify that $g^{\text{in}} = g^{\text{sat}}$ and that Eq. (4.5a) recovers the thermodynamically derived expression for R_C . Our mechanical criteria for the critical nucleus [Eqs. (4.6)] thus recover and generalize the thermodynamic criteria [Eq. (4.1)].

Equations (4.6) [or, equivalently, Eqs. (4.4) and (4.5)] are the *finite-size* coexistence criteria for the critical nucleus. In Ref. [39], finite-sized coexistence criteria were derived which appear to be distinct from those of the present work. Fundamentally, this is because pressure (referred to as “pseudopressure”), surface tension (referred to as “pseudotension”), and chemical potential have different physical interpretations in Ref. [39] than in this work. Indeed, Ref. [39] explicitly cautioned against interpreting pseudopressure and pseudotension in the same manner as mechanical pressure and mechanical surface tension. In Appendix C.2 we formally

map AMB+ to our mechanical treatment of finite-size coexistence. Despite their ambiguous physical interpretation, we find that the finite-sized coexistence criteria presented in Ref. [39] are fully consistent with those found here. The unambiguous physical interpretation of the mechanical quantities used in our work represents an advantage of a mechanical perspective which also has a clear connection to microscopics through the Irving-Kirkwood-Noll procedure [120–122].

Given a saturation density, one may use Eqs. (4.6) to solve for the unknowns ρ^{in} and R_C . We note that as ρ^{sat} approaches either binodal density, R_C will diverge; Eq. (4.4a) and Eq. (4.5a) then simplify to the macroscopic phase coexistence criteria presented in Ref. [21]. As one approaches this limit and R_C grows, $O(R_C^{-2})$ corrections to the coexistence criteria become negligible and we expect our theory to be most accurate. Solution of the coexistence criteria is complicated by the presence of γ_{mech} and γ_{ost} in Eqs. (4.6), which depend on the density profile, which in turn depends on R_C and ρ^{in} . Therefore, solution of Eqs. (4.6) requires *self-consistently* solving for the density profile using a procedure detailed in Appendix C.1. In the next section of this chapter, we apply our mechanical theory to the canonical model system we considered earlier for capillary waves: active Brownian particles.

Nucleation of Motility-Induced Phases

We consider a system of N interacting ABPs. For convenience we reiterate their dynamics here. The time variation of the position $\mathbf{r}_i$ and orientation $\mathbf{q}_i$ of the i th particle follow overdamped Langevin equations:

$$\dot{\mathbf{r}}_i = U_o \mathbf{q}_i + \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij}, \quad (4.7a)$$

$$\dot{\mathbf{q}}_i = \mathbf{q}_i \times \boldsymbol{\Omega}_i, \quad (4.7b)$$

where $\mathbf{F}_{ij}$ is the (pairwise) interparticle force, U_o is the intrinsic active speed, ζ is the translational drag coefficient, and $\boldsymbol{\Omega}_i$ is a stochastic angular velocity with

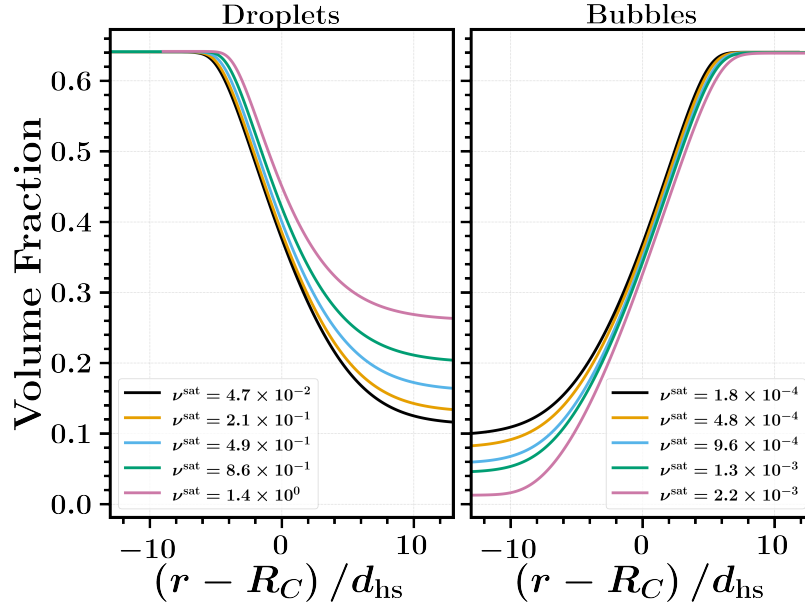


Figure 4.1: Density profiles of critical bubbles and droplets at $\ell_o/d_{\text{hs}} = 150$. All profiles have been shifted by their corresponding critical radius, given in Fig. 4.2. Here, the volume fraction is defined as the number density multiplied by the effective volume of a particle $\rho\pi d_{\text{hs}}^3/6$.

zero mean and variance $\langle \mathbf{\Omega}_i(t) \mathbf{\Omega}_j(t') \rangle = 2D_R \delta_{ij} \delta(t - t') \mathbf{I}$. Here, D_R is the rotational diffusivity, which may be athermal in origin, and is inversely related to the orientational relaxation time $\tau_R \equiv D_R^{-1}$. For the remainder of this Article, we consider purely repulsive $\mathbf{F}_{ij}$, corresponding to a sufficiently stiff interaction potential such that hard-sphere statistics are closely approximated with an effective hard-sphere diameter of d_{hs} [63, 124]. One can then define an intrinsic run length, $\ell_o/d_{\text{hs}} = U_o \tau_R$, the typical distance an ideal particle travels before reorienting.

In three dimensions, the form of $\mathcal{P}(\rho)$ and $\{\kappa_i(\rho)\}$ for ABPs are given by [21, 42]:

$$\mathcal{P}(\rho) = p_C(\rho) + \frac{\rho \zeta U_o \ell_o \bar{U}(\rho)}{6}, \quad (4.8a)$$

$$\kappa_1(\rho) = \frac{\ell_o^2}{20} \left(\bar{U}(\rho) \right)^2 \frac{\partial p_C}{\partial \rho}, \quad (4.8b)$$

$$\kappa_3(\rho) = \frac{\ell_o^2}{20} \bar{U}(\rho) \frac{\partial}{\partial \rho} \left[\bar{U}(\rho) \frac{\partial p_C}{\partial \rho} \right], \quad (4.8c)$$

$$\kappa_2(\rho) = \kappa_4(\rho) = 0, \quad (4.8d)$$

$$\mathcal{K} = 0, \quad (4.8e)$$

where $p_C(\rho)$ is the *conservative* pressure arising from particle interactions and $\bar{U}(\rho)$ is the dimensionless effective active speed. In Eqs. (4.8) we have ignored the gradient contributions of the interaction potential to Σ , which is justified when $\ell_o/d_{\text{hs}} \gg 1$ for purely repulsive interactions [19, 21, 64]. Given our expressions for $\{\kappa_i(\rho)\}$, one can solve for the generalized Maxwell construction pseudovariabe $\mathcal{E} \sim p_C(\rho)$ [21]. We also note that because $\mathcal{K} = 0$, Eq. (4.4a) reduces to the familiar Laplace pressure difference (i.e. no non-local corrections) and the negative sign of γ_{mech} [24] implies that the pressure inside of a droplet is *lower* than the pressure outside of the droplet [19]. The phase diagram, dependent on ℓ_o/d_{hs} and volume fraction $\phi \equiv \rho \pi d_{\text{hs}}^3/6$, has been well established computationally [63] and theoretically [21] with the latter predicting a critical point at $\phi \approx 0.49$ and $\ell_o \approx 16.2 d_{\text{hs}}$.

With the dynamic stress of ABPs in hand, we are able to self-consistently solve our generalized critical nucleus conditions [Eqs. (4.6)], with the resulting density profiles for both bubbles and droplets shown in Fig. 4.1 for a select activity and various supersaturations. Values of R_C and ρ^{in} obtained from self-consistent solution of the criteria at various activities and supersaturations are provided in Fig. 4.2. Here, we define a dimensionless overall supersaturation density as the relative departure of the majority phase density from the binodal density, $\nu^{\text{sat}} = |\rho^{\text{sat}} - \rho^{\text{gas,liq}}|/\rho^{\text{gas,liq}}$ and a dimensionless nucleus density as

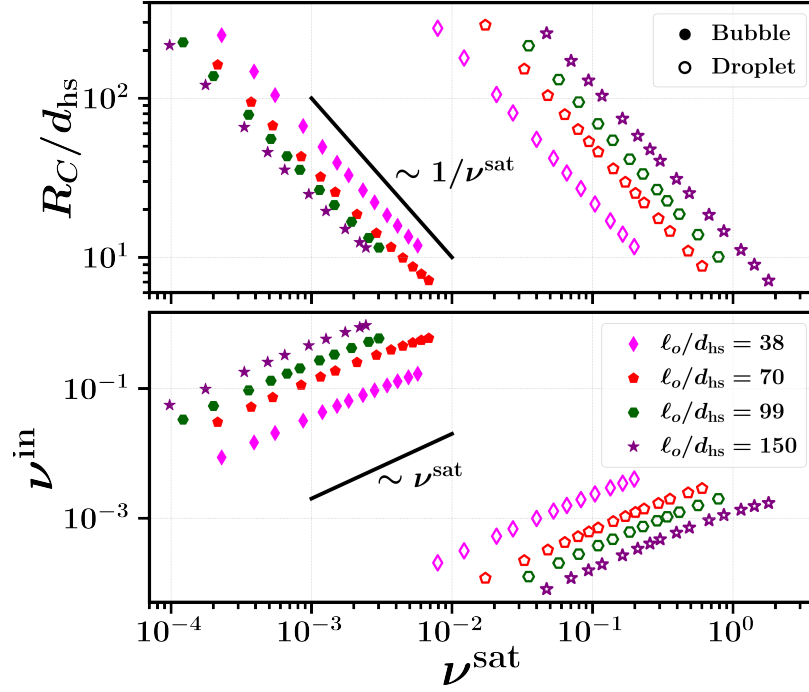


Figure 4.2: Critical radius and relative change in nucleus density (ν^{in}) as a function of supersaturation (ν^{sat}), as calculated from Eq. (4.6) using the numerical procedure outlined in Appendix C.1. Each data point requires self-consistent solution of the corresponding full spatial profile, e.g. those shown Fig. 4.1. Linear dependence of ν^{in} on ν^{sat} and inverse dependence of R_C/d_{hs} on ν^{sat} implies a Gibbs-Thomson relation with $\nu^{\text{in}} \propto 1/R$.

the relative departure of the minority phase density from the opposite binodal $\nu^{\text{in}} = |\rho^{\text{in}} - \rho^{\text{liq,gas}}|/\rho^{\text{liq,gas}}$. Our theory is expected to hold as $\nu^{\text{sat}} \rightarrow 0$. We observe a roughly linear relationship between ν^{sat} and ν^{in} as well as an inverse relationship between ν^{sat} and R_C . These results are therefore in excellent agreement with existing simulation data reporting that ABPs obey a Gibbs-Thomson relation with $\nu^{\text{in}} \propto 1/R$ [27].

Figure 4.2 displays a striking difference between critical bubbles and droplets. At all examined values of activity, critical droplets of active Brownian particles are over an order of magnitude larger than critical bubbles under similar saturation conditions. This difference between bubbles and droplets widens as activity is increased and the phases become increasingly distinct. To contextualize this order

of magnitude difference, we calculate the critical nucleus size and density for an *equilibrium* van der Waals fluid. While the size of the critical droplets are indeed larger than the size of critical bubbles in passive fluids, they remain within an order of magnitude of each other at similar saturation conditions even far from the critical point. In equilibrium, this difference is rooted in the asymmetry of the equations of state outside of the binodal densities: the vapor bubbles are more compressible than the liquid droplets. This asymmetry results in larger Laplace pressure differences for bubbles in comparison to droplets under similar saturation conditions, leading to the critical radius discrepancy. While this asymmetry in the compressibility is also present for active systems, the coexistence criteria contain uniquely nonequilibrium contributions, including a significant discrepancy between the vapor and bubble Ostwald tensions. At equilibrium these two quantities are identical, but can differ by an order of magnitude in active systems as shown in Fig. 4.4.

For a more physical understanding of the discrepancy in critical nucleus size between bubbles and droplets, we draw on the kinetic arguments independently proposed by Redner *et al.* [76] and Lee [27]. Establishing a flux-free critical nucleus with a density (and therefore interaction pressure) distinct from its surroundings requires the generation of an interfacial region of particles with a polarization pointing into the dense phase. Crucially, the rate at which particles with this orientation arrive to the interface must equal the rate at which they reorient and escape in order to satisfy the flux-free condition. The arrival rate is controlled by gas phase active speed and density while the escape rate is controlled by the minimum angle a particle must reorient in order to escape, the so-called “horizon angle [76].” For the same R_C , the rate of escape will depend greatly on whether the nucleus is the liquid phase (small horizon angle) or the gas phase (large horizon angle). As a result, for a similar supersaturation, the droplet must have a larger critical radius in order to satisfy the flux-free condition.

Langevin Dynamics of a Nucleus

Further insight into the active nucleation process can be obtained by considering the growth dynamics of nuclei near their critical size. We assume the dynamics of the

nucleus to proceed quasistatically: any given nucleus with radius R (not necessarily equal to R_C) will be in a local mechanical balance with its surroundings. We now distinguish the density just outside the nucleus (now denoted as ρ^{out}), and the density far from the nucleus which remains fixed at the saturation density, ρ^{sat} . The mechanical balance between the nucleus and its immediate surroundings will determine the intermediate density which will generally not equal the far away saturation density, $\rho^{\text{out}} \neq \rho^{\text{sat}}$. The inequality between the intermediate and far away densities will drive the transport of particles and will cause ρ^{out} to evolve in time. This, in turn, will cause R to also change in time as determined by the local mechanical balance. Cates and Nardini [78] recently demonstrated that such a description can allow for the determination of effective nucleation barriers of active fluids with dynamics described by AMB+, despite the absence of well-defined free energy for these driven systems. Deriving the stochastic growth dynamics of the nuclei requires a fluctuating hydrodynamic description of the density field which we recently obtained for interacting ABPs [42]:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \mathbf{J}, \quad (4.9a)$$

$$\mathbf{J} = \frac{1}{\zeta} \nabla \cdot \Sigma + \boldsymbol{\eta}^{\text{act}}, \quad (4.9b)$$

where ρ now is a *stochastic* density field (while in the previous sections it was an ensemble average), Σ is defined identically as the dynamic stress tensor of the previous section and $\boldsymbol{\eta}^{\text{act}}$ is an athermal stochastic contribution to the flux with zero mean and a variance of:

$$\begin{aligned} \langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \boldsymbol{\eta}^{\text{act}}(\mathbf{r}', t') \rangle = & 2 \frac{k_B T^{\text{act}}}{\zeta} \left(\rho \mathbf{I} - \frac{d}{d-1} \mathbf{Q}' \right) \\ & \times \delta(t - t') \delta(\mathbf{r} - \mathbf{r}'). \end{aligned} \quad (4.9c)$$

Here $\mathbf{Q}'$ is the traceless nematic order and we have defined $k_B T^{\text{act}} = \ell_o \zeta U_o / d(d-1)$ as the active energy scale.

We recently established a surface-area minimizing principle for liquid-gas interfaces of ABPs resulting from a positive capillary-wave tension, γ_{cw} , which

resists fluctuations to the area-minimizing interfacial shape. The strength of the fluctuations were found to be set by the athermal active energy scale $k_B T^{\text{act}}$ [42]. The ratio of the capillary tension to this energy scale – the active interfacial stiffness – controls the amplitude of long-wavelength interfacial fluctuations. A fluctuating nucleus can be well-approximated as a sphere when the average radius R satisfies $(R^2 \gamma_{\text{cw}} / k_B T^{\text{act}})^{1/2} \gg 1$. Under this condition, the stochastic density field dynamics can be connected to the radial growth dynamics of the nucleus through the following radially symmetric ansatz [78, 97]:

$$\rho(\mathbf{r}, t) = \varphi_R [r - R(t)], \quad (4.10)$$

where φ_R is the noise-averaged density profile associated with a nucleus of radius R . The density profiles calculated in the previous section correspond to φ_R when $R = R_C$. The ansatz may be substituted into our fluctuating hydrodynamics equations [Eqs. (4.9)], allowing us to obtain the following stochastic radial growth dynamics:

$$\zeta_{\text{eff}} \frac{\partial R}{\partial t} = F^D(R) + \tilde{\chi}(t), \quad (4.11a)$$

$$\langle \tilde{\chi}(t) \rangle = 0, \quad (4.11b)$$

$$\langle \tilde{\chi}(t) \tilde{\chi}(t') \rangle = 2k_B T^{\text{act}} \zeta_{\text{eff}} \delta(t - t'), \quad (4.11c)$$

$$\zeta_{\text{eff}} = \frac{4\pi \Delta \rho^2 R^3 \zeta}{\rho^{\text{sat}}}, \quad (4.11d)$$

$$F^D(R) = -8\pi \gamma_{\text{ost}} R \left[1 - \frac{R}{R_C} \right]. \quad (4.11e)$$

The radial growth dynamics take the form of a one-dimensional overdamped Langevin equation containing three distinct forces: a drag force with drag coefficient ζ_{eff} , a stochastic force with an athermal temperature scale of $k_B T^{\text{act}}$ [125], and a deterministic force $F^D(R)$ proportional to the Ostwald tension. This deterministic force vanishes when $R = R_C$ where the density outside nucleus is identical to the far away saturation density $\rho^{\text{out}} = \rho^{\text{sat}}$. Crucially, the noise statistics satisfy the fluctuation-dissipation theorem (FDT) despite the underlying microscopic

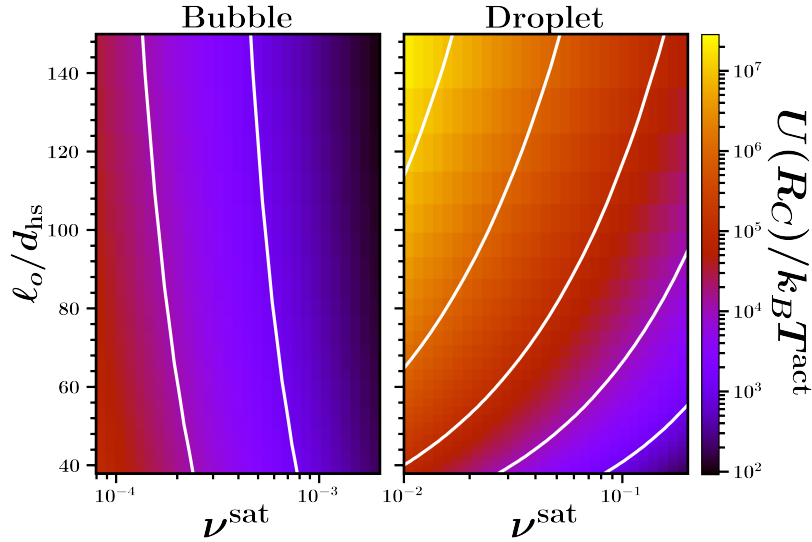


Figure 4.3: Barrier $U(R_C)$ relative to active energy scale as a function of activity and supersaturation. The distinct white contour lines distinguish values of $U(R_C)/k_B T^{\text{act}}$ separated by orders of magnitude.

dynamics [Eqs. (4.7)] violating FDT. We can plainly see that γ_{ost} is the quantity controlling the dynamics of nucleation and ripening. As first argued by Thjung *et al.* [39] a positive γ_{ost} implies R_C is an unstable fixed point in which nuclei with $R > R_C$ tend to grow while nuclei with $R < R_C$ tend to shrink. *A priori*, there is no reason to assume γ_{ost} is generally positive. In the case of a negative γ_{ost} , R_C becomes a *stable* fixed point. In such a scenario, one may expect phenomenology ranging from microphase separation to a “bubbly liquid” phase coexisting with a gas phase [39]. However, for ABPs we do not find a negative γ_{ost} [see Fig. 4.4] – consistent with existing simulation work on *three-dimensional* ABPs [34, 42, 63, 126]. In two dimensions, some simulations of ABPs have reported phenomenology consistent with bubbly liquids [34, 38]. Whether these bubbly phases are a result of a negative γ_{ost} or other effects [35] remains an open question.

As our deterministic force is a function of a single variable, R , we can define (within an integration constant) an effective conservative potential $U(R)$:

$$U(R) = 8\pi\gamma_{\text{ost}} \left[\frac{R^2}{2} - \frac{R^3}{3R_C} \right] \quad (4.12)$$

with $F^D(R) = -\partial U/\partial R$. In defining this potential, we have treated γ_{ost} as independent of R (just as in Ref. [78]), an approximation that appears to be well justified while $R \sim R_C$. This potential may be rearranged into a form familiar to equilibrium intuition:

$$U(R) = \underbrace{4\pi R^2 \gamma_{\text{ost}}}_{\text{Interface}} - \underbrace{\frac{4}{3}\pi R^3 \mathcal{V}}_{\text{Bulk}}, \quad (4.13a)$$

where:

$$\mathcal{V} = \frac{\mathcal{P}(\rho^{\text{sat}})\Delta\rho}{\rho^{\text{sat}}} - \frac{\Delta\Phi\Delta\rho}{\rho^{\text{sat}}\Delta\mathcal{E}}. \quad (4.13b)$$

At equilibrium, $\mathcal{V}$ simplifies to the expected $\mu(\rho^{\text{sat}})\Delta\rho - \Delta f$. From Eq. (4.13), one can clearly appreciate that the barrier contains two contributions: a *positive* term which depends solely on *interfacial* properties scaling as R^2 and a *negative* term which depends solely on *bulk* properties scaling as R^3 . Thus the familiar picture of homogeneous nucleation where interfacial effects compete with bulk effects in order to define a critical radius and nucleation barrier is recovered but now contains generalized volumetric driving and interfacial penalty terms.

Evaluating the potential at the critical radius allows us to compare the relative strengths of the nucleation barrier across activities and saturations, as shown in Fig. 4.3. Here we see a decrease in the nucleation barrier as ν^{sat} increases, consistent with equilibrium intuition. We further observe that the nucleation barrier height is much higher for droplets in comparison to bubbles. The disparity in nucleation barrier between droplets and bubbles has an attractive physical interpretation: much like the capillary tension of fluids comprised of ABPs [42], γ_{ost} is rooted in *nematic* flows. As we discuss in recent work [42], these nematic flows lead to particles accumulating at *convex* liquid surfaces and escaping from *concave* liquid surfaces. Then for *small* nuclei $R < R_C$, where curvature is high, droplets (concave liquid surfaces) are less favorable to grow than bubbles (convex liquid surfaces). This demonstrates that while Eqs. (4.11) satisfies the fluctuation dissipation theorem and allows for the definition of a conservative potential, the underlying physics of this conservative potential remain rooted in nonequilibrium effects.

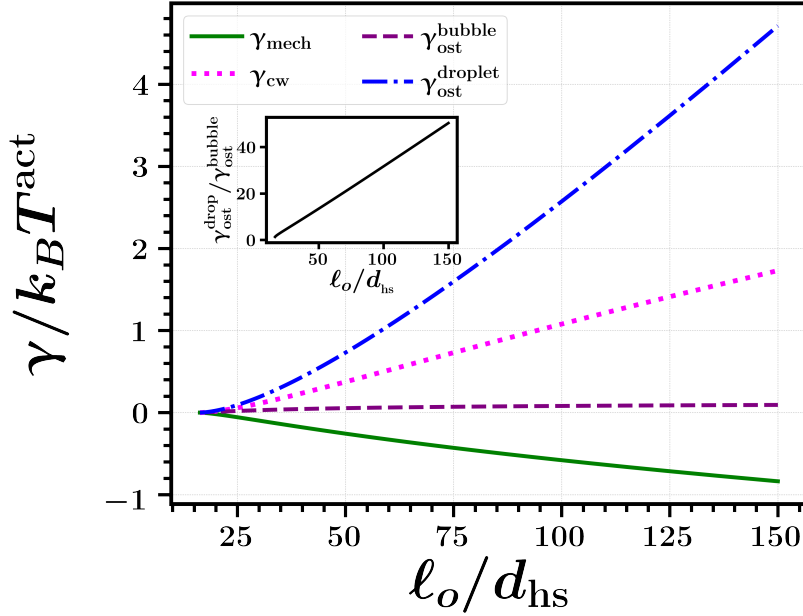


Figure 4.4: Surface tensions as defined by γ_{mech} , $k \rightarrow 0$ limit of γ_{cw} , and $R \rightarrow \infty$ limit of γ_{ost} for both bubbles and droplets as a function of run length.

The recovery of an effective detailed balance with respect to the size of a sub-critical nucleus directly implies that the probability of observing a nucleus of size $R < R_C$ is proportional to $\exp[-U(R)/k_B T^{\text{act}}]$ [78]. We note that the potential defined here relied on the assumption that the density profiles and quantities such as γ_{ost} are constant with R an approximation that clearly breaks down when $R \ll R_C$. Even in equilibrium, the nucleation potential formulated using bulk thermodynamic quantities (e.g the so-called "capillary approximation") is often used for all $R < R_C$ despite likely being beyond the range of applicability of bulk thermodynamic quantities [112, 127, 128]. Nevertheless, use of this potential for all R allows one to straightforwardly extend the arguments of Becker-Döring theory [32, 45, 112] to extract the following kinetic expression for the rate at which an active droplet nucleates per unit volume, as outlined in Appendix C.4:

$$\mathcal{R} = \frac{(\rho^{\text{sat}})^2}{\rho^{\text{in}}} U_o \sqrt{\frac{\pi \gamma_{\text{ost}}}{2 k_B T^{\text{act}}}} \exp \left[-\frac{U(R_C)}{k_B T^{\text{act}}} \right]. \quad (4.14)$$

The characteristic timescale to observe a nucleation event is given by $\tau_{\text{nuc}} = (V\mathcal{R})^{-1}$ where V is the total volume of the system. Thus the work of Redner *et al.* [76], which inferred the existence of a nucleation barrier by observing clustering statistics consistent with Becker-Döring theory from particle simulation, is well-justified on theoretical grounds, although our calculated barrier is for three dimensions while the simulations of Ref. [76] are in two. We note that the rate [Eq. (4.14)] is applicable to the nucleation of active droplets but not to active bubbles – the physical interpretation of the “clusters” posited in the derivation of Eq. (4.14) becomes unclear [112] in this context.

Active Coarsening Dynamics

While Eqs. (4.6) and Eqs. (4.11) were motivated to describe the earliest stages of nucleation, i.e. the structure and dynamics of a single nucleus forming in a supersaturated solution, they may also describe late-stage coarsening dynamics long after all nucleation events have occurred. At this stage, the system will contain several nuclei of different sizes. The coarsening of these domains can proceed through a combination of several different mechanisms, including evaporation-condensation (Ostwald ripening) and coalescence. At sufficiently low ν^{sat} , the nuclei will be separated by large enough distances that collisions between them are exceedingly rare and coarsening will be dominated by Ostwald ripening. In such a scenario, Eqs. (4.11) can describe the dynamics of each nucleus such that they are only coupled via the density of the majority phase, ρ^{sat} , which now evolves with time. The finite-size coexistence criteria presented in this work imply minor differences in density of the majority phase just outside nuclei of different sizes, inversely proportional to their radii. The majority phase therefore has an inhomogeneous density, driving transport of mass away from nuclei with enriched outside densities towards nuclei with depleted outside densities. For passive systems as well as ABPs [see Fig. 4.1], smaller droplets have enriched outside densities while smaller bubbles have depleted outside densities. Noting that transport of mass away from a nucleus would cause a droplet to shrink and a bubble to grow, this implies that large nuclei of minority phase will grow at

the expense of smaller nuclei until the system reaches a final steady state with a single macroscopic domain. Nuclei compete to absorb (or inject) particles from the majority phase and therefore R_C , the radius below which nuclei shrink and above which nuclei grow, is now a function of time. While we do not have profiles for a particular system in which $\gamma_{\text{ost}} < 0$, we suspect they would reveal smaller droplets having depleted outside densities and smaller bubbles having enriched outside densities. This would result in smaller nuclei growing at the expense of larger ones, consistent with stable radial dynamics of Eq. (4.11).

With the above physical picture in mind, we can determine the time-dependence of the post-nucleation critical radius in a similar manner as the Lifshitz-Slyozov-Wagner (LSW) theory [33]. In the limit of a vanishing phase fraction of the minority phase, the nuclei can be approximated as independent with each satisfying the Langevin equation shown in Eq. (4.11). The Fokker-Planck equation consistent with Eq. (4.11) can be straightforwardly found and its long-time asymptotic solution (while discarding terms of higher order than R^{-3}) can be determined analogously to LSW theory, as detailed in Appendix C.6. In doing so, we find that the critical radius evolves in time according to:

$$R_C(t) = \left[R_C^3(t_o) + \frac{6\rho^{\text{sat}}}{\zeta\Delta\rho^2}\gamma_{\text{ost}}Ct \right]^{1/3}, \quad (4.15)$$

where C is a dimensionless constant and t_o is an arbitrary time origin. We note that this long-time asymptotic solution also implies that the average radius is always within a dimensionless multiplicative constant of R_C [33, 114]. Equation (4.15) demonstrates that the $t^{1/3}$ growth law predicted by LSW theory should generically hold out of equilibrium while $\gamma_{\text{ost}} > 0$. This 1/3 power law has been measured experimentally in active systems [12, 16] with disparate microscopic details, suggesting that this growth law may in fact be robust. We note that the simulations of Redner *et al.* [110] measured the average number of particles per droplet (in two-dimensions) to grow slightly faster than $t^{1/2}$ at late times. This is consistent with a radial growth rate faster than $t^{1/4}$ and is perhaps consistent with our theoretical prediction.

Curiously, Ref. [110] also measured a much faster coarsening rate at early times where nuclei are spaced closely. While in this work we do not consider coarsening driven by nucleus collision, it is well-established for passive systems to also result in a $t^{1/3}$ growth law [129]. A transition from fast coarsening when nuclei are closely spaced to slow coarsening when nuclei are distant is eerily reminiscent of coarsening in systems with hydrodynamic interactions, in which collisions of nuclei (and subsequent relaxation to spherical geometry) induce flows that cause further collisions [130, 131]. While standard ABP models do not consider hydrodynamic interactions, a detailed accounting of the polarization field at the early stages of coarsening may reveal non-diffusive contributions. Furthermore, if ℓ_o greatly exceeds the spacing between nuclei, particles can be ballistically exchanged between nuclei, an effect that is also not accounted for in our perspective. A systematic treatment of early-stage active coarsening would help bridge the gap in our understanding between nucleation and late-stage coarsening and is the subject of future work.

We also find that Eqs. (4.6) may be used to treat coarsening problems with a finite phase fraction of minority phase, which is beyond the scope of LSW theory. To demonstrate this, we adopt the perspective and numerical scheme of Voorhees and Glicksman [40, 41], originally developed to study precipitation of solid phases in alloys. We now consider timescales at which the minority phase has nucleated and coarsened to the extent that the final phase fraction has been reached. All that remains to complete the phase separation process is for the disparate nuclei to coarsen to a single domain such that a single interface exists.

In the numerical scheme, the initial sizes and positions of nuclei in a fully periodic domain are prescribed. While the positions remain fixed, the sizes of the nuclei evolve with time via diffusion of particles through the majority phase. Additionally, the majority phase will relax much faster than the individual nuclei grow or shrink. We therefore consider the diffusive dynamics of *only the majority phase* (i.e., excluding the regions occupied by the nuclei) with the nuclei simply acting as particle generation terms (sources or sinks of particles). Considering only the majority phase dynamics implies that large density-gradient interfaces are ignored; we therefore discard gradient terms from Σ and arrive at the quasi-

stationary expression for the majority phase’s density field (see Appendix C.6 for further details):

$$0 = \nabla^2 \mathcal{P}(\rho) + \zeta \sum_{i=1}^{N_{\text{nuc}}} S_i(\mathbf{r}), \quad (4.16)$$

where N_{nuc} is the number of nuclei and $S_i(\mathbf{r})$ is a function quantifying its injection/absorption of particles into the majority phase. Using Eq. (4.16) to solve for the pressure field is analogous to solving for the electrostatic potential resulting from N_{nuc} charge densities. Approximating each nucleus as spherically symmetric allows one to treat the “charge densities” as “point charges”, i.e. $S_i(\mathbf{r}) \sim \delta(\mathbf{r} - \mathbf{r}_i)$. Then the effective diffusive interactions between nuclei, responsible for Ostwald ripening, can be calculated analogously to the electrostatic energy of a collection of point charges [40].

This perspective, while initially developed for passive systems, only relies on thermodynamic assumptions by employing an equilibrium Gibbs-Thomson relation as a boundary condition between each nucleus and the majority phase, allowing for the determination of the magnitude of each S_i . Our derived Eqs. (4.6) are the analogue of the Gibbs-Thomson relation for an active system, allowing us to extend the perspective of Ref. [40] to active matter.

For demonstration, we implement the active Voorhees-Glicksman theory to simulate the ripening of a periodic cell of active nuclei. Simulations of 250 nuclei with random positions [132] and sizes were executed with the nuclei separated by exceedingly large distances. It is therefore expected for the coarsening dynamics to be in agreement with LSW theory [33]. Noting again that the average radius is proportional to the critical radius, we expect the average radius across multiple activities to collapse to the same curve when scaled by the prefactor of the growth rate found in Eq. (4.15), $K \equiv (\rho^{\text{sat}} \gamma_{\text{ost}} d_{\text{hs}} / \zeta U_o \Delta \rho^2)^{1/3}$. The average nucleus size from these simulations scaled by K are shown in Fig. 4.5. Our numerical simulations reveal that the average nucleus size is in excellent agreement with the predicted $\sim t^{1/3}$ scaling with curves for both bubbles and droplets collapsing for all activities. Plots of the distribution of nucleus sizes, which display evidence of evolving towards an LSW distribution [33], are provided in Section 4 of the SM.

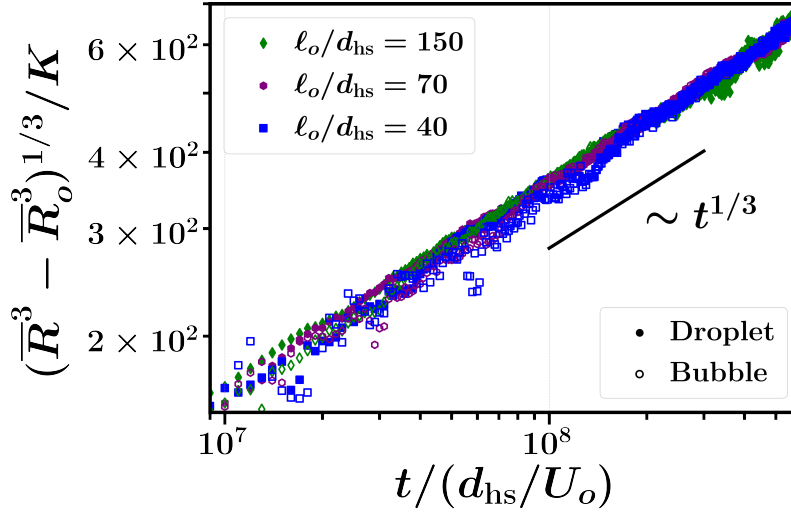


Figure 4.5: Average radius growth as a function of time for nuclei initialized with an LSW distribution and average radius $750d_{\text{hs}}$. Radius growth curves across different activities, as well as between bubbles and droplets, collapse onto one another when scaled by prefactor K .

Although the Voorhees-Glicksman theory can be conveniently applied to late-stage active coarsening dynamics via Eqs. (4.6), it has significant limitations. In particular, the positions of the nuclei are fixed in time while in reality nuclei may diffuse, which may have significant impact on the inter-nuclei diffusion interactions and in extreme cases may lead to collision of nuclei (coalescence). More sophisticated coarsening theories [113] attempting to account for these effects, such as the work of Enomoto *et al.* [133] also only draw on thermodynamic arguments in the form of a Gibbs-Thomson relation. Our work establishes a Gibbs-Thomson relation for single-component phase separation purely from mechanics, without invoking equilibrium assumptions. Then established perspectives on classical coarsening may be extended to nonequilibrium systems as long as a nonequilibrium Gibbs-Thomson relation, such as that found in this work, is used.

4.3 Conclusions

In this chapter, we derived a mechanical scheme for computing the properties of a critical nucleus arbitrarily far from equilibrium and apply this perspective to motility-induced phases comprised of active Brownian particles. From this theory we are able to derive generalized Gibbs-Thompson equations for active systems, a result which has been measured numerically but lacked theoretical justification [27]. Expanding on this theory, we use the framework proposed by Cates and Nardini [78] to find the Langevin dynamics of a nucleus. These dynamics allow us to define an effective nucleation barrier, revealing that the kinetic barrier to nucleating a droplet is substantially higher than that of a bubble. In addition, we find that the critical radius is an *unstable* fixed point for all activities where motility-induced phase separations occurs, demonstrating the nucleation dynamics of ABPs occur similarly to a passive system. By following the arguments first presented by Becker and Döring [32], we also determine the expected rate of nucleation for active droplets [76]. Our generalization of a Gibbs-Thompson relation to mechanical nonequilibrium systems allows a wide body of existing work studying the coarsening dynamics [33, 113] of passive systems to be applied to active matter. As an example, we demonstrated the applicability of Voorhees-Glicksman theory [40, 41] to active Brownian particles.

Our theory identifies physical implications of three independently defined surface tensions, shown in Fig. 4.4. The mechanical tension, γ_{mech} , which can be negative [24], controls the pressure difference between a nucleus and its surroundings *only* in the absence of non-local contributions to the dynamic stress. The capillary tension, γ_{cw} , establishes a surface area minimizing principle for an interface, which allows us to approximate fluctuating nuclei as spherically symmetric [42, 48, 78]. The Ostwald tension, γ_{ost} , plays a role in the finite-sized coexistence criteria in addition to controlling the dynamics of a nucleus [39, 78]. All three of these surface tensions are *identical* and *positive* for an equilibrium system but may take disparate, and even negative, values in active systems. Our results point to several new avenues for future work. One interesting direction would be to find rigorous connections between γ_{ost} and more palpable kinetic arguments such as those pro-

posed by Redner *et al.* [76, 110] and Lee [27], potentially aided by measurements from particle based simulations. Another would be to thoroughly investigate the physical implications of the nonlocal stress term $\mathcal{K}$ identified here, which may lead to macroscopic coexistence criteria explicitly dependent on interfacial properties.

The controversy surrounding a negative γ_{mech} has sparked a number of new definitions of surface tension in nonequilibrium systems. These methods have been rooted in a variety of perspectives, ranging from mechanical balances [29, 86] to dynamics [27, 48] to power functional theory [28]. Here, we argue that outside of equilibrium the surface tension must be defined contextually. While γ_{mech} and γ_{ost} were not found to enter the dynamics of capillary-waves [42], they are crucial for describing nucleation. Therefore one cannot haphazardly apply the definition of surface tension from one context to another; *the physically relevant surface tension must be derived from first principles*. While this work provides insight to the ongoing debates surrounding nonequilibrium surface tension, it may also serve as a starting point for other puzzles in active matter. This work could be extended to include multiple order parameters [64] in order to understand the nucleation of active crystals. Furthermore, applying the perspective of this work, in conjunction with the fluctuating hydrodynamics [42], may lead to identification of microscopic systems where γ_{ost} changes sign and novel nucleation dynamics are uncovered [39].

Nucleation events are intrinsically challenging to observe and characterize in simulation and experiment. Indeed, measuring the nucleation rates of equilibrium systems as simple as hard spheres has been a longstanding challenge [134–138]. Theories for nucleation are therefore essential for both guiding computational and experimental investigations and making predictions for conditions in which measurements can be prohibitive. To this end, CNT has provided crucial intuition for nucleation studies. It is our hope that this work, which makes use of a number of important recent developments [19, 39, 42, 48, 78], can provide a similar road map for studies of active nucleation by extending CNT and canonical coarsening theories to systems far from equilibrium.

Chapter 5

Thermal Noise, Hexatic Order, and Bubbles

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5.1 Introduction

The ubiquity of phase separation observed in environments driven far from equilibrium, in both synthetic and biological [7, 10, 12, 16, 23, 140, 141] contexts, has inspired theoretical efforts to understand purely non-equilibrium driving forces for phase transitions. One identified driving force is that of *self-propulsion*: motile particles with purely repulsive interactions can phase separate into dense and dilute regions at sufficient density and propulsive speed, a phenomenon known as motility induced phase separation (MIPS) [15]. Considerable recent work towards understanding MIPS has come from both modified Cahn-Hilliard frameworks [39, 51] as well as particle-based simulations [34, 55, 110]. These combined approaches have made significant progress towards understanding the non-equilibrium behavior of phase coexistence [19–21, 63, 142], capillary fluctuations [25, 42, 48], nucleation [76, 78, 108], and crystallization [143].

Despite this progress, the origin of the bubbles decorating the dense phase of phase separated active particles in two dimensions [34] remains an open question. The phenomenology of these bubbles resembles that predicted by Tjhung *et al.* [39] using a modified Cahn-Hilliard framework Active Model B+ (AMB+). In their minimal scalar field theory, they identified a range of parameters where the surface tension governing nucleation becomes negative. Under these conditions, AMB+ predicts a reversal of the Ostwald process resulting in a dense phase in which bubbles spontaneously form, coalesce, migrate to the boundaries of the dense phase, and merge with the surrounding vapor (liquid droplets dispersed in a vapor phase is also possible). The qualitative similarities between this prediction and the bubbles found in two-dimensional particle simulations [24, 34, 35, 110, 144] have led some to argue that the Ostwald process is reversed in active particles [37], although this has yet to be directly established.

Caporusso *et al.* [35] measured the distribution of bubble sizes in active particle simulations to be power-law distributed rather than peaked about a stable critical radius as one might naively assume a reversed Ostwald process implies. Further, they find that bubbles are present almost exclusively at the borders of hexatic domains, implying that the bubbles may originate from topological defects rather than

exotic coarsening dynamics. However, Fausti *et al.* [145] demonstrated through numerical integration of AMB+ that the distribution of bubble sizes can be anything between narrowly peaked or power-law decaying depending on the relative strengths of reversed Ostwald ripening, coalescence dynamics, and the nucleation rate. The numerical work of Shi *et al.* [38] investigated the system-size dependence of the distribution of bubbles in two-dimensional active particles, finding evidence that the bubbly fluid displays self-organized criticality (SOC). They also demonstrated that a reduced numerical model implementing reversed Ostwald ripening also displays SOC, further strengthening the argument that the Ostwald process is reversed in active particles. Recently, Yan *et al.* measured bubble *dynamics* [36] from particle simulations in addition to the size distribution. In particular, they fit a stochastic differential equation for the size dynamics of bubbles, finding that the drift term is area-independent and strictly negative, driving bubble collapse. While bubble dissolution in AMB+ under reversed Ostwald ripening is driven by bubble migration to the interface, Yan *et al.* found that 97% of bubbles in particle simulations spontaneously dissolve into the dense phase (even after accounting for coalescence) rather than escaping into the vapor. Finally, bubbles have not been observed in large-scale simulations of ABPs in three dimensions [34, 63]. The work on active nucleation of ABPs in Chapter 4 in three dimensions is consistent with these numerical findings [108].

Clearly, the origin of bubble formation in microscopic models of two-dimensional active particles remains an open question. In this Chapter, we present additional simulation data that further elucidates the dependencies of bubble formation. While previous simulation studies have focused on the dependence of the bubbles on activity, overall density, and system size, we focus on two other dependencies: polydispersity of particle size and thermalization (i.e., *translational* noise). Increasing polydispersity allows for the systematic elimination of hexatic order which was implicated in bubble formation in Ref. [35]. The reduction in hexatic order is indeed found to eliminate appreciable bubble formation. Intriguingly the presence of hexatic domains appears to be a necessary but insufficient condition for bubble formation. Translational thermal noise — while thought to play only a quantitative role in the phenomenology of MIPS — is also necessary for appreciable

bubble formation. Shockingly, we find that even a vanishingly small magnitude of thermalization is sufficient for the liquid to be populated with bubbles – only when translational noise is precisely zero do bubbles become scarce. These findings provide an increasingly complete portrait regarding the nature of bubbles in two-dimensional active matter.

5.2 Results and Discussion

Model System and Numerical Details

We conduct simulations of two-dimensional interacting active Brownian particles (ABPs) in which the time variation of the position $\mathbf{r}_i$ and (2D) orientation $\mathbf{q}_i$ of the i th particle follow overdamped Langevin equations:

$$\dot{\mathbf{r}}_i = U_o \mathbf{q}_i + \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij} + \boldsymbol{\eta}_i, \quad (5.1a)$$

$$\dot{\mathbf{q}}_i = \boldsymbol{\Omega}_i \times \mathbf{q}_i, \quad (5.1b)$$

where $\mathbf{F}_{ij}$ is a pairwise interaction force, U_o is the intrinsic active speed, ζ is the translational drag coefficient, $\boldsymbol{\eta}_i$ is a stochastic translational velocity with zero mean and variance $\langle \boldsymbol{\eta}_i(t) \boldsymbol{\eta}_j(t') \rangle = 2D_T \delta_{ij} \delta(t - t') \mathbf{I}$, and $\boldsymbol{\Omega}_i$ is a stochastic angular velocity. In two dimensions, the only component of $\boldsymbol{\Omega}_i$ relevant to the dynamics of $\mathbf{q}_i$ will be that of the out-of-plane direction, which we describe with unit vector $\mathbf{e}_z$. We can express $\boldsymbol{\Omega}_i$ as $\Omega_i \mathbf{e}_z$, where Ω_i has zero mean and variance $\langle \Omega_i(t) \Omega_j(t') \rangle = 2D_R \delta_{ij} \delta(t - t')$. Here, D_R and D_T are the rotational and translational diffusivity, respectively. The orientational relaxation time follows as $\tau_R \equiv D_R^{-1}$. Thus, the characteristic length scale that a non-interacting particle travels before reorienting is given by the intrinsic run length: $\ell_o \equiv U_o \tau_R$. The pairwise interaction forces are generated by a Weeks-Chandler-Anderson [124] potential with characteristic energy and length scales of ϵ and σ , respectively.

The relative strength of translational noise can be characterized by a dimensionless “thermalization” coefficient $\mathcal{T} \equiv D_T / (D_R \sigma^2)$. Existing simulation studies of

ABPs tend to either consider the “athermal” limit $\mathcal{T} = 0$ [29, 42, 63, 146] or $\mathcal{T} = 1/3$ [24, 34–36, 110, 147, 148], the latter of which is consistent with the three-dimensional Einstein relation [149] between thermal translation and rotation of an isolated sphere in a viscous medium. Throughout this Chapter, we will refer to simulations with $\mathcal{T} = 0$ as athermal and $\mathcal{T} > 0$ as thermal while stressing that D_T (and D_R) need not be thermal in origin. All of the simulations and theory in previous Chapters considered the $\mathcal{T} = 0$ athermal limit. We further emphasize that our particle dynamics remain stochastic and diffusive (at long times) in the athermal limit.

In order to ensure minimal overlap of disks while $\mathcal{T}$ is small, we set the characteristic interaction force to be much larger than the active force with the dimensionless stiffness set to $\mathcal{S} \equiv \epsilon/(\zeta U_o \sigma) = 50$ [63]. As detailed in Appendix D.1, our choice of a steep potential allows us to define an effective hard-sphere diameter $d = 2^{1/6} \sigma$ for systems in which the particles are monodisperse in size. We implement polydisperse simulations by radially shifting the WCA potential and can define the effective hard-disk diameter of the i th particle, d_i . We consider a Gaussian distribution of particle diameter with a mean $\langle d_i \rangle = d$ and a standard deviation of $0.09d$ (i.e., 9%). The area fraction occupied by our disks can then be defined as $\phi_o = \rho \pi \langle d_i^2 \rangle / 4$ where ρ is the areal number density.

Our system state is thus fully described by four dimensionless parameters: $\mathcal{T}$, ℓ_o/σ , $\mathcal{S}$, and ϕ_o in addition to the degree of polydispersity¹. All simulations are performed at a run length of $\ell_o/\sigma = 112.25$ (or $\ell_o/d = 100$), well beyond the critical activity of 2D MIPS [21, 142]. Monodisperse and polydisperse simulations were conducted at area fractions of $\phi_o = 0.64$ and $\phi_o = 0.49$, respectively². All simulations are performed using the HOOMD-Blue software package [65] and contain a total of 315844 particles.

¹We note that the particle activity can also be quantified by defining a Péclet number $\text{Pe} \equiv U_o \sigma / D_T$. We can express this Péclet via our dimensionless run length and thermalization coefficient: $\text{Pe} = \ell_o / (\mathcal{T} \sigma)$.

²The significance of this difference in overall area fractions, as well as simulation results for a thermal polydisperse simulation done at $\phi_o = 0.64$, is further discussed in the SM.

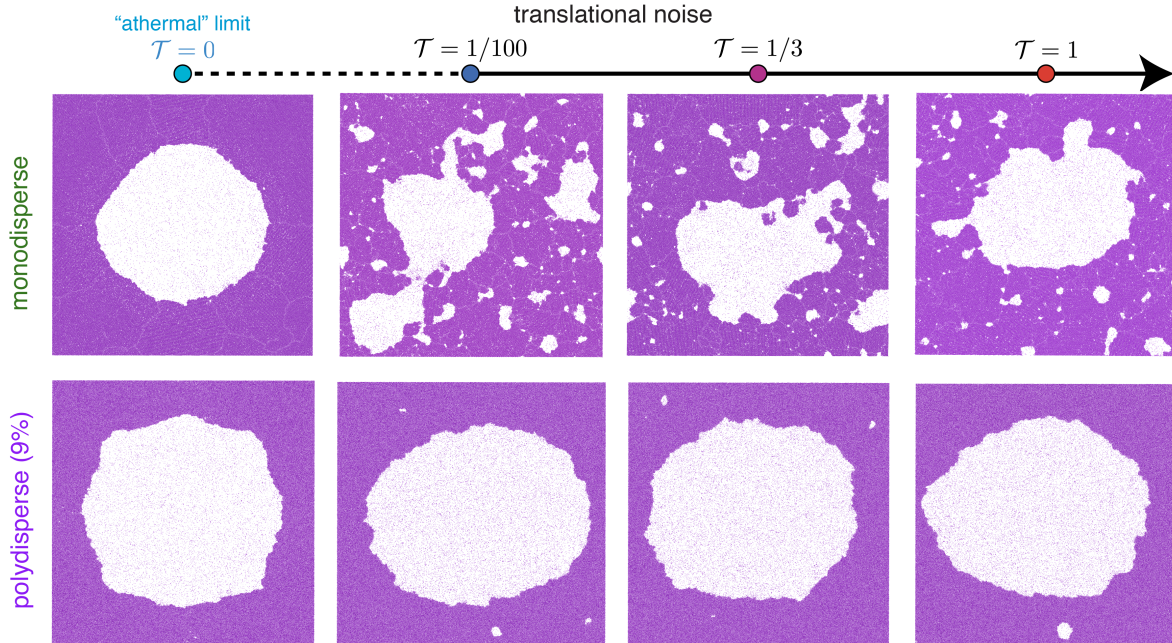


Figure 5.1: Representative snapshots of simulations at various values of thermalization for both monodisperse and polydisperse cases, rendered using Ovito [67]. Bubbles within the liquid phase were uncommon in all polydisperse and all athermal simulations.

Simulation Results

Representative snapshots of the steady-state configurations for both the monodisperse and polydisperse systems are shown in Fig. 5.1 as a function of $\mathcal{T}$. Consistent with previous works [34, 35, 38], the liquid phase in *monodisperse* and *thermal* systems is decorated with a broad distribution of bubble sizes. Intriguingly, and particularly for the smallest non-zero value of the thermal noise ($\mathcal{T} = 1/100$), the larger bubbles appear to be approaching the size of the vapor phase. This may suggest that the liquid in these systems might be in a state of self-organized criticality [38].

One may suspect that $\mathcal{T} = 1/100$ is sufficiently small that we have effectively reached the athermal limit in which translational noise plays a negligible role in the large-length scale phenomenology. However, setting the translational noise to be identically zero results in a dramatic change in the bubble distribution: bubbles

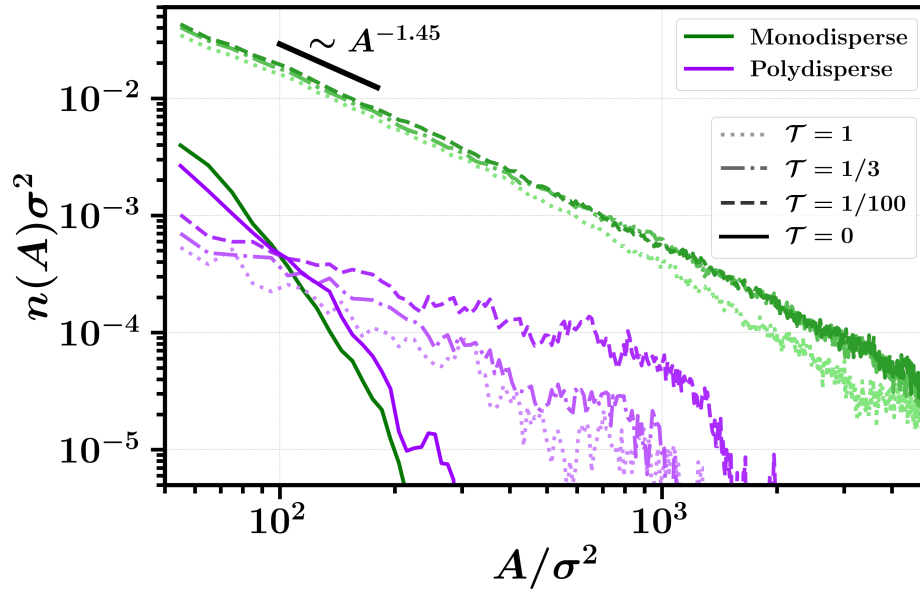


Figure 5.2: The dependence of $n(A)$ on thermal noise for both polydisperse and monodisperse systems.

become extraordinarily rare. We can quantify this dramatic change through the bubble distribution, $n(A)$, the average (over time in the steady state) number of bubbles of area A ³. The total number of bubbles (irrespective of size) in the system is thus given by $N_{\text{bub}} = \int_0^\infty n(A) dA$. We construct $n(A)$ by calculating the instantaneous coarse-grained density field to identify regions of space with gas phase density, then using the DBSCAN-enabled [150] procedure described in Ref. [35] to identify bubbles while ignoring the largest cluster [i.e., the bulk gas phase].

The sharp qualitative contrast between the thermal and athermal states are reflected in $n(A)$ [Fig. 5.2]. For the thermal monodisperse systems, we find an algebraic decay of $n(A) \sim A^{-1.45}$ that persists for over a decade of area [See Appendix D.4]. This decay cannot continue indefinitely, and eventually we observe a dramatic decrease in bubble number beyond a large cutoff area (see Appendix D.4). For the smallest bubble areas, the athermal system has an order of magnitude fewer

³ $n(A)$ is thus generally an extensive quantity that can be made intensive by, for example, dividing by the area of the liquid phase [38].

bubbles than the thermal systems. The bubble number for the athermal system sharply decays with area, further widening the difference with the thermal systems. Bubbles of sizes in excess of one hundred particle areas are essentially improbable for the athermal system while routinely observed for systems with finite translational noise.

The simulations by Caporusso et al. [35] found that bubbles are often observed at the grain boundaries of hexatic domains. To control for these effects, we now focus on polydisperse disks and show typical configurations of their steady state in Fig. 5.1. No bubble formation comparable to that of the thermal monodisperse system is observed for either the thermal or athermal polydisperse systems. $n(A)$ is nearly identical in the athermal polydisperse and monodisperse cases, with larger bubbles completely absent. As $\mathcal{T}$ is introduced, the polydisperse $n(A)$ broadens and larger bubbles are more likely to be observed, but remain uncommon.

Figure 5.3(a) displays the spatial correlation of the per-particle hexatic-order parameter (see Appendix D.4), confirming the absence of hexatic domains in our polydisperse systems, as anticipated. For the monodisperse system, hexatic correlations quantitatively increase in spatial range with decreasing translational noise but there is apparently no qualitative distinction. This difference in range can be visually appreciated in Figure 5.3 which colors the distinct domains. The size distribution can be readily computed for the thermal systems, and appears to be nearly independent of $\mathcal{T}$ [see Fig. 5.3(b)]. From this analysis, it appears unlikely that the structural differences between the athermal and thermal monodisperse systems are alone responsible for the strong disparity in their bubble statistics.

We now look to identify if there are qualitative *dynamical* distinctions which may delineate the thermal monodisperse systems from the athermal and polydisperse systems. Direct visualization of the particle dynamics indeed reveals a dependence of the dense-phase particle dynamics on the degree of thermalization and polydispersity (see Appendix D.6 for videos). In the athermal simulations, dynamics were sluggish and mediated by point vacancies (in the monodisperse case) or areas of lower density (in the polydisperse case). Monodisperse thermal simulations displayed striking collective motion, with individual hexatic domains often translating and reorienting as a group. Polydisperse thermal simulations

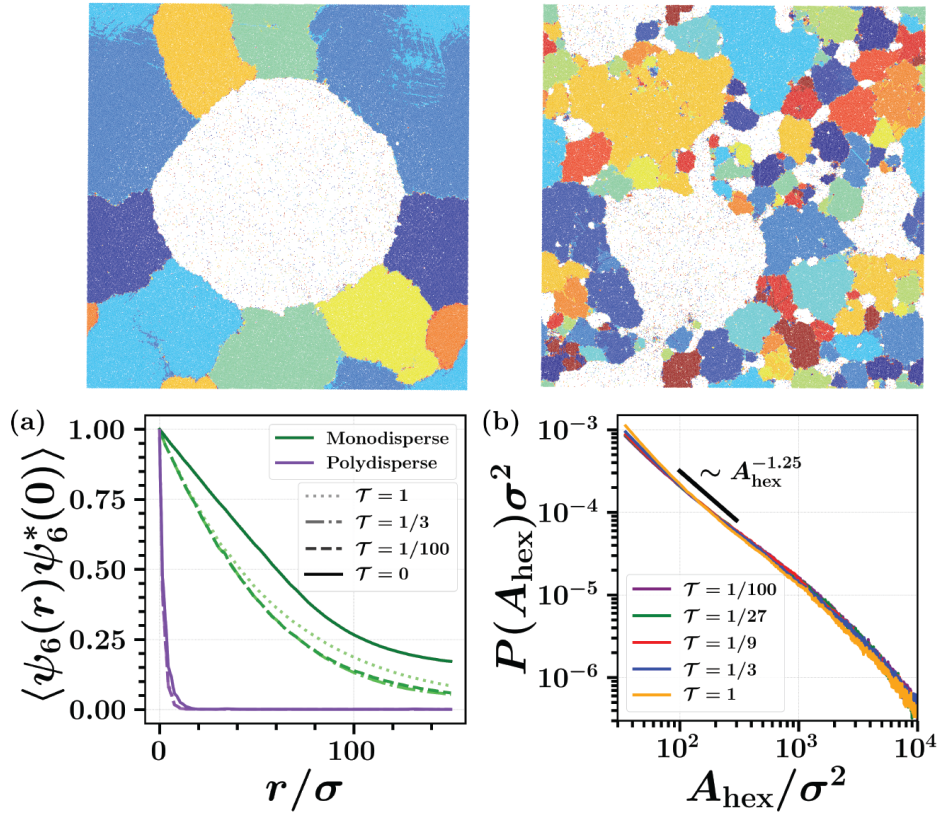


Figure 5.3: Top: Snapshots of $\mathcal{T} = 0$ (left) and $\mathcal{T} = 1/100$ (right) monodisperse simulations colored by hexatic domain orientation, rendered using Ovito [67]. (a) Hexatic order parameter spatial correlation functions. (b) Probability distribution of hexatic domain areas, A_{hex} , for thermalized monodisperse simulations. Polydisperse simulations not shown due to the lack of domains. Monodisperse $\mathcal{T} = 0$ not shown due to the sluggish kinetics and the presence of only a few large domains leading to poor statistics.

featured rapid transport of particles without noticeable collective effects. In order to quantify the collective motion observed in these videos, we calculate the spatial correlation function of the velocity orientation proposed by Caprini *et al.* [151–153]:

$$Q_i(r) = 1 - 2 \sum_j^{N_r} \frac{\theta_{ij}}{N_r \pi}, \quad (5.2)$$

where N_r is the number of particles in a shell of some thickness Δr centered at a distance r away from the i th particle and θ_{ij} is the angle between the velocities of particle i and particle j . Clearly, if the tagged particle velocity is perfectly aligned (anti-aligned) with those of the particles in the shell, we will recover $Q_i(r) = 1$ ($Q_i(r) = -1$). We can then calculate the average velocity orientation correlations $Q(r) = \sum_i Q_i(r)/N_{\text{dense}}$ by averaging Eq. (5.2) over all particles in the dense phase. The results of this calculation are shown in Fig. 5.4(a). We note that because all simulations are overdamped, the velocities used in this calculation are *average* velocities $\mathbf{v}_i(t) = (\mathbf{r}_i(t + \Delta t) - \mathbf{r}_i(t))/\Delta t$. The magnitude of $Q(r)$ therefore depends strongly on the choice of separation time Δt . We therefore calculate the correlations of the nearest-neighbor velocity orientations, $Q(r = 2^{1/6}\sigma)$, across a wide range of Δt (shown in Appendix D.4) and plot in Fig. 5.4(a) the $Q(r)$ corresponding to the Δt that maximizes nearest-neighbor correlations at each simulation condition. While $Q(r)$ allows us to quantify spatial dynamical correlations, we will characterize the rate of transport through the time dependence of the mean squared displacement (MSD), displayed in Fig. 5.4(b).

The spatial range of velocity correlations increases with thermal noise in both the monodisperse and polydisperse simulations. All monodisperse simulations had longer range velocity correlations, likely due to the longer range hexatic ordering. Any finite amount of thermalization, for both monodisperse and polydisperse simulations, increased the MSD of particles in the dense phase by multiple orders of magnitude. We note that our averaging procedure includes only dense-phase particles and excludes particles that escaped the dense phase during our observation window. This increase in both the range of dynamical correlations as well as the average speed of transport with $\mathcal{T}$ points to thermalization triggering some form

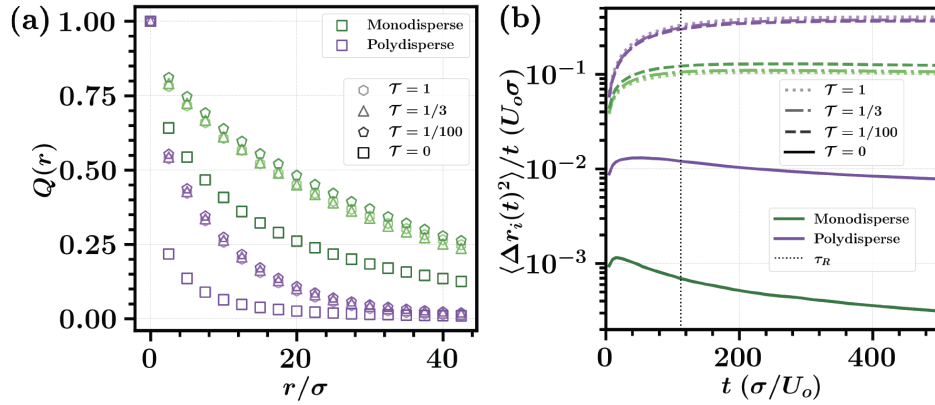


Figure 5.4: (a) Spatial correlations of velocity orientation obtained by averaging Eq. (5.2) over all particles, employing a time separation Δt that maximizes nearest-neighbor correlations. (b) Mean squared displacement of dense phase particles at each simulation condition, normalized by time. Diffusive dynamics should converge to a constant. Vertical dashed line corresponds to τ_R , the timescale beyond which we might expect diffusive dynamics.

of cooperative motion.

Discussion

The strong dependence of any measured quantity on thermalization is striking. Consider the relative magnitudes of terms in Eq. (5.1a). If we consider the effective diffusivity of an ideal ABP, it would be described by [17]:

$$D_{\text{ideal}} = \frac{U_o^2}{2D_R} + D_T = \frac{U_o \ell_o}{2} + \frac{\mathcal{T} \sigma^2 U_o}{\ell_o}. \quad (5.3)$$

Therefore, at the simulated run length of $\ell_o = 112.25\sigma$, the thermal contribution to the mobility of an ideal particle is negligible even as high as $\mathcal{T} = 1$. While it is true that the active contribution to this ideal diffusivity should be far lower in the dense phase of interacting ABPs, the dramatic increase in bubble fraction (see Table D.1 of Appendix D.4) and particle mobility with thermalization still exists at values of $\mathcal{T}$ as low as $1/100$. Capturing these effects in a coarse-grained or continuum-level model may warrant further investigation into cooperative effects that require

consideration of long-range correlations between particle dynamics that are not considered in existing efforts towards coarse-graining ABPs [108, 154].

Recently, Fausti *et al.* [145] argued that $n(A)$ should be understood as the net result of several processes including nucleation, ripening/coarsening, and coalescence. They also proposed a reduced model which allowed one to independently vary the relative strengths of coalescence and (reversed Ostwald) ripening while coupling the nucleation rate to the rate of coalescence. From these reduced models one can measure $n(A)$ to be strongly peaked at a particular area when reversed Ostwald ripening dominates or power-law distributed when coalescence/nucleation dominate. This reduced model may be a crucial tool for qualitative mechanistic insight on the $n(A)$ measured both in this study as well as previous studies [35, 36, 38]. Future work could focus on expanding these models to encompass a wider range of qualitative behaviors so that they are more readily applicable to interpreting simulation data. For example, the model reported in Ref. [145] has no mechanism for a bubble smaller than all of its neighbors to shrink in size, which is difficult to reconcile with the findings of Ref. [36] that the overwhelming majority of small bubbles dissolve into the dense phase. Other lines of expansion to the model may be to allow for forward Ostwald ripening or an uncoupling of the nucleation rate from the coalescence rate (while still maintaining mass conservation).

We can further isolate the importance of translational noise by defining $P(A)$, the probability distribution of any bubble being of size A . The size distribution can be found by normalizing $n(A)$ by its zeroth moment, N_{bub} . $P(A)$ [shown in Fig. 5.5] thus solely measures the size distribution, containing no information on the number of bubbles. Although the data remains noisy for systems in which bubbles were rare, all thermalized systems (including polydisperse) seem to display the same algebraic decay for small bubble sizes. This might suggest the possibility that the structural and dynamical distinctions between the polydisperse and monodisperse systems solely impact N_{bub} while giving rise to the same size distribution. Importantly, $P(A)$ for the *athermal* monodisperse and polydisperse systems does not collapse onto those of the thermal systems.

Hexatic order has a significant impact on the *number of bubbles* formed in the steady state but appears to be less decisive for the size distribution. This may

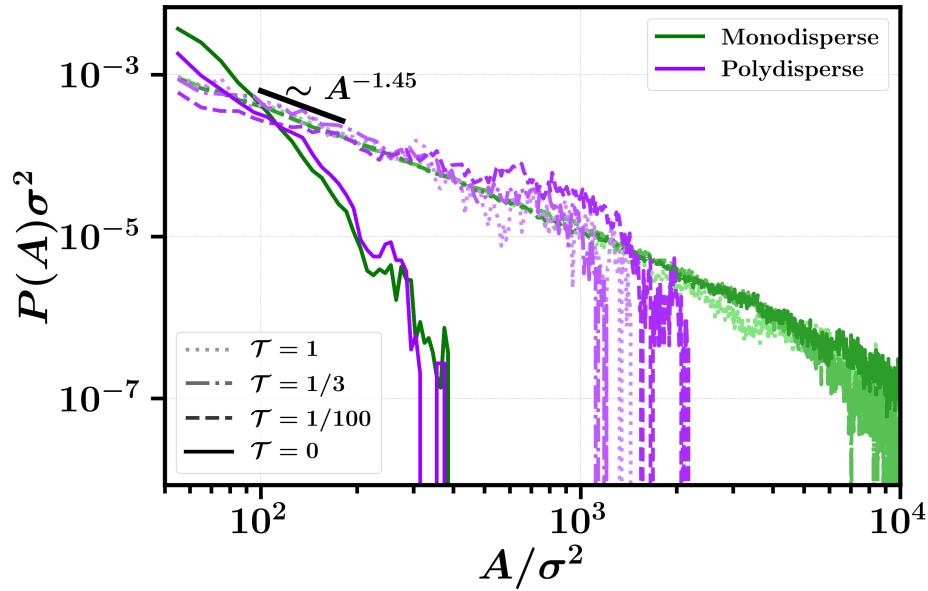


Figure 5.5: Probability distribution of observing a bubble of size A at various simulation conditions. Although the thermalized polydisperse systems are noisy due to the scarcity of bubbles, the scaling appears similar to the thermalized monodisperse systems.

explain the lack of bubbles in two-dimensional ABPs, since the liquid phase in three dimensions is disordered [63]. The apparent crucial role of hexatic order was first appreciated by Caporusso *et al.* [35], who identified that bubbles often appear at grain boundaries. It may be that these boundaries serve as preferred nucleation sites [37] in comparison to dense liquids (polydisperse) or ordered (monodisperse) domains. We note, however, that the fraction of gas phase within bubbles (see Appendix D.4) indicates that the number of observed bubbles decreases with $\mathcal{T}$ while $\mathcal{T} > 0$ (see also Fig. 5.1), despite the hexatic domain structure of monodisperse thermal simulations being indistinguishable (see Fig. 5.3).

5.3 Conclusions

In summary, we conducted simulations of phase-separated monodisperse and polydisperse ABPs at varying degrees of thermalization. We found that only monodisperse ABPs with finite (no matter how small) thermalization were well-endowed

with bubbles in the dense phase. We also found that thermalization resulted in dense phase dynamics that were both faster and longer-range in spatial correlations, suggesting that translational noise may induce cooperative motion. These findings present data that further complicate an ongoing debate regarding the origin of bubbles in two-dimensional active matter. Further work, both theoretical and numerical, is needed to understand the drastic dependence of both the bubble population and the dense phase dynamics in two-dimensional ABPs.

Chapter 6

Conclusions and outlook

This thesis directly addresses several open questions regarding active interfaces. As a prerequisite to addressing those questions, we presented a procedure for determining the fluctuating hydrodynamics of active Brownian particles. Using these fluctuating hydrodynamics, an equation of motion for the interface was derived and the resulting spectra of capillary-waves was found to be k^{-2} in agreement with large-scale Brownian dynamics simulation. Further, the interface was demonstrated to be stable despite the mechanically defined surface tension γ_{mech} being negative. This allowed for the definition of a new surface tension, γ_{cw} , which controls the stability and surface area minimization of interfaces. We then investigated the role of chirality in active interfaces by modifying the microscopic equation of motion for ABPs to include a deterministic torque that breaks spatial parity symmetry. A fluctuating hydrodynamics corresponding to these microscopic dynamics is then derived and an equation of motion for the interfacial dynamics is extracted. The interfacial dynamics of the chiral interface closely resemble those of the achiral dynamics, except for an imaginary term which generates flows in a particular direction determined by the chirality. The presence of these odd-surface flows are then verified via Brownian dynamics simulation. Nucleation was then investigated by first extending the coexistence theory of Ref. [21] to account for finite-size effects, thus generalizing the Gibbs-Thomson effect to equilibrium. Encoded within this finite-size coexistence theory is a physical meaning for the negative γ_{mech} : the interior of bubbles and droplets have a *lower* pressure than

their surroundings, reversed from equilibrium. The dynamics of nucleation were also investigated using the fluctuating hydrodynamics, and a nucleation barrier was identified. These results on nucleation were then applied to coarsening problems, and a $t^{1/3}$ coarsening rate was identified in agreement with equilibrium LSW theory. Further, another quantity with units of surface tension that reduced to γ_{mech} at equilibrium was identified. This quantity was found to control both the finite-sized coexistence as well as the nucleation barrier, and therefore became important for coarsening as well. This tension, which we labeled γ_{ost} , can be demonstrated to be equivalent to the pseudotension defined by AMB+ under certain assumptions. We found that γ_{ost} to be *positive* for all activities in three dimensional ABPs, consistent with numerical findings of standard nucleation and coarsening behavior in ABPs. Finally, we investigated the origin of the bubbles which decorate dense phases in two dimensional active matter. We found that hexatic order and thermalization are crucial for appreciable bubble formation. Bubbles were rarefied in athermal systems and polydisperse systems, while monodisperse systems with any finite degree of thermalization were populated with a large fraction of bubbles in the dense phase. Significant cooperative motion induced by thermalization was uncovered, and the magnitude of the mean squared displacements in the presense of any thermalization was found to increase by multiple orders of magnitude.

In the decade since Bialké *et al.* [24] reported a negative mechanical surface tension in ABPs, a plethora of studies have focused on resolving this apparent contradiction by *proposing* new definitions for surface tension [28–30, 155]. While such studies are well-motivated and frequently arrive at positive values, they rarely involve direct demonstration of the physical significance of these definitions. It is therefore unclear what contexts any of these proposed definitions are meaningful. The perspective of the work in this thesis has been to systematically derive the governing equations (e.g. finite-size coexistence criteria, interfacial fluctuations, etc.) of a particular non-equilibrium system, then identify the quantities playing the role that surface tension would take at equilibrium. The physical significance of those quantities is then unambiguous. This perspective is growing in popularity outside of our own work. Zakine and colleagues have made significant progress understanding the interface of active fluids with passive solids [106] and have

applied this understanding towards describing active wetting [156]. Indeed, their work on wetting derives the relevant tension from a statement of mechanical balance rather than proposing it. The resulting quantities accurately predict the three-phase contact angle between a passive solid wall, an active liquid, and an active gas.

It is therefore becoming increasingly clear that in general, different interfacial phenomena are described by different surface tensions. This distinguishes active interfaces from their equilibrium counterparts, and may complicate their experimental characterization. At equilibrium properties as diverse as the Laplace pressure difference, nucleation barrier, capillary-wave spectrum, and coarsening dynamics were all governed by a single quantity: the reversible work required to deform the interface. Away from equilibrium, the lack of a free energy to minimize implies that it is no longer guaranteed for all of these properties to be governed by the same quantity. As we find with ABPs, each distinct physical process where interfaces become relevant may be governed by a different physical quantity. One might argue that at equilibrium surface tension is a fundamental property due to the sheer number of physically relevant processes it governs. From a single measurement, one unlocks insight towards a plethora of thermodynamic and kinetic information. This work finds that no such advantage is guaranteed to exist in active systems: distinct physical processes are governed by different quantities which all reduce to γ_{mech} *only* at equilibrium. While this thesis was being written, it has come to our attention that recently Bureković *et al.* [157] found the following relationship between the γ_{mech} and γ_{ost} :

$$\gamma_{\text{mech}} = \frac{1}{\rho^{\text{liq}} - \rho^{\text{gas}}} \left[\rho^{\text{liq}} \gamma_{\text{ost}}^{\text{bubble}} - \rho^{\text{gas}} \gamma_{\text{ost}}^{\text{droplet}} \right]. \quad (6.1)$$

This relationship between the Ostwald tensions of droplets, bubbles can be confirmed numerically in Fig. 6.1. Relationships such as Eq. (6.1) reduce the total number of measurements required to characterize the interfacial behavior of non-equilibrium systems, and are therefore crucial results. Future work should not only derive the non-equilibrium surface tensions describing other notable interfacial phenomena (e.g. Marangoni flows, Rayleigh instabilities), but also determine whether relations between previously identified and newly identified tensions exist.

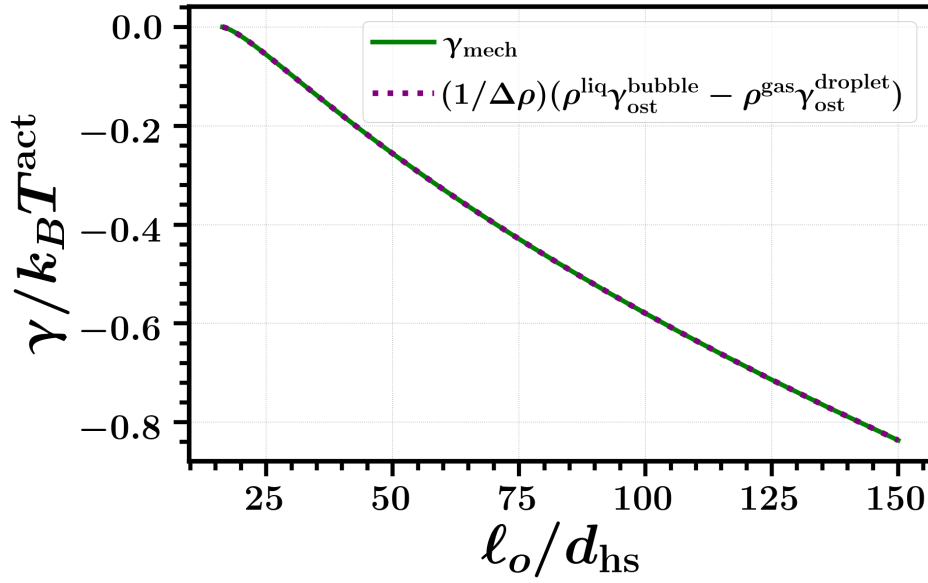


Figure 6.1: Numerical verification of Eq. (6.1).

Aside from our discussion of surface tension, this thesis provides generic tools for constructing models of active interfaces that forgo traditional thermodynamic assumptions. These tools, developed for achiral systems in mind, were able to be productively applied to chiral systems and predict new interfacial phenomena that were then confirmed within simulation. Thus, this thesis represents a step towards systematic understanding of interfacial phenomena far from equilibrium.

Appendix A

Appendix: Active Capillary Waves

The contents of Appendix A will closely follow that of the Supplemental Information of Ref. [42]. We will present the details of the derivation of the fluctuating hydrodynamics of ABPs, interfacial height fluctuations, and also discuss the details of molecular dynamics simulations used to measure the fluctuation spectra of 3d ABPs.

A.1 Fluctuating Hydrodynamics

Our derivation of the fluctuating hydrodynamics for active Brownian particles begins with the overdamped Langevin equation for the position $\mathbf{r}_i$ and orientation $\mathbf{q}_i$ of particle i :

$$\dot{\mathbf{r}}_i = U_o \mathbf{q}_i + \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij}, \quad (\text{A.1a})$$

$$\dot{\mathbf{q}}_i = \mathbf{q}_i \times \boldsymbol{\Omega}_i, \quad (\text{A.1b})$$

where $\dot{\mathbf{a}}$ denotes the time variation of $\mathbf{a}$, $\mathbf{F}_{ij}$ is the interparticle force, U_o is the intrinsic active speed, ζ is the translational drag coefficient, $\boldsymbol{\Omega}_i$ is a Brownian angular velocity with zero mean and a variance of $\langle \boldsymbol{\Omega}_i(t) \boldsymbol{\Omega}_j(t') \rangle = 2D_R \delta_{ij} \delta(t-t') \mathbf{I}$, and D_R is the rotational diffusivity. We note that, as presented, Eq. (A.1b) must be

interpreted in the Stratonovich convention to conserve the magnitude of $\mathbf{q}_i$ [61]. By adding the relevant drift term [42] we may convert Eq. (A.1b) to an equivalent statement interpreted with the Itô convention:

$$\dot{\mathbf{q}}_i = \mathbf{q}_i \times \boldsymbol{\Omega}_i - 2D_R \mathbf{q}_i. \quad (\text{A.1c})$$

We aim to describe the fluctuations of the coarse-grained density field, $\rho(\mathbf{r}; t) = \sum_{i=1}^N \Delta(\mathbf{r} - \mathbf{r}_i)$, where $\Delta(\mathbf{r} - \mathbf{r}_i)$ is a kernel of finite spatial width such that $\rho(\mathbf{r}; t)$ is spatially continuous. To do this, we first define the dynamical operator required to describe the time evolution of an observable (interpreted with the Itô convention), O , which can arbitrarily depend on the microscopic degrees of freedom, $(\mathbf{r}^N, \mathbf{q}^N)$ ¹:

$$\frac{\partial O}{\partial t} = \sum_{i=1}^N \left[\dot{\mathbf{r}}_i \cdot \frac{\partial O}{\partial \mathbf{r}_i} + \dot{\mathbf{q}}_i \cdot \frac{\partial O}{\partial \mathbf{q}_i} + \mathcal{I} \right]. \quad (\text{A.2})$$

$\mathcal{I}$ represents terms emerging from the the Itô chain rule:

$$\mathcal{I} = D_R \nabla_{\mathbf{q}_i}^2 O + 2D_R \mathbf{q}_i \cdot \frac{\partial O}{\partial \mathbf{q}_i}, \quad (\text{A.3})$$

where $\nabla_{\mathbf{q}_i} \equiv \mathbf{q}_i \times \partial / \partial \mathbf{q}_i$ is the rotational gradient operator. Substitution of Eq. (A.3) and Eq. (A.1c) into Eq. (A.2) results in:

$$\frac{\partial O}{\partial t} = \sum_{i=1}^N \left[\dot{\mathbf{r}}_i \cdot \frac{\partial O}{\partial \mathbf{r}_i} + D_R \nabla_{\mathbf{q}_i}^2 O + (\mathbf{q}_i \times \boldsymbol{\Omega}_i) \cdot \frac{\partial O}{\partial \mathbf{q}_i} \right]. \quad (\text{A.4})$$

We now define the dynamical operator $\mathcal{L}$ that evolves an arbitrary observable in time as:

$$\mathcal{L} = \sum_{i=1}^N \left[\underbrace{\dot{\mathbf{r}}_i \cdot \frac{\partial}{\partial \mathbf{r}_i} + D_R \nabla_{\mathbf{q}_i}^2}_{\text{deterministic}} + \underbrace{(\mathbf{q}_i \times \boldsymbol{\Omega}_i) \cdot \frac{\partial}{\partial \mathbf{q}_i}}_{\text{stochastic}} \right], \quad (\text{A.5})$$

¹We note that while we have denoted the time derivative O as a partial derivative, it is in fact a total derivative as the only time dependence of O is implicit through the microscopic degrees of freedom [6]. We choose to express it as a partial derivative in order to avoid confusion with a material derivative later on.

where we note that the first two terms are deterministic (i.e., solely depending on the microscopic configuration) while the last term is stochastic through the explicit dependence on Ω_i . This distinction allows us to further appreciate that the dynamical operator can be split into two contributions with:

$$\mathcal{L} = \mathcal{L}_{\text{FP}}^* + \mathcal{L}_S, \quad (\text{A.6a})$$

$$\mathcal{L}_{\text{FP}}^* = \sum_{i=1}^N \left[\dot{\mathbf{r}}_i \cdot \frac{\partial}{\partial \mathbf{r}_i} + D_R \nabla_{\mathbf{q}_i}^2 \right], \quad (\text{A.6b})$$

$$\mathcal{L}_S = \sum_{i=1}^N \left[(\mathbf{q}_i \times \Omega_i) \cdot \frac{\partial}{\partial \mathbf{q}_i} \right], \quad (\text{A.6c})$$

where we have recognized that the deterministic portion of the operator is *precisely* the adjoint of the Fokker-Planck operator for ABPs (see Ref. [21]). The dynamics of an arbitrary observable are thus given by:

$$\frac{\partial}{\partial t} O = \mathcal{L} O = \mathcal{L}_{\text{FP}}^* O + \mathcal{L}_S O. \quad (\text{A.7})$$

In what follows, we will proceed to derive the dynamics of the density and its one-body orientational moments as derived by Ref. [21]. There, exact expressions for $\mathcal{L}_{\text{FP}}^* O$ were derived and ultimately an expectation over the noise-averaged N -body distribution was performed. The latter operation eliminated the need to evaluate $\mathcal{L}_S O$ which vanishes upon averaging over the noise distribution. Here, for completeness, we will recapitulate the expressions found in Ref. [21] for $\mathcal{L}_{\text{FP}}^* O^2$ while now also determining the stochastic contributions to our field variables.

The primary field variable of interest is the density, which acts as the unambiguous order parameter in three dimensional motility-induced phase separation [63]. We will first focus on the *exact* dynamics of the coarse-grained density field before

²We note that, in this work, we are considering the dynamics of coarse-grained variables while Ref. [21] examined the dynamics of microscopic variables (averaged over the distribution). The use of coarse-grained field variables, however, will simply introduce the coarse-graining kernel in place of the Dirac delta function employed in the definition of microscopic field variables.

introducing the closures and approximations necessary to ultimately describe these dynamics solely in terms of the density field itself.

The coarse-grained density at position $\mathbf{r}$ is defined as:

$$\rho(\mathbf{r}, t) = \sum_{i=1}^N \Delta(\mathbf{r} - \mathbf{r}_i). \quad (\text{A.8})$$

Substituting $O = \rho$ into Eq. (A.7) results in simply $\partial\rho/\partial t = \mathcal{L}_{\text{FP}}^* \rho$ (and is thus fully derived in Ref. [21]) as the density contains no explicit dependence on the particle orientations. The exact evolution equation for the coarse-grained density follows as:

$$\frac{\partial\rho}{\partial t} = -\nabla \cdot \mathbf{J}, \quad (\text{A.9a})$$

where $\mathbf{J} = \sum_{i=1}^N \dot{\mathbf{r}}_i \Delta(\mathbf{r} - \mathbf{r}_i)$ is the flux of density and is given by:

$$\mathbf{J} = U_o \mathbf{m} + \frac{1}{\zeta} \nabla \cdot \boldsymbol{\sigma}^C, \quad (\text{A.9b})$$

where $\mathbf{m} = \sum_{i=1}^N \mathbf{q}_i \Delta(\mathbf{r} - \mathbf{r}_i)$ and $\boldsymbol{\sigma}^C$ are, respectively, the polar order and conservative interaction stress. For the pairwise interactions under consideration in this work, the interaction stress is:

$$\boldsymbol{\sigma}^C = -\frac{1}{2} \sum_{i=1}^N \sum_{j \neq i}^N \mathbf{F}_{ij} \mathbf{r}_{ij} b_{ij}, \quad (\text{A.10})$$

where $b_{ij}(\mathbf{r}; \mathbf{r}_i, \mathbf{r}_j) = \int_0^1 \Delta(\mathbf{r} - \mathbf{r}_j - \lambda \mathbf{r}_{ij}) d\lambda$ is the bond function [121, 122, 158, 159] and $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$.

The density flux [Eq. (A.9b)] naturally introduces the polar order field, the dynamics of which we now consider. The time evolution of the polarization is given by $\partial\mathbf{m}/\partial t = \mathcal{L}_{\text{FP}}^* \mathbf{m} + \mathcal{L}_S \mathbf{m}$ where we now must consider the stochastic operator. The resulting evolution equation follows as:

$$\frac{\partial\mathbf{m}}{\partial t} = -\nabla \cdot \mathbf{J}^{\mathbf{m}} - (d-1)D_R \mathbf{m} + \boldsymbol{\eta}^{\mathbf{m}}, \quad (\text{A.11a})$$

where we have defined a polarization noise vector as $\boldsymbol{\eta}^{\mathbf{m}} = \mathcal{L}_S \mathbf{m}$. The polarization flux $\mathbf{J}^{\mathbf{m}} = \sum_{i=1}^N \dot{\mathbf{r}}_i \mathbf{q}_i \Delta(\mathbf{r} - \mathbf{r}_i)$ (a second rank tensor) is given by:

$$\mathbf{J}^{\mathbf{m}} = U_o \mathbf{Q} + \frac{1}{\zeta} \boldsymbol{\kappa}^{\mathbf{m}} + \frac{1}{\zeta} \boldsymbol{\nabla} \cdot \boldsymbol{\Sigma}^{\mathbf{m}}. \quad (\text{A.11b})$$

The polarization flux consists of three contributions: convection at the ideal active speed by the nematic density tensor $\mathbf{Q} = \sum_{i=1}^N \mathbf{q}_i \mathbf{q}_i \Delta(\mathbf{r} - \mathbf{r}_i)$, forcing from a “body-force-like” term $\boldsymbol{\kappa}^{\mathbf{m}}$ (dimensions of force density) and forcing from a “stress-like” term $\boldsymbol{\Sigma}^{\mathbf{m}}$ (dimensions of stress). These terms emerge from the Fokker-Planck portion of the operator and are defined as:

$$\boldsymbol{\kappa}^{\mathbf{m}} = \frac{1}{2} \sum_i^N \sum_{j \neq i}^N \mathbf{F}_{ij} (\mathbf{q}_i - \mathbf{q}_j) b_{ij}, \quad (\text{A.12a})$$

$$\boldsymbol{\Sigma}^{\mathbf{m}} = -\frac{1}{2} \sum_i^N \sum_{j \neq i}^N \mathbf{r}_{ij} \mathbf{F}_{ij} \mathbf{c}_{ij}, \quad (\text{A.12b})$$

where $\mathbf{c}_{ij} = \int_0^1 d\lambda (\mathbf{q}_j - \lambda \mathbf{q}_{ij}) \Delta(\mathbf{r} - \mathbf{r}_j - \lambda \mathbf{r}_{ij})$ and $\mathbf{q}_{ij} = \mathbf{q}_i - \mathbf{q}_j$. The physical interpretation of these terms are discussed in Ref. [21]. Here, we note that $\boldsymbol{\kappa}^{\mathbf{m}}$ vanishes for two particles with the same orientation and is maximal for two particles with *opposite* orientation. One can thus interpret $\boldsymbol{\kappa}^{\mathbf{m}}$ as a force that resists the nematic convection and reduces the effective convective speed. While the first two terms in Eq. (A.11b) are associated with convection, the final term, captures the flux of polar generated by interactions across surfaces. Constitutive equations for Eq. (A.12) will later be introduced.

We now consider the stochastic term, $\boldsymbol{\eta}^{\mathbf{m}}$, appearing in Eq. (A.11a). Applying the stochastic operator on the polarization results in an expression for the polarization noise with:

$$\boldsymbol{\eta}^{\mathbf{m}} = \sum_{i=1}^N \mathbf{q}_i \times \boldsymbol{\Omega}_i \Delta(\mathbf{r} - \mathbf{r}_i). \quad (\text{A.13})$$

We can immediately recognize that this noise is nonlocal in space for finite coarse-graining width. Furthermore, describing the variance of this noise in terms of field variables is also not readily possible. An approximation that allows this noise to be expressed solely in terms of the one-body orientational fields will be later discussed.

The time evolution of the nematic order is now required. Again, like the polar order (and unlike the density), the stochastic contribution cannot be neglected. The nematic dynamics are found to be:

$$\frac{\partial \mathbf{Q}}{\partial t} = -\nabla \cdot \mathbf{J}^{\mathbf{Q}} - 2dD_R \left(\mathbf{Q} - \frac{1}{d}\rho \mathbf{I} \right) + \boldsymbol{\eta}^{\mathbf{Q}}, \quad (\text{A.14a})$$

where we have defined a polarization noise tensor as $\boldsymbol{\eta}^{\mathbf{Q}} = \mathcal{L}_S \mathbf{Q}$. The nematic flux $\mathbf{J}^{\mathbf{Q}} = \sum_{i=1}^N \dot{\mathbf{r}}_i \mathbf{q}_i \mathbf{q}_i \Delta(\mathbf{r} - \mathbf{r}_i)$ (a third rank tensor) is given by:

$$\mathbf{J}^{\mathbf{Q}} = U_o \mathbf{B} + \frac{1}{\zeta} \boldsymbol{\kappa}^{\mathbf{Q}} + \frac{1}{\zeta} \nabla \cdot \boldsymbol{\Sigma}^{\mathbf{Q}}. \quad (\text{A.14b})$$

Just as in the case of the polar order flux, the nematic flux consists of three contributions: convection at the ideal active speed by $\mathbf{B} = \sum_{i=1}^N \mathbf{q}_i \mathbf{q}_i \mathbf{q}_i \Delta(\mathbf{r} - \mathbf{r}_i)$, forcing from a “body-force-like” term $\boldsymbol{\kappa}^{\mathbf{Q}}$ (dimensions of force density) and forcing from a “stress-like” term $\boldsymbol{\Sigma}^{\mathbf{Q}}$ (dimensions of stress). Expressions for the latter two contributions have the following form:

$$\boldsymbol{\kappa}^{\mathbf{Q}} = \frac{1}{2} \sum_i^N \sum_{j \neq i}^N \mathbf{F}_{ij} (\mathbf{q}_i \mathbf{q}_i - \mathbf{q}_j \mathbf{q}_j) b_{ij}, \quad (\text{A.15a})$$

$$\boldsymbol{\Sigma}^{\mathbf{Q}} = -\frac{1}{2} \sum_i^N \sum_{j \neq i}^N \mathbf{r}_{ij} \mathbf{F}_{ij} \mathbf{d}_{ij}, \quad (\text{A.15b})$$

where $\mathbf{d}_{ij} = \int_0^1 d\lambda (\mathbf{q}_i \mathbf{q}_i - \lambda (\mathbf{q}_i \mathbf{q}_i - \mathbf{q}_j \mathbf{q}_j)) \Delta(\mathbf{r} - \mathbf{r}_j - \lambda \mathbf{r}_{ij})$. Despite the change of the tensorial rank of these quantities from those found in the polar order, the physical interpretations remains the same. The nematic noise vector takes the

following form:

$$\boldsymbol{\eta}^{\mathbf{Q}} = \sum_{i=1}^N 2\mathbf{q}_i \times \boldsymbol{\Omega}_i \mathbf{q}_i \Delta(\mathbf{r} - \mathbf{r}_i), \quad (\text{A.16})$$

and, as with $\boldsymbol{\eta}^{\mathbf{m}}$, we will later find approximations for the statistics of $\boldsymbol{\eta}^{\mathbf{Q}}$.

We can similarly obtain an evolution equation for the *traceless* nematic order, defined as:

$$\mathbf{Q}' = \mathbf{Q} - \frac{\rho}{d} \mathbf{I}. \quad (\text{A.17a})$$

Inserting this definition into Eq. (A.14) results in:

$$\frac{\partial \mathbf{Q}'}{\partial t} = -\boldsymbol{\nabla} \cdot \mathbf{J}^{\mathbf{Q}'} - 2dD_R \mathbf{Q}' + \boldsymbol{\eta}^{\mathbf{Q}}, \quad (\text{A.17b})$$

where the traceless nematic field flux is simply $\mathbf{J}^{\mathbf{Q}'} = \mathbf{J}^{\mathbf{Q}} - \frac{1}{d} \mathbf{J} \mathbf{I}$ with:

$$\mathbf{J}^{\mathbf{Q}'} = U_o \mathbf{B} + \frac{1}{\zeta} \boldsymbol{\kappa}^{\mathbf{Q}} - \frac{1}{d} \mathbf{J} \mathbf{I} + \frac{1}{\zeta} \boldsymbol{\nabla} \cdot \boldsymbol{\Sigma}^{\mathbf{Q}}. \quad (\text{A.17c})$$

In principle we could continue deriving the evolution equation of $\mathbf{B}$ and the infinite hierarchy of one-body orientation fields. However, obtaining the *linear* height field dynamics of the interface does not require the consideration of orientation fields beyond the nematic tensor. We thus conclude our derivation of the coarse-grained density dynamics of athermal ABPs and now begin to simplify our expressions with approximations consistent with obtaining a long wavelength capillary theory.

Our aim is to derive to stochastic height field dynamics associated with ABP interfaces. Doing so requires a stochastic description of the field variable used to define the location of the interface. In our case, this is the coarse-grained density. Above, we derived the *exact* stochastic dynamics of the coarse-grained density field and, in doing so, found that the density field dynamics are coupled to the dynamics of the other one-body orientational fields. An exact description of the height field dynamics would require solving the coupled dynamics of the density, polarization and nematic order *and* an ansatz that introduces less error than that

of Bray’s [49] [Eq. (2.4)]. However, as we are interested in obtaining a theory for long wavelength capillary fluctuations, we will make use of Bray’s ansatz and other simplifying approximations in our fluctuating hydrodynamics consistent with this aim. These assumptions reduce our fluctuating hydrodynamic description of the density field to *solely* consist of a single field variable, the density field itself. This proves convenient in the derivation of the interfacial dynamics, as a single field was assumed in Bray’s treatment and more recently by Fausti *et al.* [48].

We first assume that the relaxation dynamics of the polar and nematic order are faster than those of the density field. The faster relaxation dynamics of higher order one-body orientational moments can be appreciated from the conservation equations derived above. Comparison of Eqs. (A.11a) and (A.17b) in the absence of a spatial gradient reveals that the nematic order will exhibit a temporal decay that is a factor of $2d/(d-1)$ faster than that of the polar order. We neglect the time variation of both the polar and nematic order in Eqs. (A.11a) and (A.17b), respectively, as they are anticipated to be faster than those of the density field. Doing so allows us to identify that the polar order can be expressed as $\mathbf{m} = \frac{\tau_R}{d-1} (\nabla \cdot \mathbf{J}^{\mathbf{m}} - \eta^{\mathbf{m}})$. Substitution of this into the density flux [Eq. (A.11b)] allows us to define several key mechanical terms:

$$\mathbf{J} = \frac{1}{\zeta} \nabla \cdot \Sigma + \frac{\ell_o}{d-1} \eta^{\mathbf{m}}, \quad (\text{A.18a})$$

where we have defined the *dynamic* stress tensor:

$$\Sigma = \sigma^{\text{act}} + \sigma^C, \quad (\text{A.18b})$$

recognizing that the body force generated by the polarization appears to take the form of an *effective* stress upon neglecting the temporal variation of the polar order. This effective active (or “swim” [17]) stress is defined as [29, 56]:

$$\sigma^{\text{act}} = \frac{\zeta \ell_o}{d-1} \mathbf{J}^{\mathbf{m}}. \quad (\text{A.18c})$$

We now find the statistics of $\eta^{\mathbf{m}}$, which is given by:

$$\eta^{\mathbf{m}} = \sum_{i=1}^N \mathbf{q}_i \times \Omega_i \delta(\mathbf{r} - \mathbf{r}_i). \quad (\text{A.19})$$

While the mean is clearly zero, the variance is given by:

$$\langle \boldsymbol{\eta}^{\mathbf{m}}(\mathbf{r}, t) \boldsymbol{\eta}^{\mathbf{m}}(\mathbf{r}', t') \rangle = \sum_{i=1}^N \sum_{j=1}^N \langle (\mathbf{q}_i \times \boldsymbol{\Omega}_i)(\mathbf{q}_j \times \boldsymbol{\Omega}_j) \rangle \Delta(\mathbf{r} - \mathbf{r}_i) \Delta(\mathbf{r}' - \mathbf{r}_j), \quad (\text{A.20})$$

where $\langle \dots \rangle$ denotes an expectation over the distribution of the stochastic angular velocities. Straightforward manipulation allows us to express $\langle (\mathbf{q}_i \times \boldsymbol{\Omega}_i)(\mathbf{q}_j \times \boldsymbol{\Omega}_j) \rangle = 2D_R [(\mathbf{q}_i \cdot \mathbf{q}_j) \mathbf{I} - \mathbf{q}_i \mathbf{q}_j] \delta_{ij} \delta(t - t')$, substitution of which into Eq. (A.20) results in:

$$\langle \boldsymbol{\eta}^{\mathbf{m}}(\mathbf{r}, t) \boldsymbol{\eta}^{\mathbf{m}}(\mathbf{r}', t') \rangle = \sum_{i=1}^N 2D_R (\mathbf{I} - \mathbf{q}_i \mathbf{q}_i) \Delta(\mathbf{r} - \mathbf{r}_i) \Delta(\mathbf{r}' - \mathbf{r}_i) \delta(t - t'), \quad (\text{A.21})$$

where the independence of the variance of angular velocities between different particles has eliminated one of the particle summations and ensured that the noise is a one-body property. We now approximate the statistics of $\boldsymbol{\eta}^{\mathbf{m}}$ with the statistics of $\hat{\boldsymbol{\eta}}^{\mathbf{m}}$, which is the noise associated with the *microscopic* polar order flux and has a variance of:

$$\langle \hat{\boldsymbol{\eta}}^{\mathbf{m}}(\mathbf{r}, t) \hat{\boldsymbol{\eta}}^{\mathbf{m}}(\mathbf{r}', t') \rangle = \sum_{i=1}^N 2D_R (\mathbf{I} - \mathbf{q}_i \mathbf{q}_i) \delta(\mathbf{r} - \mathbf{r}_i) \delta(\mathbf{r}' - \mathbf{r}_i) \delta(t - t'). \quad (\text{A.22})$$

Using the identity $\delta(\mathbf{r} - \mathbf{r}_i) \delta(\mathbf{r}' - \mathbf{r}_i) = \delta(\mathbf{r} - \mathbf{r}') \delta(\mathbf{r} - \mathbf{r}_i)$, which is the only component of our approximation that cannot be applied to arbitrary Δ , and invoking the definitions of the microscopic density and nematic order, we arrive at our final expression for the microscopic polarization noise statistics:

$$\langle \hat{\boldsymbol{\eta}}^{\mathbf{m}}(\mathbf{r}, t) \rangle = \mathbf{0}, \quad (\text{A.23a})$$

$$\langle \hat{\boldsymbol{\eta}}^{\mathbf{m}}(\mathbf{r}, t) \hat{\boldsymbol{\eta}}^{\mathbf{m}}(\mathbf{r}', t') \rangle = 2D_R \left(\hat{\rho} \mathbf{I} - \hat{\mathbf{Q}} \right) \delta(\mathbf{r} - \mathbf{r}') \delta(t - t'), \quad (\text{A.23b})$$

where $\hat{\rho}$ and $\hat{\mathbf{Q}}$ can be evaluated at $\mathbf{r}$ or $\mathbf{r}'$. This stochastic polarization term is identical to that found by Cugliandolo *et al.* [61] in describing the fluctuating

hydrodynamics of passive dipoles with the only physical distinction being the (generally) athermal origins of D_R for active systems. By approximating the statistics of $\boldsymbol{\eta}^{\mathbf{m}}$ with the statistics of $\hat{\boldsymbol{\eta}}^{\mathbf{m}}$ we find:

$$\langle \boldsymbol{\eta}^{\mathbf{m}}(\mathbf{r}, t) \rangle = \mathbf{0}, \quad (\text{A.24a})$$

$$\langle \boldsymbol{\eta}^{\mathbf{m}}(\mathbf{r}, t) \boldsymbol{\eta}^{\mathbf{m}}(\mathbf{r}', t') \rangle = 2D_R (\rho \mathbf{I} - \mathbf{Q}) \delta(\mathbf{r} - \mathbf{r}') \delta(t - t'), \quad (\text{A.24b})$$

Furthermore, by absorbing the factor of $\ell_o/(d-1)$ present in Eq. (A.18a) into $\boldsymbol{\eta}^{\mathbf{m}}$, we define $\boldsymbol{\eta}^{\text{act}} = \ell_o/(d-1) \boldsymbol{\eta}^{\mathbf{m}}$ which has statistics following directly from those of the polar order noise [Eq. (A.24)]:

$$\langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \rangle = \mathbf{0}, \quad (\text{A.25a})$$

$$\langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \boldsymbol{\eta}^{\text{act}}(\mathbf{r}', t') \rangle = 2 \frac{k_B T^{\text{act}}}{\zeta} \left(\rho \mathbf{I} - \frac{d}{d-1} \mathbf{Q}' \right) \delta(t - t') \delta(\mathbf{r} - \mathbf{r}'), \quad (\text{A.25b})$$

where we have defined the active energy scale as $k_B T^{\text{act}} \equiv \ell_o \zeta U_o / d(d-1)$. The statistics of $\boldsymbol{\eta}^{\mathbf{Q}}$ can be approximated analogously to those of the polarization flux with:

$$\langle \boldsymbol{\eta}^{\mathbf{Q}}(\mathbf{r}, t) \rangle = \mathbf{0}, \quad (\text{A.26a})$$

$$\langle \boldsymbol{\eta}^{\mathbf{Q}}(\mathbf{r}, t) \boldsymbol{\eta}^{\mathbf{Q}}(\mathbf{r}', t') \rangle = 8D_R (\mathbf{Q} \mathbf{I} - \mathbf{C}) \delta(\mathbf{r} - \mathbf{r}') \delta(t - t'), \quad (\text{A.26b})$$

where $\mathbf{C} = \sum_{i=1}^N \mathbf{q}_i \mathbf{q}_i \mathbf{q}_i \mathbf{q}_i \Delta(\mathbf{r} - \mathbf{r}_i)$.

At this point, the unknown quantities required to describe our stochastic density flux are constitutive relations for the conservative stress [Eq. (A.10)] as well as the body-force-like and stress-like terms [Eqs. (A.12) and (A.15)] and the remaining one-body orientational moments, $\mathbf{Q}'$, $\mathbf{B}$ and $\mathbf{C}$. Before proposing constitutive equations and closure relations we recall that predicting states of coexistence *requires* the retention of terms that are at least third order in spatial gradient of the density in our flux expression [Eq. (A.18a)] and thus second order in our dynamic stress [Eq. (A.18b)].

We first focus on the required constitutive equations, beginning with the conservative stress. In the case of a homogeneous density, the conservative stress would

simply return the isotropic interaction pressure, $\sigma^C = -p_C \mathbf{I}$, an equation of state that depends on the bulk density and activity. A general second-order gradient expansion of the conservative stress has the form:

$$\sigma^C = [-p_C + \kappa_1 \nabla^2 \rho + \kappa_2 |\nabla \rho|^2] \mathbf{I} + \kappa_3 \nabla \rho \nabla \rho + \kappa_4 \nabla \nabla \rho, \quad (\text{A.27})$$

where the linear term vanishes due to inversion symmetry. In the reversible limit, which occurs as $\ell_o \rightarrow 0$, the coefficients $\{\kappa_i\}$ take the form of the Korteweg stress [43, 44, 57] as noted in the main text. We forgo providing general microscopic expressions for $\{\kappa_i\}$ that are valid for all activities as, for active hard spheres, there is likely only a narrow region of activity where these coefficients both depart significantly from the reversible Korteweg stress and are comparable in scale to the gradient terms that will arise from the active stress.

The physical interpretation of the body-force-like terms is clear: they arise from interparticle interactions and *oppose* free convection at the intrinsic active speed U_o . Following Ref. [21], we therefore propose the following constitutive relation for κ^m :

$$\kappa^m = -\zeta (U_o - U) \mathbf{Q}, \quad (\text{A.28})$$

where U is the *effective* active speed and is bound between 0 and U_o . We adopt a similar constitutive equation for the body-force-like term appearing in the nematic flux with:

$$\kappa^Q = -\zeta (U_o - U) \mathbf{B}, \quad (\text{A.29})$$

where we have assumed that the same effective active speed appears here as in Eq. (A.28). The polarization flux also requires a constitutive relation for the stress-like term, Σ^m , which cannot be *a priori* discarded on the basis of spatial gradients. However, here we neglect this contribution as it was shown to have a marginal quantitative impact on the ABP hard sphere/disk phase diagrams in Ref. [21].

With Eq. (A.28) and $\Sigma^m \approx \mathbf{0}$, the simplified polarization flux results in the active stress taking the following form:

$$\sigma^{\text{act}} = -p_{\text{act}} \mathbf{I} + \frac{\zeta \ell_o}{d-1} (U \mathbf{Q}'), \quad (\text{A.30a})$$

where the active pressure is defined as:

$$p_{\text{act}} = \frac{\zeta \ell_o U}{d(d-1)} \rho. \quad (\text{A.30b})$$

From Eq. (A.30) it is clear that the gradient contributions to the active stress will be generated by the convection of the traceless nematic order and equation of state for U (which will depend on the nature of the particle interactions) in addition to p_C .

We now turn to the traceless nematic field. Neglecting the time evolution of $\mathbf{Q}'$ in Eq. (A.17b) allows us to express $\mathbf{Q}' = \frac{\tau_R}{2d} (\nabla \cdot \mathbf{J}^{\mathbf{Q}'} - \eta^{\mathbf{Q}})$. A description of the nematic order requires expressions for $\eta^{\mathbf{Q}}$ (and thus, $\mathbf{C}$), $\mathbf{B}$, and $\Sigma^{\mathbf{Q}}$ [recall, we have already assumed the form of $\kappa^{\mathbf{Q}}$ in Eq. (A.29)]. We can now appreciate that each orientational field contains contributions related to the divergence of the next higher order field (e.g., $\mathbf{m} \sim \nabla \cdot \mathbf{Q}$) and therefore introduces higher order spatial gradients. We thus can safely assume that $\mathbf{B}$ is isotropic:

$$\mathbf{B} = \frac{1}{d+2} \alpha \cdot \mathbf{m}, \quad (\text{A.31})$$

where α is the fourth-order identity tensor given in Einstein notation as $\alpha_{ijkl} = \delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}$. Further, $\Sigma^{\mathbf{Q}}$ can be neglected as its contribution to the dynamic stress will be higher than second order in spatial gradient. We will also discard $\eta^{\mathbf{Q}}$ as its variance (at lowest order) will enter into the flux at the level of $\nabla \cdot \nabla \cdot \mathbf{QI}$. Finally, the fast relaxation of the polar order and nematic field implies that the density flux can be approximated to be vanishingly small such that $\zeta U_o \mathbf{m} \approx -\nabla \cdot \sigma^C$ [see Eq. (A.9b)]. Under these simplifications, the traceless nematic field takes the following form:

$$\mathbf{Q}' = \frac{\tau_R}{2\zeta U_o d(d+2)} \nabla \cdot \left[U \left(\nabla \cdot \sigma^C \right) \cdot \alpha \right]. \quad (\text{A.32})$$

With expressions for the bulk and gradient contributions for the conservative and active stress and the active noise statistics, we now have an expression for the density flux that *solely* depends on the density field itself. Evaluating these

expressions will require equations of state for the effective active speed, bulk interaction pressure, and any relevant interfacial coefficients in the conservative stress. We now summarize the fluctuating hydrodynamic equations for the coarse-grained density field, ρ . It is convenient to reorganize Eq. (A.32) under the simplification (consistent with our gradient theory) of $\sigma^C \approx -p_C \mathbf{I}$:

$$\mathbf{Q}' = \frac{3\ell_o}{2\zeta d(d+2)} \left[-\bar{U}(\rho) \frac{\partial p_C}{\partial \rho} \nabla^2 \rho \mathbf{I} - \frac{\partial}{\partial \rho} \left[\bar{U}(\rho) \frac{\partial p_C}{\partial \rho} \right] \nabla \rho \nabla \rho \right], \quad (\text{A.33})$$

where we have introduced the dimensionless effective active speed, $\bar{U} \equiv U/U_o$. Substitution of Eq. (A.33) into the active stress allows us to straightforwardly identify the active stress gradient terms.

The fluctuating hydrodynamics of athermal ABPs can now be summarized:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \mathbf{J}, \quad (\text{A.34a})$$

$$\mathbf{J} = \frac{1}{\zeta} \nabla \cdot \Sigma + \boldsymbol{\eta}^{\text{act}}, \quad (\text{A.34b})$$

$$\Sigma = \sigma^C + \sigma^{\text{act}}, \quad (\text{A.34c})$$

$$\sigma^C = \left[-p_C(\rho) + \kappa_1(\rho) \nabla^2 \rho + \kappa_2(\rho) |\nabla \rho|^2 \right] \mathbf{I} + \kappa_3(\rho) \nabla \rho \nabla \rho + \kappa_4(\rho) \nabla \nabla \rho, \quad (\text{A.34d})$$

$$\sigma^{\text{act}} = \left[-p_{\text{act}} + a(\rho) \nabla^2 \rho \right] \mathbf{I} + b(\rho) \nabla \rho \nabla \rho, \quad (\text{A.34e})$$

$$a(\rho) = \frac{3\ell_o^2}{2d(d-1)(d+2)} \bar{U}^2 \frac{dp_C}{d\rho}, \quad (\text{A.34f})$$

$$b(\rho) = \frac{3\ell_o^2 \bar{U}}{2d(d-1)(d+2)} \frac{d}{d\rho} \left[\bar{U} \frac{dp_C}{d\rho} \right], \quad (\text{A.34g})$$

where the statistics of the noise $\boldsymbol{\eta}^{\text{act}}$ are:

$$\langle \boldsymbol{\eta}^{\text{act}} \rangle = \mathbf{0}, \quad (\text{A.34h})$$

$$\langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \boldsymbol{\eta}^{\text{act}}(\mathbf{r}', t') \rangle = 2 \frac{k_B T^{\text{act}}}{\zeta} \left(\rho \mathbf{I} - \frac{d}{d-1} \mathbf{Q}' \right) \delta(t - t') \delta(\mathbf{r} - \mathbf{r}'), \quad (\text{A.34i})$$

and the simplified traceless nematic tensor is provided in Eq. (A.33) and its slightly more general form provided in Eq. (A.32). As stated above, our derived Langevin equations for observables such as Eq. (A.34) are to be interpreted in the Itô convention. However, as the noise appearing in Eq. (A.34) is *conserved*, the drift term associated with switching to the Stratonovich convention vanishes [58, 68–70].

The derived fluctuating hydrodynamics are general to athermal ABPs that obey the equations-of-motion described at the outset of this Appendix [Eqs. (A.1)]. As input, these fluctuating hydrodynamics require equations of state that will certainly depend on the precise form of the conservative particle interactions. Specifically, the density and activity dependence of $\bar{U}$, p_C , and the gradient coefficients $\{\kappa_i\}$ must be provided. For hard spheres, the active stress gradient coefficients scale as ℓ_o^2 [see Eqs. (A.34f) and (A.34g)] while those of the conservative interaction stress ($\{\kappa_i\}$) can only scale with powers of the hard sphere diameter d_{hs} . We therefore can safely discard $\{\kappa_i\}$ as we will exclusively focus on phase-separated hard spheres where the run length is at least an order of magnitude larger than the hard sphere diameter [$\ell_o/d_{\text{hs}} \geq \mathcal{O}(10)$].

A.2 Interfacial Height Fluctuations

We transform the density evolution to a height field evolution using the ansatz proposed by Bray *et al.*, $\rho(\mathbf{r}, t) = \varphi(z - h(\mathbf{x}, t))$ [49], where φ is the noise-averaged *stationary* density field. We assume that the conditions for a planar phase-separated state are met such that $\varphi(z)$ is simply a function of z , the normal direction to the interface. Furthermore, $\varphi(z)$ is spatially constant (at the binodal densities) at nearly all points except for within the interface, where the density transitions between the two binodal densities. The ansatz implies the following

chain-rule relations:

$$\frac{\partial \rho}{\partial t} = -\phi' \frac{\partial h}{\partial t} \quad (\text{A.35a})$$

$$|\nabla \rho|^2 = \left(1 + |\nabla_x h|^2\right) (\phi')^2 \quad (\text{A.35b})$$

$$\nabla^2 \rho = \left(1 + |\nabla_x h|^2\right) \phi'' - \nabla_x^2 h \phi', \quad (\text{A.35c})$$

where we denote the gradient in all directions other than z with ∇_x . Substitution of the continuity equation and our derived constitutive equation for the density flux [Eq.(A.34)] into Eq. (A.35a) results in:

$$-\phi' \frac{\partial h}{\partial t} = -\frac{1}{\zeta} \nabla \cdot \nabla \cdot \Sigma - \nabla \cdot \eta^{\text{act}}. \quad (\text{A.36})$$

We now express the double divergence of the dynamic stress using the above chain rule relations,

$$\begin{aligned} \nabla \cdot \nabla \cdot \Sigma = & \nabla^2 \left[-\mathcal{P}(\phi) + \left(1 + |\nabla_x h|^2\right) a(\phi) \phi'' - \nabla_x^2 h a(\phi) \phi' \right] \\ & + \nabla \cdot \nabla \cdot (b(\rho) \nabla \rho \nabla \rho), \end{aligned} \quad (\text{A.37})$$

where $\mathcal{P} \equiv p_C + p_{\text{act}}$ is the dynamic pressure. It is straightforward to show:

$$\nabla \cdot \nabla \cdot (b(\rho) \nabla \rho \nabla \rho) = -\frac{\partial}{\partial z} \left[b(\phi) (\phi')^2 \right] \nabla_x^2 h + \nabla^2 \left[b(\phi) (\phi')^2 \left(1 + |\nabla_x h|^2\right) \right]. \quad (\text{A.38})$$

Substituting Eq. (A.38) into Eq. (A.37) gives

$$\begin{aligned} \nabla \cdot \nabla \cdot \Sigma = & \nabla^2 \left[-\mathcal{P}(\phi) - \nabla_x^2 h a(\phi) \phi' + \left(1 + |\nabla_x h|^2\right) \left(a(\phi) \phi'' + b(\phi) (\phi')^2 \right) \right] \\ & - \frac{\partial}{\partial z} \left[b(\phi) (\phi')^2 \right] \nabla_x^2 h. \end{aligned} \quad (\text{A.39})$$

The height evolution is then given by

$$\begin{aligned} -\zeta \varphi' \frac{\partial h}{\partial t} = & -\nabla^2 \left[-\mathcal{P}(\varphi) - \nabla_x^2 h a(\varphi) \varphi' + \left(1 + |\nabla_x h|^2 \right) \left(a(\varphi) \varphi'' + b(\varphi) (\varphi')^2 \right) \right] \\ & + \frac{\partial}{\partial z} \left[b(\varphi) (\varphi')^2 \right] \nabla_x^2 h - \zeta \nabla \cdot \boldsymbol{\eta}^{\text{act}}. \end{aligned} \quad (\text{A.40})$$

Throughout the SM and the main text, we will denote the continuous Fourier transform of $h(\mathbf{x}, t)$ as $h(\mathbf{k}, t)$ (dimensions of $[\text{length}]^d$), defined as

$$h(\mathbf{k}, t) = \int d\mathbf{x} e^{i\mathbf{k} \cdot \mathbf{x}} h(\mathbf{x}, t), \quad (\text{A.41})$$

while we will denote the discrete (fast) Fourier transform as $\tilde{h}(\mathbf{k}, t)$ (dimensions of $[\text{length}]$). The continuous Fourier transform of Eq. (A.40) in the $\mathbf{x}$ direction (i.e. all directions except z) results in

$$\begin{aligned} -\zeta \varphi' \frac{\partial h}{\partial t} = & - \left[\frac{\partial^2}{\partial z^2} - k^2 \right] \left[-\mathcal{P}(\varphi) \delta(k) + k^2 h a(\varphi) \varphi' \right. \\ & + \mathcal{F} \left[1 + |\nabla_x h|^2 \right] \left(a(\varphi) \varphi'' + b(\varphi) (\varphi')^2 \right) \Big] \\ & - \frac{\partial}{\partial z} \left[b(\varphi) (\varphi')^2 \right] |k^2 h - \zeta \mathcal{F} \left[\nabla \cdot \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \right], \end{aligned} \quad (\text{A.42})$$

where we have defined the wavevector magnitude as $k = |\mathbf{k}|$ and the Fourier transform of $f(\mathbf{x})$ as $\mathcal{F} [f(\mathbf{x})]$. The statistics of the Fourier transformed noise, as well as all subsequent transformations to the noise, will be derived in later in this Appendix.

The Green's function of a Laplace operator that has been Fourier transformed in all directions but the z -direction is defined by [160]:

$$\underbrace{\left[\frac{\partial^2}{\partial z^2} - k^2 \right]}_{\text{Laplacian}} \underbrace{\left[\frac{1}{2k} e^{-k|z-z'|} \right]}_{\text{Green's Function}} = -\delta(z - z'). \quad (\text{A.43})$$

We thus multiply Eq. (A.42) by the Green's function and integrate across all z to find:

$$\begin{aligned} \zeta \frac{\partial h}{\partial t} \int dz' e^{-k|z-z'|} \varphi'(z') = & -2k^3 h a(\varphi) \varphi' \\ & - 2k \mathcal{F} [|\nabla_x h|^2] \left(a(\varphi) \varphi'' + b(\varphi) (\varphi')^2 \right) \\ & + k^2 h \int dz' e^{-k|z-z'|} \frac{\partial}{\partial z'} [b(\varphi) (\varphi')^2] | \\ & + \zeta \int dz' e^{-k|z-z'|} \mathcal{F} [\nabla \cdot \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t)], \end{aligned} \quad (\text{A.44})$$

where we have discarded terms proportional to $k\delta(k) = 0$. At this point, Eq. (A.44) retains a dependence on the z -dimension and contains nonlinear terms (i.e., $\mathcal{F} [|\nabla_x h|^2]$) that are outside of the scope of the ansatz. Fausti *et al.* [48] recognized that both the z -dependence and the nonlinear terms can be eliminated by multiplying Eq. (A.44) by the z -derivative of a density-dependent pseudovariabe $\mathcal{E}$ and integrating across the z -dimension. Under the ansatz, this pseudovariabe coincides with the same pseudovariabe used in equal-area constructions to determine nonequilibrium binodals [20, 21, 71]. Here $\mathcal{E}$, first introduced by Aifantis and Serrin [71], is defined by:

$$\frac{\partial^2 \mathcal{E}}{\partial \rho^2} = \frac{2b(\rho) - a'(\rho)}{a(\rho)} \frac{\partial \mathcal{E}}{\partial \rho}. \quad (\text{A.45})$$

We therefore multiply Eq. (A.44) by the z -derivative of the pseudovariabe, $\mathcal{E}$, that depends *solely* on density (and is nonzero only within the interface) and integrate over all $u = z - h$. We note that while integrating over u , the error incurred by approximating this integral as solely over z (i.e., neglecting h) has been shown to introduce error on the order of $k^2 h^3$ [49], which is higher order than the error introduced by using the ansatz. Such a treatment also implies that we may freely switch between φ and ρ in all integrals with respect to u . Use of the pseudovariabe results in:

$$\int du \frac{\partial \mathcal{E}}{\partial u} [a(\varphi) \varphi'' + b(\varphi) (\varphi')^2] = 0. \quad (\text{A.46})$$

Up to a constant of integration, $\partial\mathcal{E}/\partial z$ can be solved for using the definitions of $b(\varphi)$ and $a(\varphi)$ as

$$\frac{\partial\mathcal{E}}{\partial z} = C \frac{\partial\rho}{\partial z} \frac{\left(p'_c(\rho)\bar{U}(\rho)\right)^2}{a(\rho)} \sim \frac{\partial\rho}{\partial z} \frac{\partial p_C}{\partial\rho}, \quad (\text{A.47})$$

which is in agreement with the pseudovisible for active Brownian particles found in Ref. [21]. We note that in the equilibrium limit, $\ell_o/d_{\text{hs}} \rightarrow 0$, the coefficients associated with Eq. (A.45) are dictated by the Korteweg expansion [57] and the resulting solution becomes $\mathcal{E} \sim 1/\rho$.

Multiplication of Eq. (A.44) by $\partial\mathcal{E}/\partial u$ and integrating over all $u = z - h$ results in:

$$\begin{aligned} \zeta A(k) \frac{\partial h}{\partial t} = & -2k^3 h \int du a(\varphi) \varphi' \frac{\partial\mathcal{E}}{\partial u} \\ & + \zeta \int du \frac{\partial\mathcal{E}}{\partial u} \int dz' e^{-k|u-z'|} \mathcal{F} [\nabla \cdot \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t)] \\ & + k^2 h \int du \frac{\partial\mathcal{E}}{\partial u} \int dz' e^{-k|u-z'|} \frac{\partial}{\partial z'} [b(\varphi)(\varphi')^2], \end{aligned} \quad (\text{A.48})$$

where $A(k)$ is defined as

$$A(k) = \int \int du dz e^{-k|u-z|} \frac{\partial\mathcal{E}}{\partial u}(u) \varphi'(z). \quad (\text{A.49})$$

Consider the second integral on the RHS of Eq. (A.48). Integrating by parts gives

$$\int dz' e^{-k|u-z'|} \frac{\partial}{\partial z'} [b(\varphi)(\varphi')^2] = - \int dz' k \text{sgn}(u - z') e^{-k|u-z'|} b(\varphi)(\varphi')^2, \quad (\text{A.50})$$

where sgn denotes the sign function which returns 1 (−1) for any positive (negative) argument. Then the equation of motion for the height field simplifies to

$$\zeta_{\text{eff}} \frac{\partial h}{\partial t} = -k^3 \gamma_{\text{cw}} h + \chi(k, t) + O(k^3 h^2), \quad (\text{A.51})$$

where the error is due to both the ansatz and approximating integrals with respect to u as if they were with respect to z . The effective drag coefficient is defined as:

$$\zeta_{\text{eff}} = \frac{\zeta A^2(k)}{2B(k)\rho^{\text{surf}}}. \quad (\text{A.52})$$

γ_{cw} is the capillary-wave tension:

$$\begin{aligned} \gamma_{\text{cw}}(k) = & \frac{A(k)}{\rho^{\text{surf}} B(k)} \left[\int_{-\infty}^{\infty} du \frac{\partial \mathcal{E}}{\partial u} \varphi' a(\varphi) \right. \\ & \left. + \int_{-\infty}^{\infty} du \frac{\partial \mathcal{E}}{\partial u} \int_{-\infty}^{\infty} dz' \text{sgn}(u - z') e^{-k|u-z'|} b(\varphi) (\varphi')^2 \right], \end{aligned} \quad (\text{A.53})$$

where $\rho^{\text{surf}} \equiv (\rho^{\text{liq}} + \rho^{\text{gas}})/2$. The noise χ has the form:

$$\chi(k, t) = \frac{A(k)\zeta}{B(k)2\rho^{\text{surf}}} \int du \frac{\partial \mathcal{E}}{\partial u} \int dz' e^{-k|u-z'|} \mathcal{F} [\nabla \cdot \boldsymbol{\eta}^{\text{act}}], \quad (\text{A.54})$$

where the function $B(k)$ is defined as:

$$B(k) = \int \int du du' e^{-k|u-u'|} \frac{\partial \mathcal{E}}{\partial u}(u) \frac{\partial \mathcal{E}}{\partial u'}(u'). \quad (\text{A.55})$$

The emergence of ρ^{surf} and $B(k)$ occurs when solving for the statistics of χ , which we will now discuss. We first solve for the statistics of $\xi(\mathbf{k}, z, t) = \mathcal{F} [\nabla \cdot \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t)]$. The variance of ξ is given by

$$\langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle = \int \int d\mathbf{x} d\mathbf{x}' e^{i\mathbf{x} \cdot \mathbf{k}} e^{i\mathbf{x}' \cdot \mathbf{k}'} \nabla \cdot \nabla' \cdot \langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \boldsymbol{\eta}^{\text{act}}(\mathbf{r}', t') \rangle. \quad (\text{A.56})$$

Then by substituting Eq. (A.25b) into Eq. (A.56) we find:

$$\begin{aligned} \langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle = & -2 \frac{k_B T^{\text{act}}}{\zeta} \delta(t - t') \\ & \times \int \int d\mathbf{x} d\mathbf{x}' e^{i\mathbf{x} \cdot \mathbf{k}} e^{i\mathbf{x}' \cdot \mathbf{k}'} \nabla \cdot \left(\left(\rho \mathbf{I} - \frac{d}{d-1} \mathbf{Q}' \right) \cdot \nabla \delta(\mathbf{r} - \mathbf{r}') \right). \end{aligned} \quad (\text{A.57})$$

Using Eq. (A.33), and the definitions of $a(\rho)$ and $b(\rho)$,

$$\left(\rho \mathbf{I} - \frac{d}{d-1} \mathbf{Q}' \right) = \rho \mathbf{I} + \frac{d}{\ell_o^2 D_R \zeta \bar{U}(\rho)} \left[a(\rho) \nabla^2 \rho \mathbf{I} + b(\rho) \nabla \rho \nabla \rho \right]. \quad (\text{A.58})$$

Then the statistics of ξ can be split into three contributions

$$\langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle = \sum_{i=1}^3 \langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle_i, \quad (\text{A.59})$$

with the three contributions defined as:

$$\begin{aligned} \langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle_1 &= -2 \frac{k_B T^{\text{act}}}{\zeta} \delta(t - t') \int \int d\mathbf{x} d\mathbf{x}' \\ &\quad \times e^{i\mathbf{x} \cdot \mathbf{k}} e^{i\mathbf{x}' \cdot \mathbf{k}'} \nabla \cdot (\rho \nabla \delta(\mathbf{r} - \mathbf{r}')), \end{aligned} \quad (\text{A.60a})$$

$$\begin{aligned} \langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle_2 &= -\frac{2k_B T^{\text{act}} d}{\ell_o^2 \zeta^2 D_R} \delta(t - t') \int \int d\mathbf{x} d\mathbf{x}' \\ &\quad \times e^{i\mathbf{x} \cdot \mathbf{k}} e^{i\mathbf{x}' \cdot \mathbf{k}'} \nabla \cdot \left(\frac{1}{\bar{U}(\rho)} a(\rho) \nabla^2 \rho \nabla \delta(\mathbf{r} - \mathbf{r}') \right), \end{aligned} \quad (\text{A.60b})$$

$$\begin{aligned} \langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle_3 &= -\frac{2k_B T^{\text{act}} d}{\ell_o^2 \zeta^2 D_R} \delta(t - t') \int \int d\mathbf{x} d\mathbf{x}' \\ &\quad \times e^{i\mathbf{x} \cdot \mathbf{k}} e^{i\mathbf{x}' \cdot \mathbf{k}'} \nabla \cdot \left(\frac{1}{\bar{U}(\rho)} b(\rho) \nabla \rho \nabla \rho \cdot \nabla \delta(\mathbf{r} - \mathbf{r}') \right). \end{aligned} \quad (\text{A.60c})$$

We evaluate these terms separately to lowest order in k , beginning with the first. Consider the term:

$$\nabla \cdot (\rho \nabla \delta(\mathbf{r} - \mathbf{r}')) = \frac{\partial}{\partial z} \left(\rho \frac{\partial}{\partial z} \delta(\mathbf{r} - \mathbf{r}') \right) + \nabla_x \cdot (\rho \nabla_x \delta(\mathbf{r} - \mathbf{r}')). \quad (\text{A.61})$$

Upon substitution of Eq. (A.61) into Eq. (A.60a), and ignoring the dependence of φ on h , we integrate by parts to find that

$$\begin{aligned} \langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle_1 &= 2 \frac{k_B T^{\text{act}}}{\zeta} (2\pi)^{d-1} \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') \\ &\times \left(k^2 \varphi \delta(z - z') - \frac{\partial}{\partial z} \left(\varphi \frac{\partial}{\partial z} \delta(z - z') \right) \right). \end{aligned} \quad (\text{A.62})$$

The same lines of argument may be used for Eq. (A.60b) to show:

$$\begin{aligned} \langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle_2 &= \frac{2k_B T^{\text{act}} d}{\ell_o^2 \zeta^2 D_R} (2\pi)^{d-1} \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') \\ &\times \left(\frac{k^2 a(\varphi)}{\bar{U}(\varphi)} \varphi'' \delta(z - z') - \frac{\partial}{\partial z} \left(\frac{a(\varphi) \varphi''}{\bar{U}(\varphi)} \frac{\partial}{\partial z} \delta(z - z') \right) \right). \end{aligned} \quad (\text{A.63})$$

The lowest order in k contribution from Eq. (A.60c) will be the term where all derivatives are in the z -direction, giving

$$\begin{aligned} \langle \xi(\mathbf{k}, z, t) \xi(\mathbf{k}', z', t') \rangle_3 &= - \frac{2k_B T^{\text{act}} d}{\ell_o^2 \zeta^2 D_R} (2\pi)^{d-1} \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') \\ &\times \frac{\partial}{\partial z} \left(\frac{b(\rho)}{\bar{U}(\rho)} (\varphi')^2 \frac{\partial}{\partial z} \delta(z - z') \right). \end{aligned} \quad (\text{A.64})$$

Now we introduce $L(\mathbf{k}, t)$:

$$L(\mathbf{k}, t) = \int du \frac{\partial \mathcal{E}}{\partial u} \int dz' e^{-k|u-z'|} \xi(\mathbf{k}, z', t). \quad (\text{A.65})$$

The variance of L is given by:

$$\begin{aligned} \langle L(\mathbf{k}, z, t) L(\mathbf{k}', z', t') \rangle &= \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int \int dz' dz'' \\ &\times e^{-k|u-z'|} e^{-k|u'-z''|} \langle \xi(\mathbf{k}, z', t) \xi(\mathbf{k}', z'', t') \rangle. \end{aligned} \quad (\text{A.66})$$

Then, as with ξ , we split up the variance of L into three contributions,

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1 &= \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int \int dz' dz'' \\ &\times e^{-k|u-z'|} e^{-k|u'-z''|} \langle \xi(\mathbf{k}, z', t) \xi(\mathbf{k}', z'', t') \rangle_1, \end{aligned} \quad (\text{A.67a})$$

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_2 &= \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int \int dz' dz'' \\ &\times e^{-k|u-z'|} e^{-k|u'-z''|} \langle \xi(\mathbf{k}, z', t) \xi(\mathbf{k}', z'', t') \rangle_2, \end{aligned} \quad (\text{A.67b})$$

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_3 &= \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int \int dz' dz'' \\ &\times e^{-k|u-z'|} e^{-k|u'-z''|} \langle \xi(\mathbf{k}, z', t) \xi(\mathbf{k}', z'', t') \rangle_3. \end{aligned} \quad (\text{A.67c})$$

We begin with the first contribution, Eq. (A.67a):

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1 &= 2 \frac{k_B T^{\text{act}}}{\zeta} (2\pi)^{d-1} \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \\ &\times \int \int dz' dz'' e^{-k|u-z'|} e^{-k|u'-z''|} \left(k^2 \varphi \delta(z' - z'') - \frac{\partial}{\partial z'} \left(\varphi \frac{\partial}{\partial z'} \delta(z' - z'') \right) \right). \end{aligned} \quad (\text{A.68})$$

The first contribution may be separated into two subcontributions,

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1^a &= 2 \frac{k_B T^{\text{act}}}{\zeta} (2\pi)^{d-1} \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int \int dz' dz'' \\ &\times e^{-k|u-z'|} e^{-k|u'-z''|} \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') \left(k^2 \varphi \delta(z' - z'') \right), \end{aligned} \quad (\text{A.69a})$$

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1^b &= -2 \frac{k_B T^{\text{act}}}{\zeta} (2\pi)^{d-1} \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int \int dz' dz'' \\ &\times e^{-k|u-z'|} e^{-k|u'-z''|} \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') \frac{\partial}{\partial z'} \left(\varphi \frac{\partial}{\partial z'} \delta(z' - z'') \right). \end{aligned} \quad (\text{A.69b})$$

In Eq. (A.69a), the delta function immediately eliminates one of the integrals, leaving us with

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1^a &= \frac{2D_R(d-1)}{d} (2\pi)^{d-1} \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int dz' \\ &\times e^{-k|u-z'|} e^{-k|u'-z'|} \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') k^2 \varphi. \end{aligned} \quad (\text{A.70})$$

Because $\partial \mathcal{E} / \partial z$ is zero everywhere except within the interface, we will approximate φ as its value halfway between the binodal densities, i.e., $\varphi \approx (\rho^{\text{liq}} + \rho^{\text{gas}}) / 2 = \rho^{\text{surf}}$. In addition, by splitting up the integral over z' into its contributions when z' is less than, greater than, or between u, u' one can show that

$$\int dz' e^{-k|u-z'|} e^{-k|u'-z'|} = \frac{1}{k} e^{-k|u-u'|}. \quad (\text{A.71})$$

As a result, Eq. (A.69a) simplifies to:

$$\langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1^a = \frac{2k_B T^{\text{act}} \rho^{\text{surf}}}{\zeta} (2\pi)^{d-1} k \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') B(k).$$

Next we simplify Eq. (A.69b). Integrating by part results in:

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1^b &= 2 \frac{k_B T^{\text{act}}}{\zeta} (2\pi)^{d-1} \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int \int dz' dz'' \\ &\times e^{-k|u-z'|} e^{-k|u'-z''|} \text{sgn}(u - z') k \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') \varphi \frac{\partial}{\partial z'} \delta(z' - z''), \end{aligned} \quad (\text{A.72})$$

followed by another integration by parts:

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1^b &= 2 \frac{k_B T^{\text{act}}}{\zeta} (2\pi)^{d-1} \int du \frac{\partial \mathcal{E}}{\partial u} \int du' \frac{\partial \mathcal{E}}{\partial u'} \int dz' \\ &\times e^{-k|u-z'|} e^{-k|u'-z'|} \text{sgn}(u - z') \text{sgn}(u' - z') k^2 \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') \varphi. \end{aligned} \quad (\text{A.73})$$

Because u' and u will only be evaluated where their magnitudes are very small, and z' will go from negative infinity to infinity, we will approximate $\text{sgn}(u -$

$z')\text{sgn}(u' - z')$ as one. Then, via the same lines of argument, b simplifies to the same contribution as a ,

$$\langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1^b = \frac{2k_B T^{\text{act}} \rho^{\text{surf}}}{\zeta} (2\pi)^{d-1} k \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') B(k). \quad (\text{A.74})$$

Which results in

$$\langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_1 = \frac{4k_B T^{\text{act}} \rho^{\text{surf}}}{\zeta} (2\pi)^{d-1} k \delta(\mathbf{k} + \mathbf{k}') \delta(t - t') B(k). \quad (\text{A.75})$$

For Eq. (A.67b), it is convenient to define the quantity $C(k)$,

$$C(k) = \int \int du du' \frac{\partial \mathcal{E}}{\partial u}(u) \frac{\partial \mathcal{E}}{\partial u'}(u') \int \int dz' dz'' e^{-k|u-z'|} e^{-k|u'-z''|} \\ \times \left(k^2 \frac{a(\rho)}{\bar{U}} \varphi''(z) \delta(z - z') - \frac{\partial}{\partial z} \left(\frac{a(\rho)}{\bar{U}} \varphi''(z) \frac{\partial}{\partial z} \delta(z - z') \right) \right). \quad (\text{A.76})$$

Then Eq. (A.67b) can be rewritten as

$$\langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_2 = \frac{2k_B T^{\text{act}} d}{\ell_o^2 \zeta^2 D_R} \delta(t - t') \delta(\mathbf{k} + \mathbf{k}') (2\pi)^{(d-1)} C(k). \quad (\text{A.77})$$

Similarly, Eq. (A.67c) can be rewritten by defining the quantity $D(k)$,

$$D(k) = \int \int du du' \frac{\partial \mathcal{E}}{\partial u}(u) \frac{\partial \mathcal{E}}{\partial u'}(u') \int \int dz' dz'' \\ \times e^{-k|u-z'|} e^{-k|u'-z''|} \frac{\partial}{\partial z'} \left[\frac{b(\rho)}{\bar{U}} (\varphi'(z'))^2 \frac{\partial}{\partial z'} \delta(z - z') \right], \quad (\text{A.78})$$

which results in

$$\langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle_3 = \frac{2k_B T^{\text{act}} d}{\ell_o^2 \zeta^2 D_R} \delta(t - t') \delta(\mathbf{k} + \mathbf{k}') (2\pi)^{(d-1)} D(k). \quad (\text{A.79})$$

Altogether, we find the full noise correlator as

$$\begin{aligned} \langle L(\mathbf{k}, t) L(\mathbf{k}', t') \rangle = & 2 \frac{k_B T^{\text{act}}}{\zeta} \delta(t - t') \delta(\mathbf{k} + \mathbf{k}') (2\pi)^{(d-1)} \\ & \times \left[2\rho^{\text{surf}} k B(k) + \frac{d}{\ell_o^2 \zeta D_R} C(k) + \frac{d}{\ell_o^2 \zeta D_R} D(k) \right], \end{aligned} \quad (\text{A.80})$$

where $B(k)$, $C(k)$, and $D(k)$ are all to be evaluated numerically. Finally, we define $\chi(\mathbf{k}, t)$ with the coefficients as defined in Eq. (A.54). We then break up the noise into two independent contributions, $\chi = \chi^{\text{iso}} + \chi^{\text{aniso}}$ which have zero mean and variances:

$$\langle \chi^{\text{iso}}(\mathbf{k}, t) \chi^{\text{iso}}(\mathbf{k}', t') \rangle = 2k(k_B T)^{\text{act}} \zeta_{\text{eff}} \delta(t - t') \delta(\mathbf{k} + \mathbf{k}') (2\pi)^{(d-1)}, \quad (\text{A.81a})$$

$$\langle \chi^{\text{aniso}}(\mathbf{k}, t) \chi^{\text{aniso}}(\mathbf{k}', t') \rangle = \frac{\zeta_{\text{eff}}(C(k) + D(k))}{(d-1)\rho^{\text{surf}} B(k)} \delta(t - t') \delta(\mathbf{k} + \mathbf{k}') (2\pi)^{(d-1)}. \quad (\text{A.81b})$$

A.3 Height Correlations, Power Spectrum, and Area Minimization

Now with the complete height-field Langevin equation, we proceed to evaluating the capillary fluctuations of the interface, i.e., evaluate $\langle h(\mathbf{k}) h(-\mathbf{k}) \rangle$. If we multiply Eq. (A.51) by $h(-\mathbf{k}, t)$, average over the noise, and consider only the steady state, we find

$$k^3 \gamma_{\text{cw}} \langle h(\mathbf{k}) h(-\mathbf{k}) \rangle = \langle \chi(\mathbf{k}, t) h(-\mathbf{k}, t) \rangle. \quad (\text{A.82})$$

Determining the stationary fluctuations thus require evaluating the noise-averaged correlation of the height field with the noise. The implicit solution for h is given by,

$$h(-\mathbf{k}, t) = h(-\mathbf{k}, 0) \exp\left(-\frac{t}{\tau(k)}\right) + \frac{1}{\zeta_{\text{eff}}} \int_0^t dt' \exp\left(-\frac{t-t'}{\tau(k)}\right) \chi(-\mathbf{k}, t'), \quad (\text{A.83})$$

where the timescale for the relaxation of a capillary wave τ is given by

$$\tau(k) \equiv \frac{\zeta_{\text{eff}}}{k^3 \gamma_{\text{cw}}}. \quad (\text{A.84})$$

We now multiply Eq. (A.83) by $\chi(\mathbf{k}, t)$ and take an expectation:

$$\langle \chi(\mathbf{k}, t) h(-\mathbf{k}) \rangle = \frac{1}{\zeta_{\text{eff}}} \int_0^t dt' \exp\left(-\frac{t-t'}{\tau(k)}\right) \langle \chi(-\mathbf{k}, t') \chi(\mathbf{k}, t) \rangle. \quad (\text{A.85})$$

We substitute in the statistics of χ , Eq. (A.81a), and Eq. (A.81b) to find

$$\begin{aligned} \langle \chi(\mathbf{k}, t) h(-\mathbf{k}) \rangle &= \frac{1}{\zeta_{\text{eff}}} \int_0^t dt' \exp\left(-\frac{t-t'}{\tau(k)}\right) \delta(t-t') L^{d-1} \\ &\quad \times \left[2\zeta_{\text{eff}} k (k_B T^{\text{act}}) + \frac{\zeta_{\text{eff}} (C(k) + D(k))}{(d-1)\rho^{\text{surf}} B(k)} \right], \end{aligned} \quad (\text{A.86})$$

where we have noted that $(2\pi)^{d-1} \delta(\mathbf{k} = \mathbf{0}) = L^{d-1}$ for a system of finite size (in three dimensions this is the area projected by the interface onto the (x, y) plane). The stationary fluctuations can now be determined:

$$\langle h(\mathbf{k}) h(-\mathbf{k}) \rangle = \frac{L^{(d-1)}}{\gamma_{\text{cw}}} \left[\frac{k_B T^{\text{act}}}{k^2} + \frac{C(k) + D(k)}{k^3 (d-1) 2\rho^{\text{surf}} B(k)} \right]. \quad (\text{A.87})$$

As shown in Fig. 2 of the main text, the anisotropic contributions to the fluctuations can be safely ignored at low k and low run length. Under these conditions we are left with

$$\langle h(\mathbf{k}) h(-\mathbf{k}) \rangle = \frac{L^{(d-1)} k_B T^{\text{act}}}{\gamma_{\text{cw}} k^2}. \quad (\text{A.88})$$

We can also Fourier transform Eq. (A.51) in time to find:

$$i\omega \zeta_{\text{eff}} h(\mathbf{k}, \omega) = -k^3 \gamma_{\text{cw}} h(\mathbf{k}, \omega) + \chi(\mathbf{k}, \omega). \quad (\text{A.89})$$

After rearranging Eq. (A.89) and multiplying both sides of the equation by its own complex conjugate we find:

$$h(\mathbf{k}, \omega)h(-\mathbf{k}, -\omega) = \frac{1}{\zeta_{\text{eff}}^2 \omega^2 + k^6 \gamma_{\text{cw}}^2} \chi(\mathbf{k}, \omega) \chi(-\mathbf{k}, \omega). \quad (\text{A.90})$$

We can then average over the noise to extract the power spectra:

$$\langle |h(\mathbf{k}, \omega)|^2 \rangle = \frac{L^{d-1} \Gamma \zeta_{\text{eff}}}{\zeta_{\text{eff}}^2 \omega^2 + k^6 \gamma_{\text{cw}}^2} \left[2k(k_B T)^{\text{act}} + \frac{C(k) + D(k)}{(d-1)\rho^{\text{surf}} B(k)} \right], \quad (\text{A.91})$$

where we have used $2\pi\delta(\omega = 0) = \Gamma$, the total time of a finite duration trajectory. At low k and low run length, we ignore the anisotropic contributions to the power spectra and find:

$$\langle |h(k, \omega)|^2 \rangle = \frac{2kL^{d-1}\Gamma k_B T^{\text{act}}}{\zeta_{\text{eff}}(\omega^2 + \tau(k)^{-2})}. \quad (\text{A.92})$$

In order to demonstrate that Eq. (A.51) implies area minimization, we wish to find a condition such that interfacial dynamics (at steady state) recover a Boltzmann distribution *weighted by the interfacial area*. In other words,

$$P[h] \sim \exp \left[-C \int d\mathbf{x} \sqrt{1 + |\nabla_x h|^2} \right], \quad (\text{A.93})$$

where C is a physical constant. For a weakly fluctuating interface where $|\nabla_x h|^2 \ll 1$, this area may be Taylor expanded as:

$$P[h] \sim \exp \left[-C \int d\mathbf{x} \left(1 + \frac{1}{2} |\nabla_x h|^2 \right) \right]. \quad (\text{A.94})$$

The value of $\int d\mathbf{x} [1]$ is simply the area of a flat interface, which is an inconsequential constant:

$$P[h] \sim \exp \left[-\frac{C}{2} \int d\mathbf{x} |\nabla_x h|^2 \right]. \quad (\text{A.95})$$

We now express the discrete Fourier series expansion of $h(\mathbf{x})$ as:

$$h(\mathbf{x}) = \sum_{\mathbf{k}} h_{\mathbf{k}} \exp [-i\mathbf{k} \cdot \mathbf{x}]. \quad (\text{A.96})$$

Then the gradient of $h(\mathbf{x})$ is:

$$\nabla_x h(\mathbf{x}) = -i \sum_{\mathbf{k}} h_{\mathbf{k}} \mathbf{k} \exp [-i\mathbf{k} \cdot \mathbf{x}]. \quad (\text{A.97})$$

We can then solve for square gradient of $h(\mathbf{x})$ as:

$$|\nabla_x h(\mathbf{x})|^2 = - \sum_{\mathbf{k}} \sum_{\mathbf{k}'} h_{\mathbf{k}} h_{\mathbf{k}'} \mathbf{k} \cdot \mathbf{k}' \exp [-i(\mathbf{k} + \mathbf{k}') \cdot \mathbf{x}]. \quad (\text{A.98})$$

Then our distribution is proportional to:

$$P[h] \sim \exp \left[\frac{C}{2} \int d\mathbf{x} \sum_{\mathbf{k}} \sum_{\mathbf{k}'} h_{\mathbf{k}} h_{\mathbf{k}'} \mathbf{k} \cdot \mathbf{k}' \exp [-i(\mathbf{k} + \mathbf{k}') \cdot \mathbf{x}] \right]. \quad (\text{A.99})$$

Evaluating the integral allows us to only consider the cases where $\mathbf{k} = -\mathbf{k}'$, and the symmetry of $h_{\mathbf{k}} = h_{-\mathbf{k}}$ for the even components and $h_{\mathbf{k}} = -h_{-\mathbf{k}}$ for the odd components allows us to identify:

$$P[h_{\mathbf{k}}] \sim \exp \left[-\frac{C}{2} |h_{\mathbf{k}}|^2 |\mathbf{k}|^2 \right]. \quad (\text{A.100})$$

We now wish to determine whether the linearized interfacial dynamics derived earlier in this Appendix exhibits this distribution at steady-state. We rewrite Eq. (A.51):

$$\zeta_{\text{eff}}(k) \frac{\partial h}{\partial t} = -k^3 \gamma_{\text{cw}}(k) h + \chi(k, t). \quad (\text{A.101})$$

For small k , the tension and effective drag become constants:

$$\frac{\partial h}{\partial t} = -k^3 \frac{\gamma_{\text{cw}}}{\zeta_{\text{eff}}} h + \chi'(k, t), \quad (\text{A.102})$$

where the noise χ' has zero average and variance:

$$\langle \chi'(k, t) \chi'(k', t') \rangle = \left(2k \frac{(k_B T^{\text{act}})}{\zeta_{\text{eff}}} + \mathcal{A}(k) \right) \delta(t - t') \delta(k + k') (2\pi)^{d-1}. \quad (\text{A.103})$$

Here $\mathcal{A}(k)$ is the component noise variance originating from the anisotropic noise,

$$\mathcal{A}(k) = \frac{\zeta_{\text{eff}}(C(k) + D(k))}{(d-1)\rho^{\text{surf}}B(k)}. \quad (\text{A.104})$$

The Fokker-Planck equation describing the distribution of $P[h]$ is then straightforwardly found:

$$\frac{\partial}{\partial t} P[h] = -\frac{\partial}{\partial h} \left[-k^3 \frac{\gamma_{\text{cw}}}{\zeta_{\text{eff}}} h P[h] \right] + \frac{1}{2} L^{d-1} \left(2k \frac{(k_B T^{\text{act}})}{\zeta_{\text{eff}}} + \mathcal{A}(k) \right) \frac{\partial^2}{\partial h^2} [P[h]]. \quad (\text{A.105})$$

Solving for the steady state of Eq. (A.105) results in:

$$0 = \frac{\partial}{\partial h} \left[k^3 \frac{\gamma_{\text{cw}}}{\zeta_{\text{eff}}} h P[h] + \frac{1}{2} L^{d-1} \left(2k \frac{(k_B T^{\text{act}})}{\zeta_{\text{eff}}} + \mathcal{A}(k) \right) \frac{\partial}{\partial h} [P[h]] \right]. \quad (\text{A.106})$$

After rearranging Eq. (A.106) and integrating we find:

$$P[h] \sim \exp \left[-2L^{1-d} k^2 \gamma_{\text{cw}} h^2 \left(2(k_B T^{\text{act}}) + \frac{1}{k} \mathcal{A}(k) \right)^{-1} \right]. \quad (\text{A.107})$$

At small wavevectors, $\mathcal{A}(k)$ is negligible and we find:

$$P[h] \sim \exp \left[-2k^2 h^2 \frac{L^{1-d} \gamma_{\text{cw}}}{k_B T^{\text{act}}} \right], \quad (\text{A.108})$$

which is indeed a Boltzmann distribution weighted by the interfacial area and the interfacial stiffness $\gamma_{\text{cw}}/k_B T^{\text{act}}$. However, if $\mathcal{A}(k)$ is not negligible, then this distribution will only be recovered if $\mathcal{A}(k)$ *scales linearly with k* . Based on inspection of Eqs. (A.55), (A.76), and (A.78), $\mathcal{A}(k)$ scales as k^2 in the low- k limit. This implies that when the anisotropic contributions to the noise are non-negligible, a Boltzmann distribution with respect to an effective free energy proportional to the interfacial area is not recovered.

A.4 Brownian Dynamics Simulations

We take the interparticle force $\mathbf{F}_{ij}$ to result from a Weeks-Chandler-Anderson (WCA) potential [124], characterized by an energy scale ε and Lennard-Jones diameter d_{LJ} . Despite our use of a continuous potential, the finite and constant amplitude of the active force along with the use of a stiff WCA allow us to achieve hard-sphere statistics. A stiffness of $\mathcal{S} \equiv \varepsilon/\zeta U_o d_{\text{LJ}} = 50$ was found to be sufficient to achieve effective hard-sphere interactions with a hard-sphere diameter of $d_{\text{hs}} = 2^{1/6} d_{\text{LJ}}$. In this hard-sphere limit, the system state is independent of the active force magnitude and is fully described by two parameters: the density and the intrinsic run length ℓ_0 . We will focus on the case of three-dimensional ABPs for the reasons outlined in the main text.

Before choosing the specific details of our simulations, we consider the computational scaling. As demonstrated above, the *number of timesteps* required to observe the relaxation of a capillary wave is derived to scale as k^{-3} . In addition, obtaining lower k requires expansion of a simulation cell which, at constant density, requires additional particles. An isotropic expansion of the simulation cell will thus result in the scaling $1/k \sim N^d$. The computational cost of a single timestep scales linearly with N . Therefore, in order to relax capillary waves of increasingly small k , the computational cost of every time step scales as $(1/k)^d$ while the number of timesteps to obtain statistically significant data scales as $(1/k)^3$. As a result, for a square interface in 3d the total computational cost of relaxing a capillary-wave scales as $(1/k)^6$, placing intense resource limitations on sampling a low k limit. In addition, because of the vanishing capillary-wave tension at low λ found above, we expect simulations at lower activities to have significantly longer relaxation times and therefore higher statistical uncertainty for the same number of simulation time steps as higher activities.

Brownian dynamics simulations of the above system were then performed using 631444 particles with intrinsic run lengths of $\ell_0/d_{\text{hs}} = \{40.09, 63.70, 89.09, 148.3\}$ for a length of 89000 d_{hs}/U_o using HOOMD-blue [65]. Rectangular simulations with dimensions of $L_z/d_{\text{hs}} = 221.1$, $L_x/d_{\text{hs}} = 196.7$, $L_y/d_{\text{hs}} = 19.2$ were employed. This combination of cell dimensions and number of particles corresponds

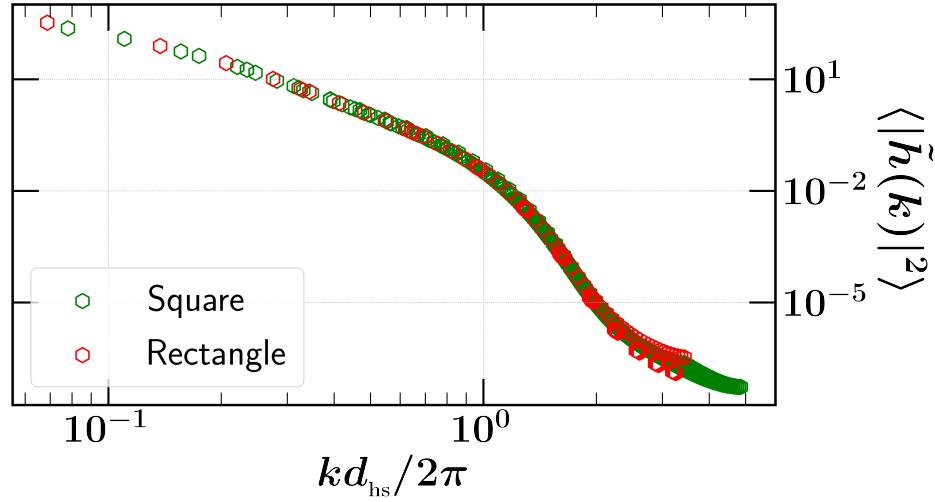


Figure A.1: Interfacial fluctuation spectrum for square ($80.5 \times 80.5 d_{\text{hs}}^2$) and rectangular ($91.4 \times 19.7 d_{\text{hs}}^2$) interfaces. Simulations were conducted at an activity of $\ell_o/d_{\text{hs}} = 89$.

to an overall volume fraction of $\phi = 0.397$.

Simulation cell dimensions were unequal in the directions tangential to the interface in order to maximize the length of one dimension (and therefore access lower wave vector fluctuations) without incurring additional computational expense. For our 3d system, this allows the cost of each time step to scale as $(1/k)^2$ rather than $(1/k)^3$. *A priori*, it was unclear whether introducing unequal dimensions would introduce artefacts into the measured height fluctuation spectra. We therefore measured the height fluctuations of (smaller) systems with square and rectangular interfaces at an activity of $\ell_o/d_{\text{hs}} = 89.09$. The dimensions of the square and rectangular interfaces were $L_x/d_{\text{hs}} = L_y/d_{\text{hs}} = 80.5$ and $L_x/d_{\text{hs}} = 91.4$, $L_y/d_{\text{hs}} = 19.7$, respectively. Their height fluctuation spectra are plotted in Fig. A.1. The fluctuation spectrum from the systems with equal and unequal dimensions are indistinguishable until wave vector far outside the scope of a low- k theory, justifying the unequal simulation cell dimensions used in our production runs.

The instantaneous location of interfaces was identified using the algorithm proposed by Willard and Chandler [66]. The coarse grained density field was

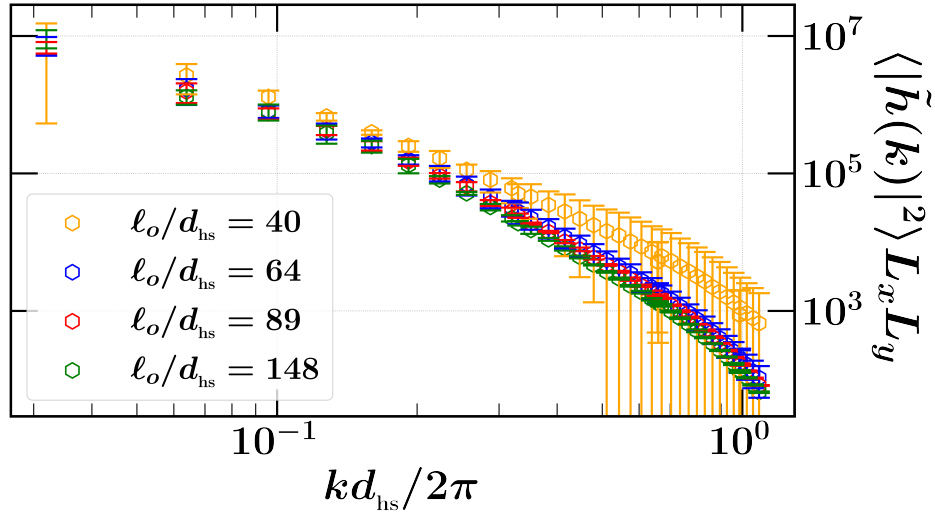


Figure A.2: Interfacial fluctuation spectrum for production runs with error bars included.

defined by specifying the kernel $\Delta(\mathbf{r} - \mathbf{r}_i)$ to be a Gaussian,

$$\rho(\mathbf{r}, t) = \left(2\pi\xi^2\right)^{-d/2} \sum_{i=1}^N \exp\left[-\frac{|\mathbf{r} - \mathbf{r}_i(t)|^2}{2\xi^2}\right], \quad (\text{A.109})$$

where ξ is a coarse-graining length. The value of this field was calculated every $4.45 d_{\text{hs}}/U_0$ with $\xi/d_{\text{hs}} = 1.78$ on a cubic grid of points with spacing $0.89 d_{\text{hs}}$. The $(d-1)$ dimensional surface at which the density field was equal ρ^{surf} was then determined through linear interpolation.

The statistical uncertainty of the height fluctuation spectra (as well as the exponent ω , stiffness K_s , and later power spectra fits) was determined by calculating dividing the trajectory into five equally spaced periods in space, calculating the average fluctuation spectrum of each period independently, and taking the standard deviation between the periods. The spread on the data corresponding to $\ell_o/d_{\text{hs}} = 40.09$ was significantly higher than the other data points. This is because the relaxation time of capillary excitations at this run length was up to an order of magnitude higher than the other activities simulated for any given wave vector, as shown by Eq. (A.84). We also attempt to collapse the height fluctuation spectra by multiplying each curve by ℓ_o . This collapse was first observed by Patch *et al.* [25]

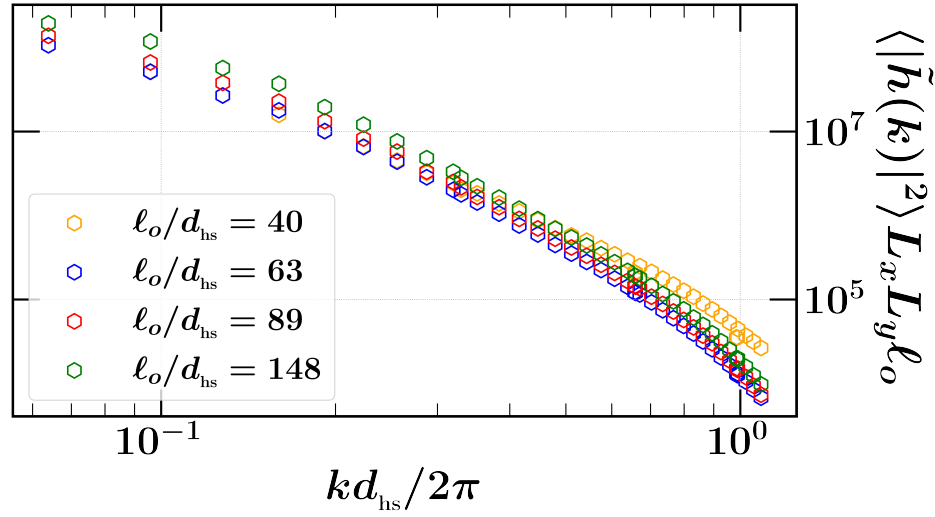


Figure A.3: Interfacial fluctuation spectrum multiplied by ℓ_o . At k low enough to be described by Eq. (A.88), all plots should collapse to a single point.

for 2d ABPs and a theoretical justification for this collapse is discussed in the main text. This curve collapse should, in principle, apply to 3d ABPs at sufficiently low k but it is unclear whether our sampling reached low enough values of k for Eq. (A.88) to hold. In addition, the statistical uncertainty found in Fig. A.2 may be concealing any potential curve collapse.

From a fit of the height spectrum data to $\langle |\tilde{h}(k)|^2 \rangle = K_s k^\omega$ we obtain the interfacial stiffness K_s^{-1} . We find that the interfacial stiffness measured from simulation is within an order of magnitude agreement to the predictions of the theory, as plotted by Fig. A.4.

The power spectra data with $\omega d_{hs} / 2\pi U_o = 2.82 \times 10^{-4}$ fixed is plotted in Fig. A.5. The lowest twelve points of the power spectra was fit to $\langle |h(k, \omega)|^2 \rangle \sim k^\beta$ and values of β are included in the inset of Fig. A.5. This plot is interested in probing the $\omega \rightarrow 0$ limit as closely as possible, so the lowest two values of k were not included in the plot or fit as $\tau(k)$ as predicted by Eq. (A.84) was on the order of ω^{-1}

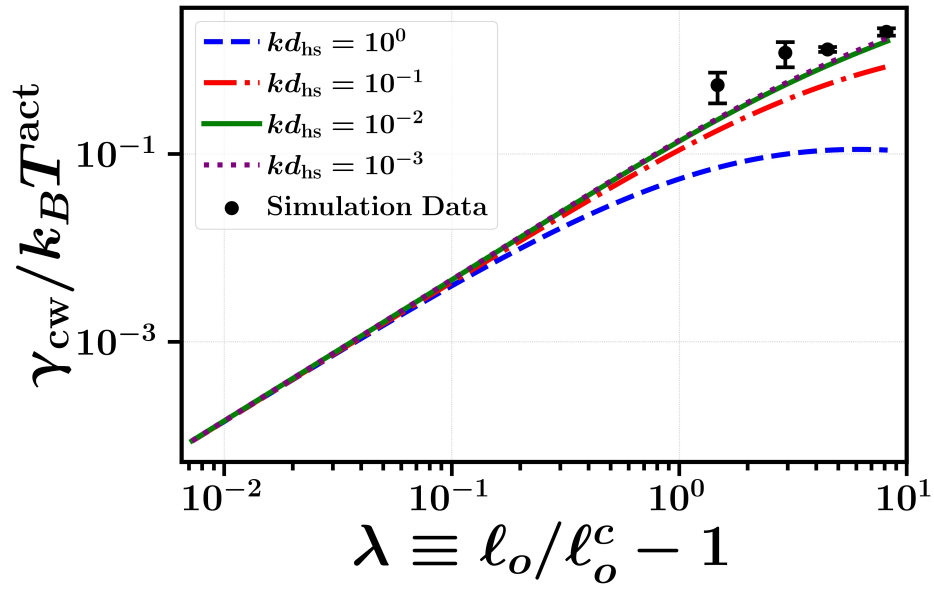


Figure A.4: Interfacial stiffness $\gamma_{cw}/k_B T^{\text{act}}$ as predicted by theory (lines) and as measured via simulation data (points). The stiffness is presented in units of d_{hs}^{-2} . The simulation data points were fit from the low- k ($kd_{\text{hs}}/2\pi < 0.5$) data.

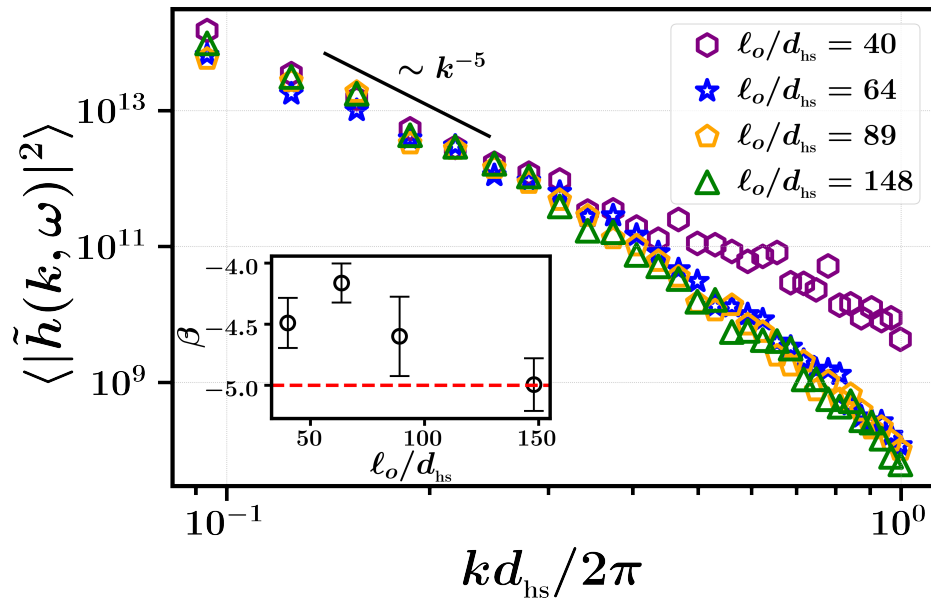


Figure A.5: Power spectra $\langle |h(k, \omega)|^2 \rangle$ at each simulated activity as a function of k with $\omega d_{\text{hs}}/2\pi U_o = 2.82 \times 10^{-4}$ fixed. (Inset) Power law scaling obtained from logarithmic fits to $\langle |h(k, \omega)|^2 \rangle \sim k^\beta$, considering low k such that $k d_{\text{hs}}/2\pi < 0.5$. Lowest two values of k not included in plot or fit.

Appendix B

Appendix: Chiral Active Interfaces

The contents of Appendix B will closely follow that of the Supplemental Information of Ref. [85]. We will present the details of the mechanical derivation of chiral active phase coexistence as well as capillarity and odd-surface flows at chiral active interfaces.

B.1 Coexistence of Chiral Active Particles

Here we summarize the theory used to solve for the binodals and stationary density profile of macroscopically coexisting phases of chiral active Brownian particles (cABPs). The theory applies the framework of Ref. [21] which was also applied to achiral ABPs in that work. We consider N particles with positional degrees of freedom $\mathbf{r}_i$ and orientational degrees of freedom $\mathbf{q}_i$. Then the phase space vector $\mathbf{\Gamma} \equiv [\mathbf{r}^N, \mathbf{q}^N]$ is a $2dN$ dimensional vector in d spatial dimensions. The evolution of the position and orientation of the i th particle is given by

$$\dot{\mathbf{r}}_i = U_o \mathbf{q}_i + \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij}, \quad (\text{B.1a})$$

$$\dot{\mathbf{q}}_i = \boldsymbol{\omega}_o \times \mathbf{q}_i + (\boldsymbol{\Omega}_i \mathbf{e}_z) \times \mathbf{q}_i, \quad (\text{B.1b})$$

where U_o is the active speed, ζ is the drag and $\boldsymbol{\omega}_o$ is the constant angular velocity applied to the particles. This constant torque gives the system an overall chirality.

The noise Ω_i has zero average and variance $2D_R\delta(t-t')\delta_{ij}$. We interpret Eq. (A.1b) in the Stratonovich convention, which is necessary to preserve the modulus of $\mathbf{q}_i$. In the limit of vanishing noise variances, an isolated particle here will trace out a circle with radius inversely proportional to $|\omega_o|$. By dividing the active speed by D_R , one can define the run length $\ell_o \equiv U_o/D_R$ as the typical distance a particle travels before reorienting in the limit $\omega_o \rightarrow 0$. Note that Eqs. (B.1) are equivalent to standard ABPs [See Eqs. (A.1)] in the limit that $\omega \rightarrow 0$. The Fokker-Planck equation associated with Eqs. (B.1) is given by:

$$\frac{\partial}{\partial t}f(\mathbf{\Gamma};t) = \mathcal{L}f(\mathbf{\Gamma};t), \quad (\text{B.2})$$

where $f(\mathbf{\Gamma};t)$ is the microstate probability density and $\mathcal{L}$ is the dynamical (Fokker-Planck) operator consistent with Eq. (B.1) with:

$$\mathcal{L} \equiv \sum_{i=1}^N \left[\frac{\partial}{\partial \mathbf{r}_i} \cdot \left(-U_o \mathbf{q}_i - \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij} \right) + \nabla_i^R \cdot (D_R \nabla_i^R - \omega_o) \right], \quad (\text{B.3})$$

where we have defined the rotational gradient operator as $\nabla_i^R \equiv \mathbf{q}_i \times \partial/\partial \mathbf{q}_i$. The adjoint of the dynamical operator can be similarly solved for as:

$$\mathcal{L}^* \equiv \sum_{i=1}^N \left[\left(U_o \mathbf{q}_i + \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij} \right) \cdot \frac{\partial}{\partial \mathbf{r}_i} + (D_R \nabla_i^R + \omega_o) \cdot \nabla_i^R \right]. \quad (\text{B.4})$$

The average of an arbitrary macroscopic observable O is:

$$\langle O \rangle = \int d\mathbf{\Gamma} O f(\mathbf{\Gamma};t), \quad (\text{B.5})$$

while the average evolution O can be expressed as:

$$\frac{\partial \langle O \rangle}{\partial t} = \int d\mathbf{\Gamma} O \mathcal{L}f(\mathbf{\Gamma};t), \quad (\text{B.6})$$

or equivalently as:

$$\frac{\partial \langle O \rangle}{\partial t} = \int d\Gamma f(\Gamma; t) \mathcal{L}^* O. \quad (\text{B.7})$$

We now define the density ρ as $\rho(\mathbf{r}) \equiv \langle \sum_{i=1}^N \delta(\mathbf{r}_i - \mathbf{r}) \rangle$. Then, proceeding in the same manner as Ref. [21], the evolution of the density is found to be:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \left(U_o \mathbf{m} + \frac{1}{\zeta} \nabla \cdot \boldsymbol{\sigma}^C \right), \quad (\text{B.8})$$

where $\nabla \equiv \partial/\partial \mathbf{r}$ and we have defined the polar order density $\mathbf{m} \equiv \langle \sum_{i=1}^N \mathbf{q}_i \delta(\mathbf{r}_i - \mathbf{r}) \rangle$ and interaction stress $\boldsymbol{\sigma}^C$ (see Refs. [21] or Appendix A.1 for microscopic expression). The evolution of the density is dependent on the polar order, and an expression for the evolution of the polar order can be found in a similar fashion:

$$\frac{\partial \mathbf{m}}{\partial t} = -\nabla \cdot \mathbf{J}^m + (1 - d) D_R \mathbf{m} + \boldsymbol{\omega}_o \times \mathbf{m}, \quad (\text{B.9a})$$

where we have defined a flux of polar order $\mathbf{J}^m$ as:

$$\mathbf{J}^m = U_o \left(\mathbf{Q} + \frac{\rho}{d} \mathbf{I} \right) + \frac{1}{\zeta} \boldsymbol{\kappa}^m + \frac{1}{\zeta} \nabla \cdot \boldsymbol{\Sigma}^m. \quad (\text{B.9b})$$

Here $\boldsymbol{\kappa}^m$, $\boldsymbol{\Sigma}^m$, are the “stress” and “body-force”-like contributions to the flux of polar order, with microscopic expressions given by:

$$\boldsymbol{\kappa}^m = \left\langle \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i}^N \mathbf{F}_{ij} (\mathbf{q}_i - \mathbf{q}_j) \delta(\mathbf{r} - \mathbf{r}_i) \right\rangle, \quad (\text{B.10})$$

$$\boldsymbol{\Sigma}^m = - \left\langle \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i}^N (\mathbf{r}_i - \mathbf{r}_j) \mathbf{F}_{ij} \mathbf{q}_i \int_0^1 d\lambda \delta(\mathbf{r} - \mathbf{r}_j - \lambda (\mathbf{r}_i - \mathbf{r}_j)) \right\rangle. \quad (\text{B.11})$$

In the expression for the polar flux we have also introduced the traceless nematic order tensor as $\mathbf{Q} \equiv \left\langle \sum_{i=1}^N \left(\mathbf{q}_i \mathbf{q}_i - \frac{1}{d} \mathbf{I} \right) \delta(\mathbf{r}_i - \mathbf{r}) \right\rangle$. The evolution of the polar order

is dependent on the traceless nematic order, and an expression for the evolution of the traceless nematic order can be found in a similar fashion:

$$\frac{\partial \mathbf{Q}}{\partial t} = -\nabla \cdot \mathbf{J}^Q + \omega_o \times \left(\mathbf{Q} + \frac{\rho}{d} \mathbf{I} \right) + \left[\omega_o \times \left(\mathbf{Q} + \frac{\rho}{d} \mathbf{I} \right) \right]^T - 2dD_R \mathbf{Q}, \quad (\text{B.12a})$$

where we have defined a flux of nematic order $\mathbf{J}^Q$ as:

$$\mathbf{J}^Q = U_o \tilde{\mathbf{B}} + \frac{1}{\zeta} \boldsymbol{\kappa}^Q + \frac{1}{\zeta} \nabla \cdot \boldsymbol{\Sigma}^Q - \frac{U_o}{d} \mathbf{m} \mathbf{I} - \frac{1}{d\zeta} \left(\nabla \cdot \boldsymbol{\sigma}^C \right) \mathbf{I}. \quad (\text{B.12b})$$

Here $\boldsymbol{\kappa}^Q$, $\boldsymbol{\Sigma}^Q$, are the “stress” and “body force”-like contributions to the flux of nematic order, with microscopic expressions given by:

$$\boldsymbol{\kappa}^Q = \left\langle \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i}^N \mathbf{F}_{ij} (\mathbf{q}_i \mathbf{q}_i - \mathbf{q}_j \mathbf{q}_j) \delta(\mathbf{r} - \mathbf{r}_i) \right\rangle, \quad (\text{B.13})$$

$$\boldsymbol{\Sigma}^Q = - \left\langle \frac{1}{2} \sum_{i=1}^N \sum_{j \neq i}^N (\mathbf{r}_i - \mathbf{r}_j) \mathbf{F}_{ij} \mathbf{q}_i \mathbf{q}_i \int_0^1 d\lambda \delta(\mathbf{r} - \mathbf{r}_j - \lambda(\mathbf{r}_i - \mathbf{r}_j)) \right\rangle. \quad (\text{B.14})$$

In the expression for the flux of nematic order we have also defined ($\tilde{\mathbf{B}} \equiv \langle \sum_{i=1}^N \mathbf{q}_i \mathbf{q}_i \mathbf{q}_i \delta(\mathbf{r}_i - \mathbf{r}) \rangle$). Although the cross product of a second rank tensor is not defined, we have employed the following notation to describe the object which (using indicial notation) is expressed as:

$$\omega_o \times \left(\mathbf{Q} + \frac{\rho}{d} \mathbf{I} \right) + \left[\omega_o \times \left(\mathbf{Q} + \frac{\rho}{d} \mathbf{I} \right) \right]^T = \left\langle \omega_o^\gamma \epsilon^{\gamma\omega\mu} \sum_{i=1}^N \left(\delta^{\mu\alpha} q_i^\beta q_i^\omega + \delta^{\mu\beta} q_i^\alpha q_i^\omega \right) \right\rangle. \quad (\text{B.15})$$

In the above indicial notation expression we use Latin subscripts to denote particle label and Greek superscripts to denote spatial component. We find that the evolution of the traceless nematic order is dependent on $\tilde{\mathbf{B}}$. The coexistence theory proposed in Ref. [21] requires a second order expansion of the dynamic stress tensor, or equivalently a third order expansion of the density flux. Because the

dynamics of the (n) th orientational moment is dependent on the divergence of the $(n + 1)$ th orientational moment, each additional level included in the hierarchy of equation serves to increase the spatial order. Truncation of the hierarchy of equations at the nematic order is then sufficient for understanding coexistence. The closure of the above hierarchy of equations can be greatly simplified by focusing on a system confined to two dimensions ($d = 2$). In this case, the only relevant component of ω_o is that in the out-of-plane direction, which we pick to be parallel to the unit vector $\mathbf{e}_z$. All orientations are constrained to the (x, y) plane. We can then write $\omega_o = \omega_o \mathbf{e}_z$. Then two relevant inverse time scales become apparent from Eq. (B.1): D_R and ω_o . We then define $\chi \equiv \omega_o/D_R$ as a dimensionless parameter capturing the relative strength of ω_o and D_R , which serves as a measure of the extent to which spatial parity between x and y has been broken. In two dimensions, the term $\omega_o \times \mathbf{m}$ can be rewritten as:

$$\omega_o \times \mathbf{m} = \omega_o m_x \mathbf{e}_y - \omega_o m_y \mathbf{e}_x, \quad (\text{B.16})$$

and Eq. (B.15) can be rewritten as:

$$\omega_o \times \left(\mathbf{Q} + \frac{\rho}{d} \mathbf{I} \right) + \left[\omega_o \times \left(\mathbf{Q} + \frac{\rho}{d} \mathbf{I} \right) \right]^T = \omega_o \begin{bmatrix} 2Q_{xy} & Q_{xx} - Q_{yy} \\ Q_{xx} - Q_{yy} & 2Q_{xy} \end{bmatrix}. \quad (\text{B.17})$$

We now consider the steady-state. At steady-state, the time derivative of ρ , $\mathbf{m}$, and $\mathbf{Q}$ must all be zero. Then the density-flux [the term in the parentheses of Eq. (B.8)] must have zero divergence. Therefore the density-flux must be given by the curl of some vector field $\mathbf{A}$. We can then express the polar order as:

$$\mathbf{m} = \frac{1}{U_o} \left(\nabla \times \mathbf{A} - \frac{1}{\zeta} \nabla \cdot \sigma^C \right). \quad (\text{B.18})$$

We now close the hierarchy of equations by assuming the $\tilde{\mathbf{B}}$ field to be isotropic:

$$\tilde{\mathbf{B}} = \frac{1}{4} \alpha \cdot \mathbf{m}, \quad (\text{B.19})$$

where α is the fourth order identity tensor represented in indicial notation by $\alpha^{\mu\gamma\nu\omega} = \delta^{\mu\gamma} \delta^{\nu\omega} + \delta^{\mu\nu} \delta^{\gamma\omega} + \delta^{\mu\omega} \delta^{\gamma\nu}$. Following Ref. [21], we also discard Σ^m , and

Σ^Q , an approximation that was found to be accurate in achiral ABPs, and introduce the following constitutive relations for κ^m , κ^Q :

$$\kappa^m = -\zeta U_o \left(1 - \bar{U}(\rho)\right) \left(\mathbf{Q} + \frac{\rho}{d}\mathbf{I}\right), \quad (\text{B.20a})$$

$$\kappa^Q = -\zeta U_o \left(1 - \bar{U}(\rho)\right) \tilde{\mathbf{B}}, \quad (\text{B.20b})$$

where $\bar{U} \in [0, 1]$ is the dimensionless active speed and is an equation of state provided later in this section. The motivation behind this constitutive law is noting that the microscopic definitions of κ^m and κ^Q are maximal when particles are antiparallel and vanishes when particles are parallel, and thus has similar microscopic origins to the reduction in effective swim speed [21]. Finally, as is standard in the active particle literature [19, 21, 64], we ignore gradient contributions to the interaction stress and approximate $\sigma^C \approx -p_C(\rho)\mathbf{I}$, where $p_C(\rho)$ is an equation of state for the interaction pressure. By substituting Eq. (B.18), Eqs. (B.20), and Eq. (B.19) into Eqs. (B.12); setting the time derivative of $\mathbf{Q}$ to zero; and assuming that at steady states gradients in the density are orthogonal to the residual flux $\nabla \times \mathbf{A}$, one can solve for the components of the traceless nematic order tensor as:

$$Q_{xx} = -\frac{3}{16\zeta D_R} \left(\frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial x} \right)^2 + \frac{\chi}{8} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left[\left(\frac{\partial \rho}{\partial x} \right)^2 - \left(\frac{\partial \rho}{\partial y} \right)^2 \right] \right. \\ \left. + \frac{\chi}{2} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial x} \frac{\partial \rho}{\partial y} \right) + \bar{U} \frac{\partial p_C}{\partial \rho} \nabla^2 \rho \right), \quad (\text{B.21a})$$

$$Q_{yy} = -\frac{3}{16\zeta D_R} \left(\frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial y} \right)^2 + \frac{\chi^2}{8} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left[\left(\frac{\partial \rho}{\partial x} \right)^2 - \left(\frac{\partial \rho}{\partial y} \right)^2 \right] \right. \\ \left. + \frac{\chi}{2} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial x} \frac{\partial \rho}{\partial y} \right) + \bar{U} \frac{\partial p_C}{\partial \rho} \nabla^2 \rho \right), \quad (\text{B.21b})$$

$$Q_{xy} = -\frac{3}{16\zeta D_R} \left(\frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial x} \frac{\partial \rho}{\partial y} \right) + \frac{\chi}{4} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\left(\frac{\partial \rho}{\partial x} \right)^2 - \left(\frac{\partial \rho}{\partial y} \right)^2 \right) \right). \quad (\text{B.21c})$$

We can now consider the evolution of the polar order [Eq. (B.9)] at steady-state:

$$\mathbf{m} = -\nabla \cdot \left(\ell_o \bar{U} \mathbf{Q} + \frac{\ell_o \bar{U} \rho}{2} \mathbf{I} \right) + \frac{1}{D_R} \omega_o \times \mathbf{m}. \quad (\text{B.22})$$

Breaking up Eq. (B.22) into its x and y components results in two equations to solve for two unknowns m_x and m_y . Solving for m_x and m_y then recombining into vector notation results in:

$$\mathbf{m} (1 + \chi^2) = -\nabla \cdot \left(\ell_o \bar{U} \mathbf{Q} + \frac{\ell_o \bar{U} \rho}{2} \mathbf{I} \right) - \frac{1}{D_R} \omega_o \times \left(\nabla \cdot \left(\ell_o \bar{U} \mathbf{Q} + \frac{\ell_o \bar{U} \rho}{2} \mathbf{I} \right) \right). \quad (\text{B.23})$$

The density flux is dependent on the divergence of the interaction stress and the polar order. We have approximated the interaction stress as proportional to the interaction pressure, which is given by an equation of state solely dependent on density. Additionally, through Eqs. (B.21), we have closed the traceless nematic order fully in terms of the density and equations of state. We now have an expressions for the polar order dependent only on density, the traceless nematic order, and equations of state. Thus we have found an expression for the density flux that is fully closed in terms of the density field:

$$\mathbf{J} = \frac{1}{\zeta} \nabla \cdot [\mathbf{T} + \chi \mathbf{T}^{\text{cross}} + \mathbf{T}^{\text{odd}}] \quad (\text{B.24a})$$

$$\mathbf{T} = \begin{bmatrix} -\mathcal{P} - \frac{\zeta U_o \ell_o \bar{U} Q_{xx}}{1+\chi^2} & -\frac{\zeta U_o \ell_o \bar{U} Q_{xy}}{1+\chi^2} \\ -\frac{\zeta U_o \ell_o \bar{U} Q_{xy}}{1+\chi^2} & -\mathcal{P} - \frac{\zeta U_o \ell_o \bar{U} Q_{yy}}{1+\chi^2} \end{bmatrix} \quad (\text{B.24b})$$

$$\mathbf{T}^{\text{cross}} = \begin{bmatrix} \frac{\chi \zeta U_o \ell_o \bar{U} Q_{xy}}{1+\chi^2} & -\frac{\chi \zeta U_o \ell_o \bar{U} Q_{xx}}{1+\chi^2} \\ -\frac{\chi \zeta U_o \ell_o \bar{U} Q_{xx}}{1+\chi^2} & -\frac{\chi \zeta U_o \ell_o \bar{U} Q_{xy}}{1+\chi^2} \end{bmatrix} \quad (\text{B.24c})$$

$$\mathbf{T}^{\text{odd}} = \begin{bmatrix} 0 & -\frac{\chi \zeta U_o \ell_o \bar{U} \rho}{2(1+\chi^2)} \\ \frac{\chi \zeta U_o \ell_o \bar{U} \rho}{2(1+\chi^2)} & 0 \end{bmatrix} \quad (\text{B.24d})$$

$$\mathcal{P} = p_C(\rho) + \frac{\zeta U_o \ell_o \bar{U} \rho}{2(1 + \chi^2)} \quad (\text{B.24e})$$

We now seek the criteria at which the system macroscopically phase separates with a planar interface. In this limit we may restrict density gradients to be in a single direction. Picking this direction to be $\mathbf{e}_x$, we demand that the flux in the x direction is zero, i.e. $\mathbf{J} \cdot \mathbf{e}_x = 0$. This constraint results in:

$$0 = -\frac{\partial \mathcal{P}}{\partial \rho} \frac{\partial \rho}{\partial x} - \frac{\ell_o \zeta U_o}{(1 + \chi^2)} \frac{\partial}{\partial x} [\bar{U} Q_{xx}] + \frac{\chi U_o \zeta \ell_o}{1 + \chi^2} \frac{\partial}{\partial x} [\bar{U} Q_{xy}]. \quad (\text{B.25})$$

Integration of Eq. (B.25) from $x = -\infty$ to $x = +\infty$, and noting that the bulk phases have no nematic order, results in:

$$\mathcal{P}(\rho^{\text{gas}}) = \mathcal{P}(\rho^{\text{liq}}) = \mathcal{P}^{\text{coexist}}. \quad (\text{B.26})$$

Indefinite integration of Eq. (B.25) results in:

$$\mathcal{P}(\rho) - \mathcal{P}^{\text{coexist}} = -\frac{\ell_o \zeta U_o \bar{U} Q_{xx}}{1 + \chi^2} + \frac{\chi \ell_o \zeta U_o \bar{U} Q_{xy}}{1 + \chi^2}, \quad (\text{B.27})$$

where we have identified the constant of integration as the coexistence pressure $\mathcal{P}^{\text{coexist}}$. Substitution of Eqs. (B.21) into Eq. (B.27) results in:

$$\begin{aligned} \mathcal{P}(\rho) - \mathcal{P}^{\text{coexist}} &= \frac{3\ell_o^2}{16(1 + \chi^2)} \left(\bar{U} \right)^2 \frac{\partial p_C}{\partial \rho} \frac{\partial^2 \rho}{\partial x^2} \\ &+ \left(1 - \frac{\chi^2}{8} \right) \frac{3\ell_o^2}{16(1 + \chi^2)} \bar{U} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial x} \right)^2. \end{aligned} \quad (\text{B.28})$$

We now define the following constants:

$$\mathbf{a}(\rho) = \frac{3\ell_o^2}{16(1 + \chi^2)} \left(\bar{U} \right)^2 \frac{\partial p_C}{\partial \rho}, \quad (\text{B.29a})$$

$$\mathbf{b}(\rho) = \left(1 - \frac{\chi^2}{8} \right) \frac{3\ell_o^2}{16(1 + \chi^2)} \bar{U} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right], \quad (\text{B.29b})$$

where substitution of these constants into Eq. (B.28) results in the compact notation:

$$\mathcal{P}(\rho) - \mathcal{P}^{\text{coexist}} = \mathfrak{a}(\rho) \frac{\partial^2 \rho}{\partial x^2} + \mathfrak{b}(\rho) \left(\frac{\partial \rho}{\partial x} \right)^2. \quad (\text{B.30})$$

Eq. (B.30) is in a form amenable to the macroscopic coexistence theory of Ref. [21]. We may define the pseudovariabe:

$$\frac{\partial^2 \mathcal{E}}{\partial \rho^2} = \frac{2\mathfrak{b}(\rho) - \frac{\partial \mathfrak{a}}{\partial \rho}}{\mathfrak{a}(\rho)} \frac{\partial \mathcal{E}}{\partial \rho}. \quad (\text{B.31})$$

Substitution of Eqs. (B.29) into Eq. (B.31) and solution of the differential equation results in:

$$\frac{\partial \mathcal{E}}{\partial \rho} \sim \left(\overline{U} \right)^{-\frac{\chi^2}{4}} \left(\frac{\partial p_C}{\partial \rho} \right)^{1-\frac{\chi^2}{4}}. \quad (\text{B.32})$$

In the limit $\chi \rightarrow 0$, the pseudovariabe recovers the same solutions as found for achiral ABPs [21, 42, 64]. Integration of Eq. (B.30) with respect to the pseudovariabe from one binodal to the other results in:

$$\int_{\rho^{\text{gas}}}^{\rho^{\text{liq}}} [\mathcal{P}(\rho) - \mathcal{P}^{\text{coexist}}] \frac{\partial \mathcal{E}}{\partial \rho} d\rho. \quad (\text{B.33})$$

Together, Eqs. (B.26) and (B.33) are the coexistence criteria for chiral ABPs. They represent two equations for which two unknowns, the coexisting densities in each phase, can be solved for. Further, as we discussed in Ref. [42] and extended to finite-size phases in Ref. [108], the coexistence criteria can be used to solve for the density profile connecting the two phases. While macroscopic coexistence demands that $\mathbf{J} \cdot \mathbf{e}_x = 0$, the tangential flux $\mathbf{J} \cdot \mathbf{e}_y$ may be non-zero. For a system with no gradients in the y direction, the tangential flux is found to be:

$$\mathbf{J} \cdot \mathbf{e}_y = -\frac{U_o \ell_o}{1 + \chi^2} \frac{\partial}{\partial x} [Q_{xy} \overline{U} + \chi Q_{xx} \overline{U}] + \frac{\chi U_o \ell_o}{2(1 + \chi^2)} \frac{\partial}{\partial x} [\overline{U} \rho]. \quad (\text{B.34})$$

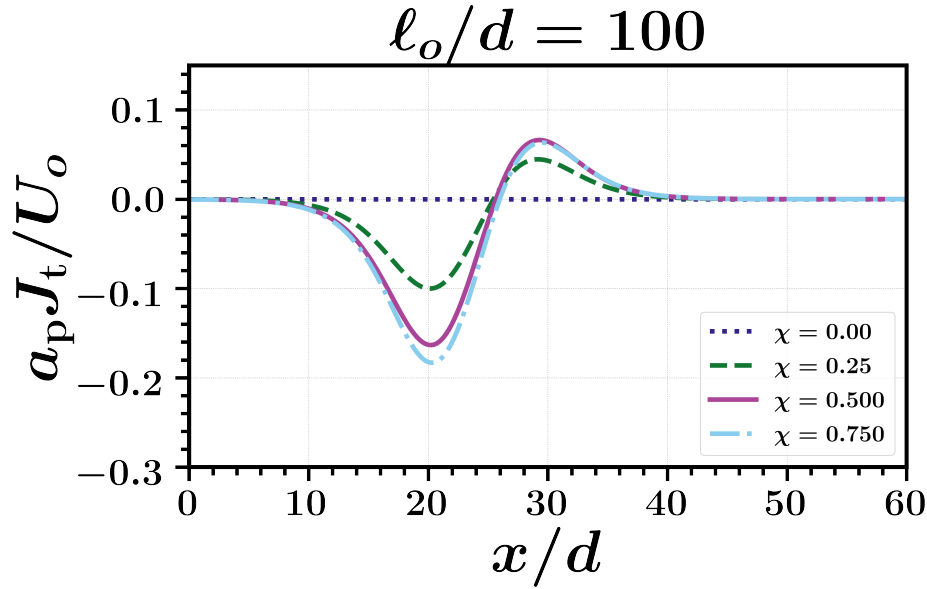


Figure B.1: Theoretically predicted flux tangential to the interface for $\ell_o/d = 75$ and selected values of χ . Here $a_p = \pi d^2/4$ is the area of a particle and $J_t = \mathbf{J} \cdot \mathbf{e}_y$ is the tangential flux.

Noting that $Q_{xy} = 0$ when $\chi = 0$, the tangential flux vanishes in the limit $\chi \rightarrow 0$. Therefore tangential fluxes at steady state in ABPs can only be supported upon violation of spatial parity symmetry. Substitution of the closure relations for $\mathbf{Q}$ [Eq. (B.21)] and the density profiles which may be solved for using the coexistence criteria results will yield steady-state tangential flux profiles, which we report in Figs. B.1- B.3

Evaluation of the steady-state binodals, density profiles, and tangential fluxes requires equations of state for the interaction pressure $p_C(\rho)$ and the dimensionless active speed $\bar{U}$. For simplicity, we choose equations of state that capture the expected qualitative behavior while satisfying known physical limits. For example, in the limit of zero area fraction we demand that the interaction pressure is zero and the dimensionless active speed is one. In the limit of close-packing ($\phi \rightarrow \phi_o$) we demand that the interaction pressure diverges and the effective dimensionless active

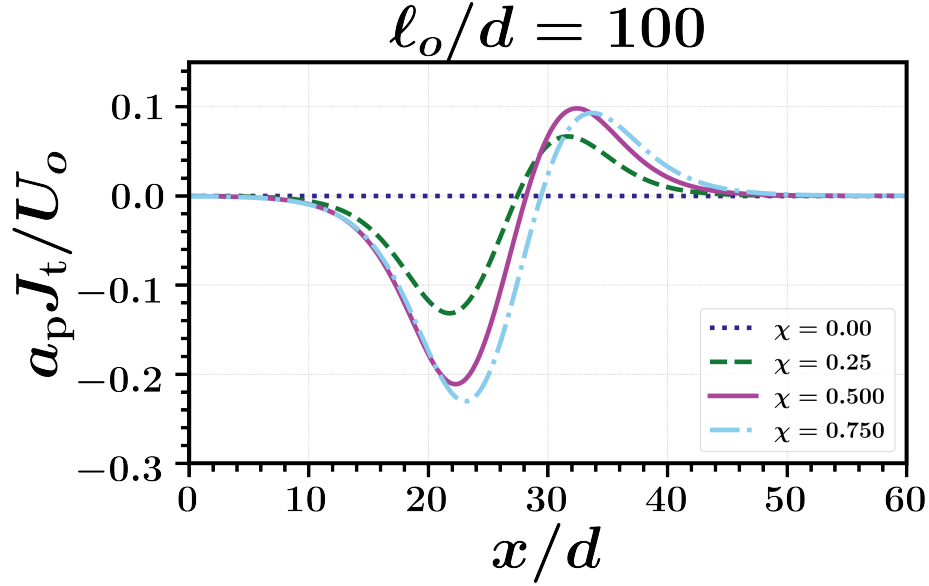


Figure B.2: Theoretically predicted flux tangential to the interface for $\ell_o/d = 100$ and selected values of χ . Here $a_p = \pi d^2/4$ is the area of a particle and $J_t = \mathbf{J} \cdot \mathbf{e}_y$ is the tangential flux.

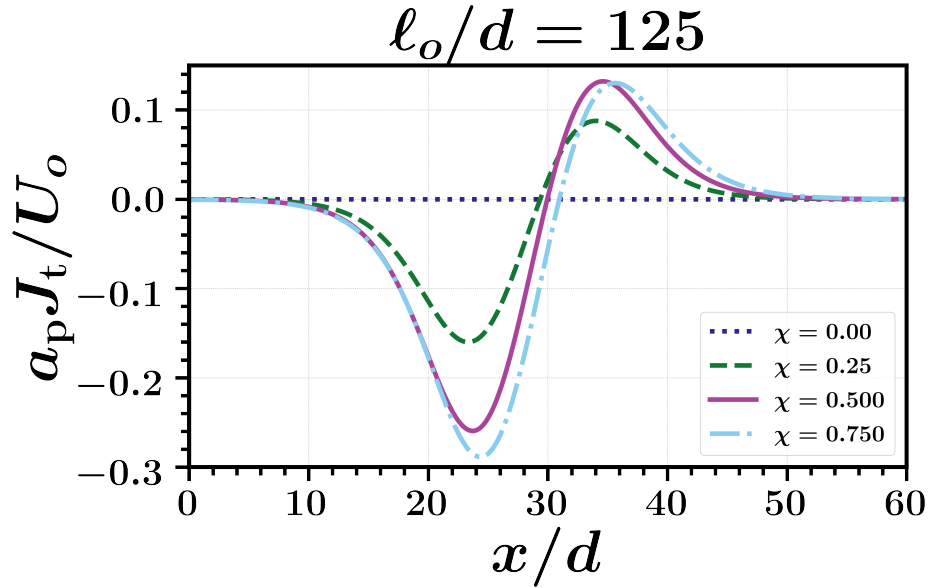


Figure B.3: Theoretically predicted flux tangential to the interface for $\ell_o/d = 125$ and selected values of χ . Here $a_p = \pi d^2/4$ is the area of a particle and $J_t = \mathbf{J} \cdot \mathbf{e}_y$ is the tangential flux.

speed is zero. We therefore adopt the interaction pressure proposed in Ref. [18]:

$$\frac{p_C d}{\zeta U_o} = \frac{8}{\pi} \phi^2 \left(1 - \frac{\phi}{\phi_o} \right)^{-1}, \quad (\text{B.35})$$

where $\phi \equiv \pi \rho d^2 / 4$ is the area fraction and a value of 0.9 is used for the close-packed area fraction ϕ_o . As discussed in Ref. [18], this form of interaction pressure is proportional to the area fraction times the value of the radial distribution function at contact. Choosing a different interaction potential would then modify the equation of state to reflect the change in radial distribution function. Further, we note that the function used to approximate the radial distribution function at contact was reported for monodisperse hard disks and may not be accurate for the polydisperse hard disks simulated in this study. We use the following form for the effective active speed:

$$\overline{U} = \left[1 + \phi \left(1 - \frac{\phi}{\phi_o} \right)^{-1} \right]^{-1}. \quad (\text{B.36})$$

The ability of our coexistence theory to quantitatively predict the results of particle simulations is highly dependent on the accuracy of our equation of states in modeling $p_C(\rho)$ and $\overline{U}(\rho)$. We do not expect quantitative accuracy out of the above equations of state, as they are minimal functions which satisfy relevant physical limits and have not been rigorously fit to simulation data. For completeness, we report the values of the liquid and gas phase area fractions measured from the simulations described in the main text and compare them to the value calculated from the theory presented in this section.

ℓ_o/d	χ	$\phi_{\text{sim}}^{\text{gas}}$	$\phi_{\text{sim}}^{\text{liquid}}$	$\phi_{\text{theory}}^{\text{gas}}$	$\phi_{\text{theory}}^{\text{liquid}}$
75	0.00	0.083	0.839	0.207	0.851
100	0.00	0.064	0.844	0.180	0.860
125	0.00	0.053	0.846	0.164	0.866
75	0.25	0.089	0.841	0.212	0.849
100	0.25	0.069	0.844	0.185	0.859
125	0.25	0.057	0.846	0.169	0.864
75	0.50	0.106	0.841	0.226	0.845
100	0.50	0.084	0.845	0.198	0.855
125	0.50	0.070	0.848	0.183	0.861
75	0.75	0.133	0.840	0.247	0.838
100	0.75	0.106	0.846	0.218	0.850
125	0.75	0.089	0.849	0.205	0.854

Table B.1: Comparison between binodal values measured in simulation and predicted in theory using the equations of state Eqs. B.35 and B.36.

While the qualitative trends, in particular widening of the binodal with increased ℓ_o and narrowing of the binodal with increased χ are correct, the quantitative agreement is poor particularly for the gas phase binodal values.

Thus far, we have considered macroscopic coexistence of cABPs which has an interface well approximated by a flat line. In the case of a finite droplet or bubble coexisting with a surrounding fluid, the interface will in general be circular. We therefore adapt the above results to cylindrical coordinates and solve for the corrections to the coexistence criteria due to interfaces with finite curvature. We will denote the density inside the circular bubble or droplet as ρ^{in} and the density of the surrounding fluid as ρ^{sat} . For the remainder of this section, when writing tensors in matrix form we will adopt the notation:

$$\mathbf{M} = \begin{bmatrix} M_{rr} & M_{r\theta} \\ M_{\theta r} & M_{\theta\theta} \end{bmatrix}. \quad (\text{B.37})$$

We can then write the density flux in cylindrical coordinates as:

$$\mathbf{J} = \frac{1}{\zeta} \nabla \cdot [\mathbf{T} + \chi \mathbf{T}^{\text{cross}} + \mathbf{T}^{\text{odd}}] \quad (\text{B.38a})$$

$$\mathbf{T} = \begin{bmatrix} -\mathcal{P} - \frac{\zeta U_o \ell_o \bar{U} Q_{rr}}{1+\chi^2} & -\frac{\zeta U_o \ell_o \bar{U} Q_{r\theta}}{1+\chi^2} \\ -\frac{\zeta U_o \ell_o \bar{U} Q_{r\theta}}{1+\chi^2} & -\mathcal{P} - \frac{\zeta U_o \ell_o \bar{U} Q_{\theta\theta}}{1+\chi^2} \end{bmatrix} \quad (\text{B.38b})$$

$$\mathbf{T}^{\text{cross}} = \begin{bmatrix} \frac{\chi \zeta U_o \ell_o \bar{U} Q_{r\theta}}{1+\chi^2} & -\frac{\chi \zeta U_o \ell_o \bar{U} Q_{rr}}{1+\chi^2} \\ -\frac{\chi \zeta U_o \ell_o \bar{U} Q_{rr}}{1+\chi^2} & -\frac{\chi \zeta U_o \ell_o \bar{U} Q_{r\theta}}{1+\chi^2} \end{bmatrix} \quad (\text{B.38c})$$

$$\mathbf{T}^{\text{odd}} = \begin{bmatrix} 0 & -\frac{\chi \zeta U_o \ell_o \bar{U} \rho}{2(1+\chi^2)} \\ \frac{\chi \zeta U_o \ell_o \bar{U} \rho}{2(1+\chi^2)} & 0 \end{bmatrix} \quad (\text{B.38d})$$

We now consider a radially symmetric state free of flux in the r direction. Such a state has no derivatives in the θ direction, so the components of the nematic order tensor can be solved for as:

$$Q_{rr} = -\frac{3}{16\zeta D_R} \left(\frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial r} \right)^2 + \frac{\chi^2}{8} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial r} \right)^2 + \bar{U} \frac{\partial p_C}{\partial \rho} \nabla^2 \rho \right), \quad (\text{B.39a})$$

$$Q_{\theta\theta} = -\frac{3}{16\zeta D_R} \left(\frac{\chi^2}{8} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial r} \right)^2 + \bar{U} \frac{\partial p_C}{\partial \rho} \nabla^2 \rho \right), \quad (\text{B.39b})$$

$$Q_{r\theta} = -\frac{3}{16\zeta D_R} \frac{\chi}{4} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \left(\frac{\partial \rho}{\partial r} \right)^2. \quad (\text{B.39c})$$

We now demand that $\mathbf{J} \cdot \mathbf{e}_r = 0$. The r component of the divergence of $\mathbf{T}^{\text{odd}}$ will be zero, so setting the r component of the flux results in:

$$0 = -\frac{\partial \mathcal{P}}{\partial \rho} \frac{\partial \rho}{\partial r} - \frac{\zeta U_o \ell_o}{1+\chi^2} \frac{\partial}{\partial r} [\bar{U} Q_{rr}] + \frac{\zeta U_o \ell_o \chi}{1+\chi^2} \frac{\partial}{\partial r} [\bar{U} Q_{r\theta}] + \frac{\zeta U_o \ell_o \bar{U}}{r(1+\chi^2)} [Q_{\theta\theta} - Q_{rr} + 2\chi Q_{r\theta}]. \quad (\text{B.40})$$

We can then integrate from the interior of the domain to the surroundings and arrive at an expression for the Laplace pressure difference:

$$\mathcal{P}(\rho^{\text{in}}) - \mathcal{P}(\rho^{\text{sat}}) = \frac{\gamma_{\text{mech}}}{R}, \quad (\text{B.41})$$

where we have defined the mechanical surface tension as:

$$\gamma_{\text{mech}} = \frac{3\ell_o^2}{16} \left(\frac{\chi^2}{2} - 1 \right) \int_0^\infty dr \left[\bar{U} \frac{\partial}{\partial \rho} \left(\bar{U} \frac{\partial p_C}{\partial \rho} \right) \left(\frac{\partial \rho}{\partial r} \right)^2 \right]. \quad (\text{B.42})$$

Note that a similar expression for the mechanical surface tension of achiral ABPs was found in Appendix C. The mechanical surface tension of achiral ABPs can be related to the mechanical surface tension of cABPs:

$$\gamma_{\text{mech}}^{\text{cABP}} = \frac{1 - \frac{\chi^2}{2}}{1 + \chi^2} \gamma_{\text{mech}}^{\text{ABP}}, \quad (\text{B.43})$$

which implies that at $\chi = \sqrt{2}$ the mechanical surface tension of cABPs transitions from a negative to a positive value. Eq. (B.41) is analogous to the pressure equality coexistence criterium found earilier in this section, but corrected to account for curvature. Similar to before, we can find the other coexistence crtierium by integrateing Eq. (B.40) indefinitely with respect to r :

$$\mathcal{P}(\rho(r)) = -\frac{\zeta U_o \ell_o}{1 + \chi^2} \bar{U} (Q_{rr} - \chi Q_{r\theta}) - F(r) + C. \quad (\text{B.44})$$

Here C is a constant of integration and we have defined $F(r)$ as:

$$F(r) = \frac{3\ell_o^2}{16} \left(\frac{\chi^2}{2} - 1 \right) \int_0^r dr \frac{1}{r} \left[\bar{U} \frac{\partial}{\partial \rho} \left(\bar{U} \frac{\partial p_C}{\partial \rho} \right) \left(\frac{\partial \rho}{\partial r} \right)^2 \right]. \quad (\text{B.45})$$

When $r = 0$, $F(r) = 0$ and when $r = \infty$, $F(r) = \gamma_{\text{mech}}/R$. The pressure must equal $\mathcal{P}(\rho^{\text{sat}})$ at $r = \infty$ and $\mathcal{P}(\rho^{\text{in}}) = \mathcal{P}(\rho^{\text{sat}}) + \gamma_{\text{mech}}/R$ at $r = 0$, so we can identify C as:

$$C = \mathcal{P}(\rho^{\text{sat}}) + \frac{\gamma_{\text{mech}}}{R} = \mathcal{P}(\rho^{\text{in}}). \quad (\text{B.46})$$

From this solution for C and by substituting the expressions for the nematic order components into Eq. (B.44), we find:

$$\mathcal{P}(\rho(r)) - \mathcal{P}(\rho^{\text{sat}}) - \frac{\gamma_{\text{mech}}}{R} + F(r) = a(\rho) \left(\frac{\partial}{\partial r} + \frac{1}{r} \right) \frac{\partial \rho}{\partial r} + b(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2. \quad (\text{B.47})$$

We now multiply Eq. (B.47) by $\partial \mathcal{E} / \partial r$ and integrate from $r = 0$ to $r = \infty$:

$$\begin{aligned} \int_{\rho^{\text{in}}}^{\rho^{\text{sat}}} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] \frac{\partial \mathcal{E}}{\partial \rho} d\rho - \frac{\gamma_{\text{mech}}}{R} (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}}) \\ + \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} F(r) = \int_0^\infty dr \frac{1}{r} a(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2. \end{aligned} \quad (\text{B.48})$$

The integral over $F(r)$ can be rewritten using integration by parts as:

$$\int_0^\infty \frac{\partial \mathcal{E}}{\partial r} F(r) dr = \mathcal{E}^{\text{sat}} \frac{\gamma_{\text{mech}}}{R} - \int_0^\infty \frac{1}{r} \mathcal{E} \mathcal{G} dr, \quad (\text{B.49})$$

where we have defined $\mathcal{G}$ as:

$$\mathcal{G} = \frac{3\ell_o^2}{16} \left(\frac{\frac{\chi^2}{2} - 1}{1 + \chi^2} \right) \left[\bar{U} \frac{\partial}{\partial \rho} \left(\bar{U} \frac{\partial p_C}{\partial \rho} \right) \left(\frac{\partial \rho}{\partial r} \right)^2 \right] \quad (\text{B.50})$$

Then Eq. (B.48) can be expressed as:

$$\int_{\rho^{\text{in}}}^{\rho^{\text{sat}}} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] \frac{\partial \mathcal{E}}{\partial \rho} d\rho = \frac{\gamma_{\text{ost}} \rho^{\text{sat}} (\mathcal{E}^{\text{in}} - \mathcal{E}^{\text{sat}})}{(\rho^{\text{in}} - \rho^{\text{sat}}) R}, \quad (\text{B.51})$$

where we have defined γ_{ost} , which has units of mechanical surface tension but is in general not equal, as:

$$\gamma_{\text{ost}} = \frac{(\rho^{\text{in}} - \rho^{\text{sat}})}{\rho^{\text{sat}} (\mathcal{E}^{\text{in}} - \mathcal{E}^{\text{sat}})} \int_0^\infty dr \left[a(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{G} (\mathcal{E} - \mathcal{E}^{\text{in}}) \right]. \quad (\text{B.52})$$

Together, Eqs. (B.41) and (B.51) represent the corrections to the coexistence criteria due to interfaces with finite curvature.

B.2 Interfacial Dynamics of Coexisting Chiral Active Particles

The fluctuating hydrodynamics of achiral ABPs have been laid out in Appendix A.1, following the procedures set by Refs. [53, 61]. These dynamics, valid when the magnitude of the flux $\mathbf{J}$ is small and on timescales larger than $1/D_R$, can straightforwardly be extended to include the external torque $\mathbf{M}_o$. In Appendix A.1 we found that the deterministic terms of the fluctuating dynamics would be exactly the same as those found via the Fokker-Planck equation, except sampled by a coarse-graining kernel $\Delta(\mathbf{r} - \mathbf{r}_i)$ rather than ensemble-averaged. Then the fluctuating hydrodynamics of chiral ABPs is given by:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \mathbf{J}, \quad (\text{B.53a})$$

$$\mathbf{J} = \frac{1}{\zeta} \left(\mathbf{T} + \mathbf{T}^{\text{cross}} + \mathbf{T}^{\text{odd}} \right) + \boldsymbol{\eta}^{\text{act}}, \quad (\text{B.53b})$$

where $\mathbf{T}$, $\mathbf{T}^{\text{cross}}$, and $\mathbf{T}^{\text{odd}}$ were provided in Section B.1 and $\boldsymbol{\eta}^{\text{act}}$ is a zero-average noise with covariance:

$$\langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \boldsymbol{\eta}^{\text{act}}(\mathbf{r}', t') \rangle = 2 \frac{k_B T^{\text{act}}}{\zeta} (\rho \mathbf{I} - 2\mathbf{Q}) \delta(t - t') \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{B.53c})$$

Here $k_B T^{\text{act}} \equiv \zeta U_o \ell_o / 2$ is the athermal energy scale of the fluctuations. It is worth noting that although $\mathbf{T}^{\text{odd}}$ enters the flux, it will never affect the overall density evolution as two divergences of an antisymmetric tensor must be zero. Despite $\mathbf{T}^{\text{odd}}$ not impacting the overall density evolution, the presence of parity-violating terms in $\mathbf{T}$, $\mathbf{T}^{\text{cross}}$ such as $\partial_x \rho \partial_y \rho$. It is convenient to express $\mathbf{T}$ in terms of derivatives of the density field. This can be done by substituting the closure of the traceless

nematic order Eq. (B.21) into Eq. (B.24):

$$\begin{aligned} \mathbf{T} = & \left[-\mathcal{P}(\rho) + a(\rho) \nabla^2 \rho + \frac{\chi}{2} b(\rho) \frac{\partial \rho}{\partial x} \frac{\partial \rho}{\partial y} + \frac{\chi^2}{8} b(\rho) \left[\left(\frac{\partial \rho}{\partial x} \right)^2 - \left(\frac{\partial \rho}{\partial y} \right)^2 \right] \right] \mathbf{I} \\ & + b(\rho) \nabla \rho \nabla \rho + \frac{\chi}{4} b(\rho) \left[\left(\frac{\partial \rho}{\partial x} \right)^2 - \left(\frac{\partial \rho}{\partial y} \right)^2 \right] (\mathbf{e}_x \mathbf{e}_y + \mathbf{e}_y \mathbf{e}_x), \end{aligned} \quad (\text{B.54})$$

where we have defined

$$a(\rho) = \frac{3\ell_o^2}{16(1+\chi^2)} \bar{U}^2 \frac{\partial p_C}{\partial \rho}, \quad (\text{B.55})$$

and

$$b(\rho) = \frac{3\ell_o^2}{16(1+\chi^2)} \bar{U} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right]. \quad (\text{B.56})$$

Additionally, $\mathbf{T}^{\text{cross}}$ can be compactly expressed as:

$$\mathbf{T}^{\text{cross}} = T_{xy} (\mathbf{e}_y \mathbf{e}_y - \mathbf{e}_x \mathbf{e}_x) + \chi (T_{xx} + \mathcal{P}) (\mathbf{e}_x \mathbf{e}_y + \mathbf{e}_y \mathbf{e}_x) \quad (\text{B.57})$$

We connect the dynamics of the density field to the dynamics of an interfacial height field via the ansatz [49]:

$$\rho(\mathbf{r}, t) = \varphi[x - h(y)], \quad (\text{B.58})$$

where φ is the stationary density profile solved for from the coexistence criteria. We proceed in the same manner as Appendix A.3, substituting Eq. (B.58) into Eqs. (B.53), Fourier transforming in the y direction, and applying the Green's function of the Laplace operator. The pseudovvariable which eliminates nonlinear terms in the interfacial dynamics is found to be different than the one used for the generalized Maxwell construction:

$$\frac{\partial \mathcal{E}_{\text{cw}}}{\partial \rho} \sim \left(\bar{U} \right)^{\frac{\chi^2}{4}} \left(\frac{\partial p_C}{\partial \rho} \right)^{1+\frac{\chi^2}{4}}. \quad (\text{B.59})$$

The pseudovvariable used to construct the binodals was chosen to eliminate nonlinear gradients in the x direction while the pseudovvariable used to derive the interfacial dynamics was chosen to eliminate nonlinear gradients in the y direction. The terms responsible for the $\chi^2/4$ exponent in the pseudovvariable came from a term which in the flux is proportional to a *difference* in the squared spatial gradients. Thus the pseudovvariable must have a corresponding antisymmetry for eliminating nonlinear gradients perpendicular to the interface as opposed to tangential to the interface. The difference in squared derivative terms leading to this antisymmetry are only possible in systems that violate spatial parity. Following integration with respect to $\mathcal{E}_{\text{cw}}$, we find the following interfacial dynamics:

$$\zeta_{\text{eff}} \frac{\partial h}{\partial t} = -k^2 |k| \gamma_{\text{cw}} h - i k |k| \nu_{\text{odd}} h + \xi^{\text{iso}} + \xi^{\text{aniso}} + \mathcal{O}(h^2 k^3), \quad (\text{B.60a})$$

where we have defined the capillary tension γ_{cw} , the odd flow coefficient ν_{odd} , an effective drag ζ_{eff} , and the noises $\chi^{\text{iso/aniso}}$ which arise from the result of the described mathematical procedure on $-\nabla \cdot \boldsymbol{\eta}^{\text{act}}$. The capillary tension γ_{cw} is found to be:

$$\gamma_{\text{cw}} = \frac{A(k)}{\rho^{\text{surf}} B(k)} \left[\int dx \frac{\partial \mathcal{E}_{\text{cw}}}{\partial x} a(\rho) \varphi' - \left(1 - \frac{\chi^2}{2} \right) \int dx \frac{\partial \mathcal{E}_{\text{cw}}}{\partial x} \int dx' e^{-|k||x-x'|} \text{sgn}(x-x') b(\rho) (\varphi')^2 \right], \quad (\text{B.60b})$$

the odd flow coefficient ν_{odd} is found as:

$$\begin{aligned} \nu_{\text{odd}} = \frac{A(k)}{\rho^{\text{surf}} B(k)} & \left[|k| \chi \int dx \frac{\partial \mathcal{E}_{\text{cw}}}{\partial x} \int dx' e^{-|k||x-x'|} b(\rho) (\varphi')^2 \right. \\ & + \frac{\chi}{2} \int dx \frac{\partial \mathcal{E}_{\text{cw}}}{\partial x} b(\rho) (\varphi')^2 \\ & \left. - k \chi \int dx \frac{\partial \mathcal{E}_{\text{cw}}}{\partial x} \int dx' e^{-|k||x-x'|} \text{sgn}(x-x') a(\rho) \varphi' \right], \end{aligned} \quad (\text{B.60c})$$

and we have defined the effective drag as:

$$\zeta_{\text{eff}} = \frac{\zeta A^2(k)}{2B(k)\rho^{\text{surf}}}, \quad (\text{B.60d})$$

where $\rho^{\text{surf}} \equiv (\rho^{\text{liq}} + \rho^{\text{gas}})/2$. The functions $A(k)$ and $B(k)$ are defined as:

$$A(k) = \int dx' \int dx e^{-|k||x-x'|} \varphi'(x) \frac{\mathcal{E}_{\text{cw}}}{\partial x'}, \quad (\text{B.60e})$$

$$B(k) = \int dx' \int dx e^{-|k||x-x'|} \frac{\partial \mathcal{E}_{\text{cw}}}{\partial x} \frac{\mathcal{E}_{\text{cw}}}{\partial x'}. \quad (\text{B.60f})$$

The noises are uncorrelated, with zero average and variances:

$$\langle \xi^{\text{iso}}(k, t) \xi^{\text{iso}}(k', t') \rangle = 2|k|k_B T^{\text{act}} \zeta_{\text{eff}} (2\pi) \delta(t - t') \delta(k + k'), \quad (\text{B.60g})$$

and

$$\langle \xi^{\text{aniso}}(k, t) \xi^{\text{aniso}}(k', t') \rangle = \frac{\zeta_{\text{eff}} (C(k) + D(k) + R(k))}{\rho^{\text{surf}} B(k)} (2\pi) \delta(t - t') \delta(k + k'). \quad (\text{B.60h})$$

Here we have defined the functions $C(k)$, $D(k)$, and $R(k)$ as:

$$\begin{aligned} C(k) = \int \int dud u' \frac{\partial \mathcal{E}_{\text{cw}}}{\partial u} \frac{\partial \mathcal{E}_{\text{cw}}}{\partial u'} \int \int dx dx' e^{-|k||u-x|} e^{-|k||u'-x'|} \\ \times \left(\frac{\varphi'' k^2 a(\rho)}{\bar{U}} \delta(x - x') - \frac{\partial}{\partial x} \left(\frac{\varphi'' a(\rho)}{\bar{U}} \frac{\partial}{\partial x} \delta(x - x') \right) \right), \end{aligned} \quad (\text{B.60i})$$

$$\begin{aligned} D(k) = \int \int dud u' \frac{\partial \mathcal{E}_{\text{cw}}}{\partial u} \frac{\partial \mathcal{E}_{\text{cw}}}{\partial u'} \int \int dx dx' e^{-|k||u-x|} e^{-|k||u'-x'|} \\ \times \frac{\partial}{\partial x} \left(\frac{(\varphi')^2 b(\rho)}{\bar{U}} \frac{\partial}{\partial x} \delta(x - x') \right), \end{aligned} \quad (\text{B.60j})$$

$$\begin{aligned}
R(k) = & \int \int du du' \frac{\partial \mathcal{E}_{\text{cw}}}{\partial u} \frac{\partial \mathcal{E}_{\text{cw}}}{\partial u'} \int \int dx dx' e^{-|k||u-x|} e^{-|k||u'-x'|} \\
& \times \left(\frac{(\varphi')^2 k^2 \chi^2 b(\rho)}{8\bar{U}} \delta(x-x') - \frac{\partial}{\partial x} \left(\frac{(\varphi')^2 \chi^2 b(\rho)}{8\bar{U}} \frac{\partial}{\partial x} \delta(x-x') \right) \right)
\end{aligned} \tag{B.60k}$$

The interfacial coefficients γ_{cw} and ν_{odd} are functions of ℓ_o , χ , and k . As discussed in the main text, we expect $\gamma_{\text{cw}}/k_B T^{\text{act}}$ and ν_{odd} to collapse across all values of χ when plotted against reduced run length $\lambda = \ell_o/\ell_o^c - 1$. We show these plots for $kd/2\pi = \{0, 0.1, 1.0\}$ in Fig. B.4, Fig. B.5, and Fig. B.6. The predicted collapse is most robust for $k \rightarrow 0$ but continues to be approximately correct even at the highest k s. In order to understand the k dependence of each coefficient, we calculate γ_{cw} , ν_{odd} , and ζ_{eff} normalized by their $k \rightarrow 0$ limits as a function of k for selected values of ℓ_o/d and χ and show the results in Figs. B.7 B.8, and B.9. From these calculations one can appreciate that the capillary tension converges to a value independent of k at long wavelengths, allowing for a clean k^{-2} scaling to be extracted from Eq. (B.72).

We now seek to determine whether the *deterministic* terms of Eq. (B.60a) are stable against the fluctuations induced by the stochastic terms. Breaking Eq. (B.60a) into real and imaginary components, we find:

$$\frac{\partial \text{Re}(h)}{\partial t} = -k^2 |k| \gamma_{\text{cw}} \text{Re}(h) + k |k| \nu_{\text{odd}} \text{Im}(h), \tag{B.61a}$$

$$\frac{\partial \text{Im}(h)}{\partial t} = -k |k| \nu_{\text{odd}} \text{Re}(h) - k^2 |k| \gamma_{\text{cw}} \text{Im}(h). \tag{B.61b}$$

The fixed point of these equations is $\text{Re}(h) = \text{Im}(h) = 0$. Then, rewriting $h = 0 + \delta h$, we find the dynamics of a small perturbation to the fixed point as:

$$\frac{\partial \text{Re}(\delta h)}{\partial t} = -k^2 |k| \gamma_{\text{cw}} \text{Re}(\delta h) + k |k| \nu_{\text{odd}} \text{Im}(\delta h), \tag{B.62a}$$

$$\frac{\partial \text{Im}(\delta h)}{\partial t} = -k |k| \nu_{\text{odd}} \text{Re}(\delta h) - k^2 |k| \gamma_{\text{cw}} \text{Im}(\delta h). \tag{B.62b}$$

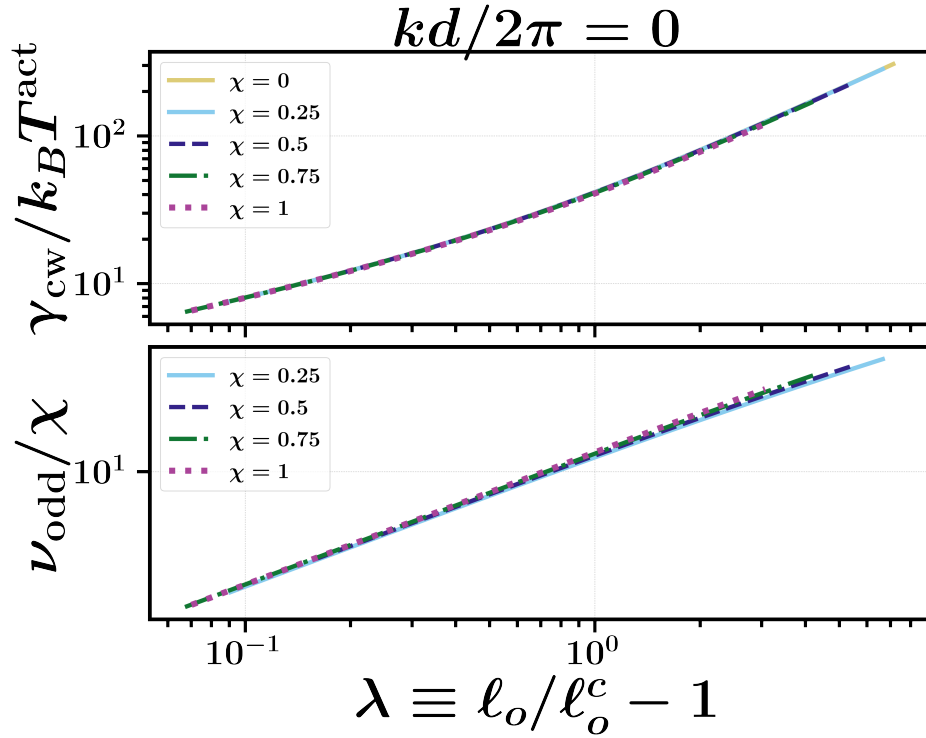


Figure B.4: $k \rightarrow 0$ limit of $\gamma_{\text{cw}}/k_B T^{\text{act}}$ and ν_{odd}/χ as a function of reduced run length.

The stability of the fixed point $h = 0$ can then be found by solving for the eigenvalues of the following matrix:

$$\begin{bmatrix} -k^2|k|\gamma_{\text{cw}} & k|k|\nu_{\text{odd}} \\ -k|k|\nu_{\text{odd}} & -k^2|k|\gamma_{\text{cw}} \end{bmatrix}. \quad (\text{B.63})$$

This matrix has two complex eigenvalues: $-k^2|k|\gamma_{\text{cw}} \pm ik|k|\nu_{\text{odd}}$. The real parts of these eigenvalues are negative on the condition γ_{cw} is positive. Therefore Eq. (B.60a) is linearly stable against fluctuations provided $\gamma_{\text{cw}} > 0$.

We now seek to describe the correlations of $h(k)$. We begin with Eq. (B.60a) evaluated at $+k$ and multiply by $h(-k)$:

$$\begin{aligned} \zeta_{\text{eff}} \frac{\partial h(k)}{\partial t} h(-k) = & -k^2|k|\gamma_{\text{cw}} h(k) h(-k) - ik|k|\nu_{\text{odd}} h(k) h(-k) \\ & + (\xi^{\text{iso}}(k) + \xi^{\text{aniso}}(k)) h(-k), \end{aligned} \quad (\text{B.64})$$

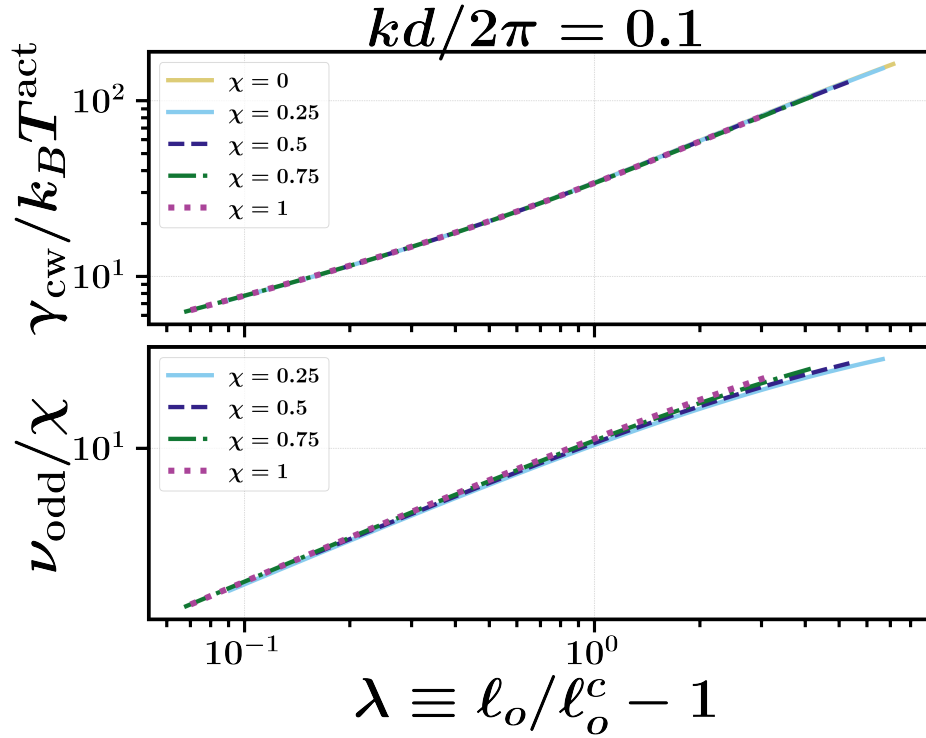


Figure B.5: $\gamma_{\text{cw}}/k_B T^{\text{act}}$ and ν_{odd}/χ when $kd/2\pi = 0.1$ as a function of reduced run length.

where we consider are considering low k and thus the k dependence of γ_{cw} , ζ_{eff} , and ν_{odd} are negligible. We also consider Eq. (B.60a) evaluated at $-k$ and multiply by $h(+k)$:

$$\zeta_{\text{eff}} \frac{\partial h(-k)}{\partial t} h(k) = -k^2 |k| \gamma_{\text{cw}} h(-k) h(k) + ik |k| \nu_{\text{odd}} h(-k) h(k) + (\xi^{\text{iso}}(-k) + \xi^{\text{aniso}}(-k)) h(k). \quad (\text{B.65})$$

Adding Eq. (B.64) to Eq. (B.65) results in:

$$\zeta_{\text{eff}} \frac{\partial}{\partial t} (h(k) h(-k)) = -2k^2 |k| \gamma_{\text{cw}} h(k) h(-k) + (\xi^{\text{iso}}(-k) + \xi^{\text{aniso}}(-k)) h(k) + (\xi^{\text{iso}}(k) + \xi^{\text{aniso}}(k)) h(-k). \quad (\text{B.66})$$

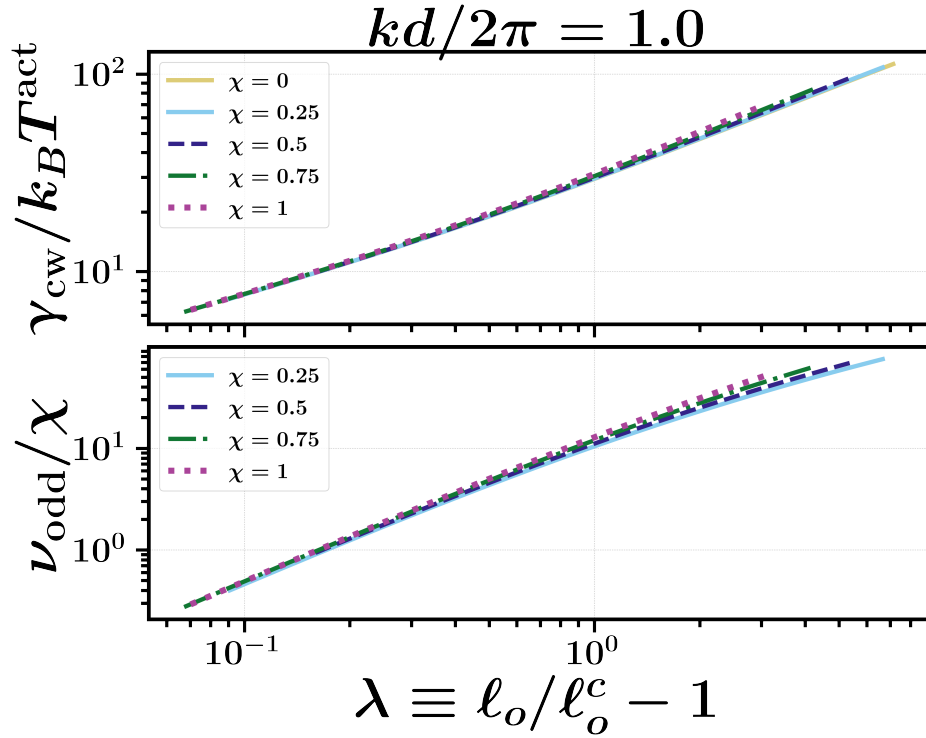


Figure B.6: $\gamma_{\text{cw}}/k_B T^{\text{act}}$ and ν_{odd}/χ when $kd/2\pi = 1.0$ as a function of reduced run length.

Averaging over the noise and taking the steady state results in:

$$2k^2|k|\gamma_{\text{cw}}\langle|h(k)|^2\rangle = \langle(\xi^{\text{iso}}(-k) + \xi^{\text{aniso}}(-k))h(k)\rangle + \langle(\xi^{\text{iso}}(k) + \xi^{\text{aniso}}(k))h(-k)\rangle. \quad (\text{B.67})$$

The implicit solution to Eq. (B.60a) is given by:

$$h(k, t) = h(k, 0) \exp \left[\frac{-t (k^2 |k| \gamma_{\text{cw}} + i k |k| \nu_{\text{odd}})}{\zeta_{\text{eff}}} \right] + \frac{1}{\zeta} \int_0^t dt' \exp \left[\frac{-(t-t') (k^2 |k| \gamma_{\text{cw}} + i k |k| \nu_{\text{odd}})}{\zeta_{\text{eff}}} \right] \left(\xi^{\text{iso}}(k, t') + \xi^{\text{aniso}}(k, t') \right) \quad (\text{B.68})$$

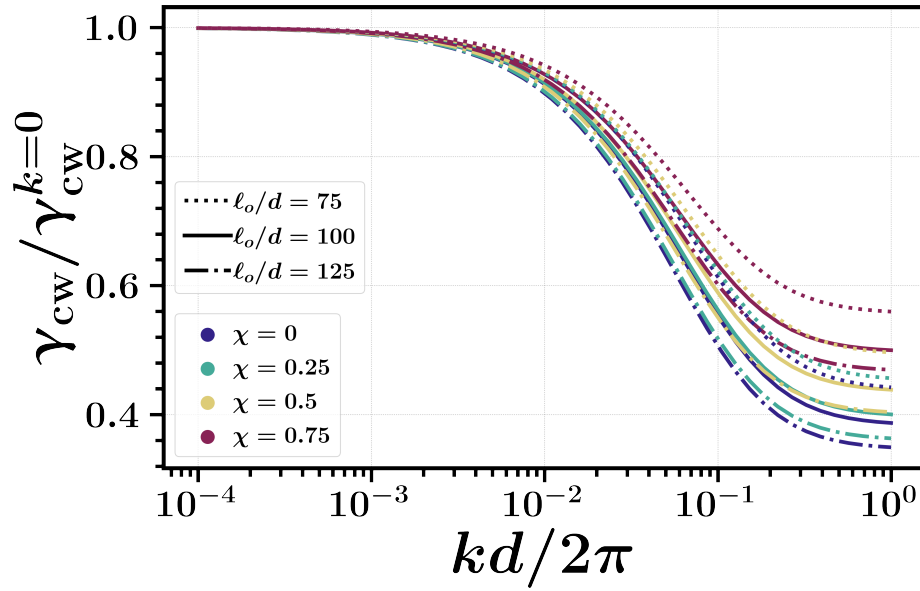


Figure B.7: γ_{cw} as a function of k and normalized by its $k \rightarrow 0$ limit for selected values of χ and ℓ_o/d .

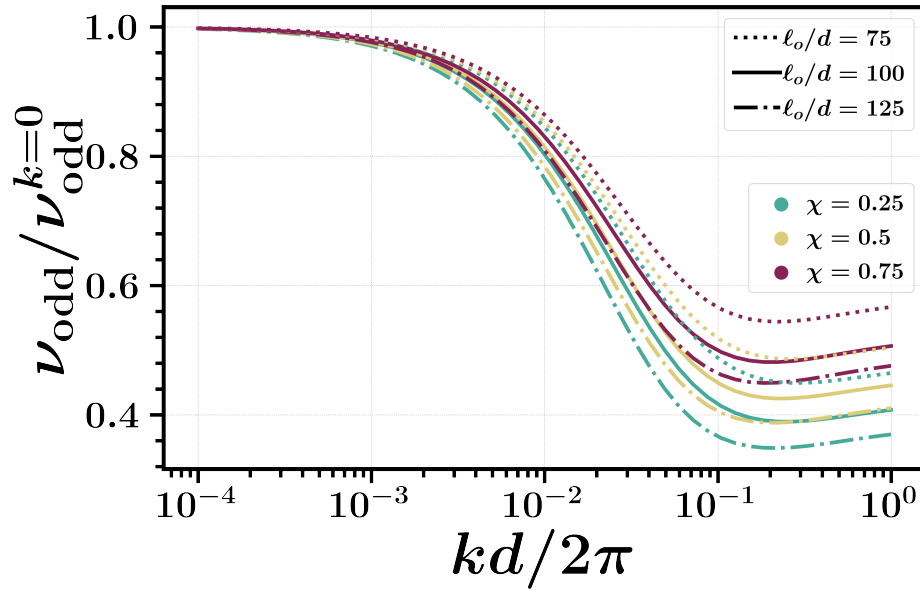


Figure B.8: ν_{odd} as a function of k and normalized by its $k \rightarrow 0$ limit for selected values of χ and ℓ_o/d .

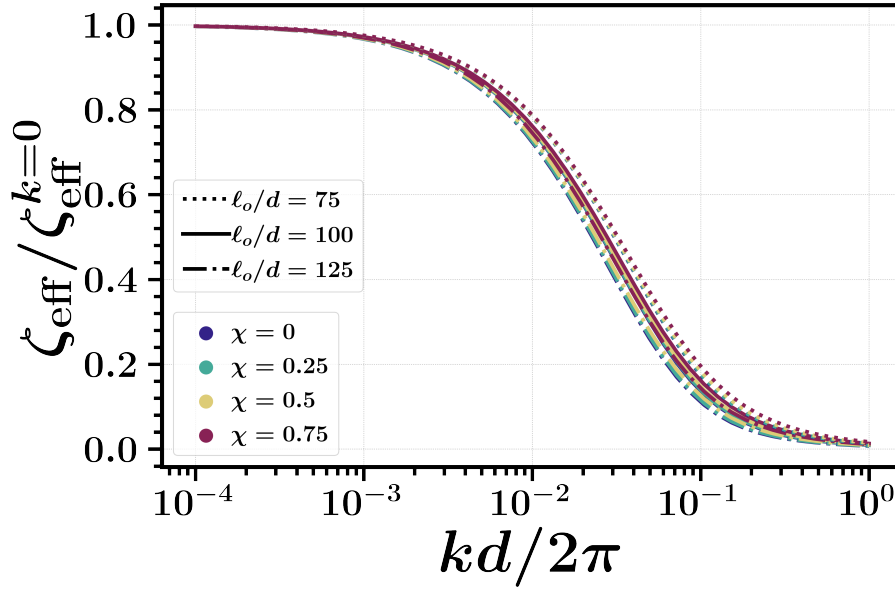


Figure B.9: ζ_{eff} as a function of k and normalized by its $k \rightarrow 0$ limit for selected values of χ and ℓ_o/d .

We now multiply by $(\xi^{\text{iso}}(-k, t) + \xi^{\text{aniso}}(-k, t))$ and average, finding:

$$\langle h(k, t)(\xi^{\text{iso}}(-k, t) + \xi^{\text{aniso}}(-k, t)) \rangle = |k|k_B T^{\text{act}} L + \frac{1}{2} \mathcal{A}(k), \quad (\text{B.69})$$

where we have defined:

$$\mathcal{A}(k) = L \frac{C(k) + D(k) + R(k)}{\rho^{\text{surf}} B(k)}. \quad (\text{B.70})$$

Then it is straightforward to show that:

$$\langle |h(k)|^2 \rangle = \frac{L|k|k_B T^{\text{act}} + 0.5\mathcal{A}(k)}{k^2 |k| \gamma_{\text{cw}}}, \quad (\text{B.71})$$

and in the limit $k\ell_o \ll 1$, where $\mathcal{A}$ was found to be negligible [42]:

$$\langle |h(k)|^2 \rangle = \frac{Lk_B T^{\text{act}}}{k^2 \gamma_{\text{cw}}}. \quad (\text{B.72})$$

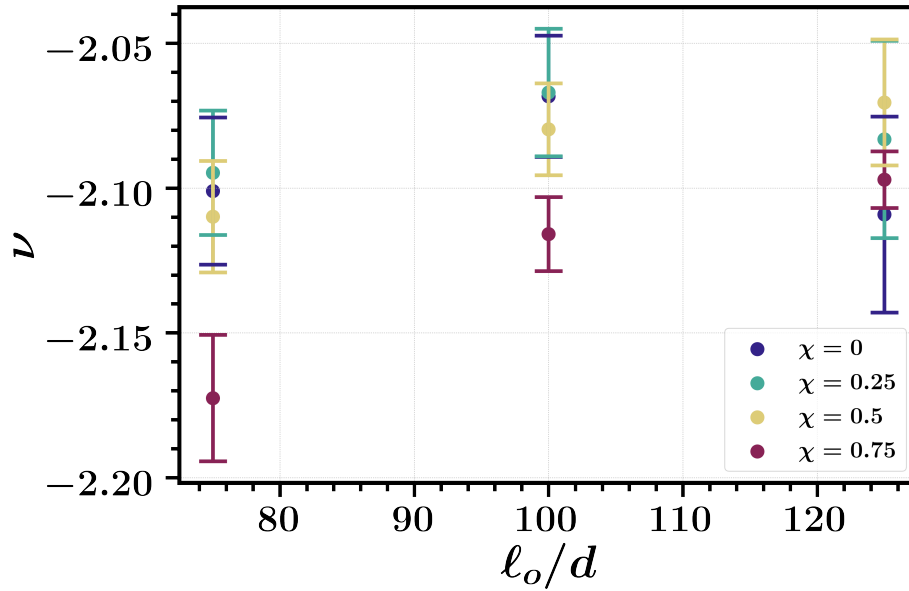


Figure B.10: ν as measured from a fit of the fluctuation spectrum to $\langle |h(k)|^2 \rangle = K_s k^\nu$. ν is near the theoretically predicted -2 for all combinations of ℓ_o and χ .

As discussed in the main text, we measure Eq. (B.72) from particle-based simulation and plotted the results in Fig. 3. We fit the measured fluctuation spectra to the power law form $\langle |h(k)|^2 \rangle = K_s k^\nu$ in order to extract a scaling with k . We repeated this calculation after splitting all simulation data into five evenly sized time epochs and calculated the standard deviation of the ν fit between the five epochs. The average result of ν and associated errorbars are plotted in Fig. B.10. Additionally, we compare the simulation spectra to that predicted by Eq. (B.72) and show the results in Fig. B.11 While the quantitative agreement is poor, likely due to our crude equation of state, the qualitative scaling and collapse across all values of ℓ_o and χ upon multiplication by λ are in agreement.

In order to determine dynamical information and the role of ν_{odd} , we now solve for the power spectrum. We will do so by Fourier transforming in time:

$$i\omega\zeta_{\text{eff}}h = -|k|^3\gamma_{\text{cw}}h + ik|k|\nu_{\text{odd}}h + \tilde{\chi}^{\text{iso}} + \tilde{\chi}^{\text{aniso}}, \quad (\text{B.73})$$

where the noise correlations of $\tilde{\chi}$ are the same as their time-space counterparts but with $\delta(t - t')$ switched with $\delta(\omega + \omega')$. We can now rearrange the time Fourier

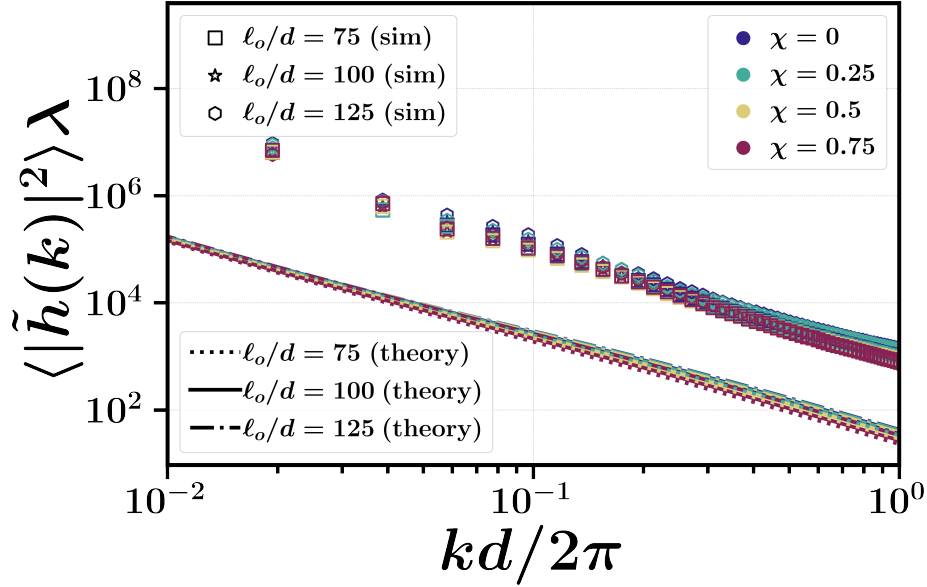


Figure B.11: $\langle |\tilde{h}(k)|^2 \rangle \lambda$ as measured from simulation and predicted by Eq. (B.72).

transformed evolution equation to find:

$$h(k, \omega) \left[|k|^3 \gamma_{\text{cw}} + i(\omega \zeta_{\text{eff}} + k|k| \nu_{\text{odd}}) \right] = \tilde{\chi}^{\text{iso}} + \tilde{\chi}^{\text{aniso}}. \quad (\text{B.74})$$

Now we multiply both sides of the above equation by its own complex conjugate ($h(-k, -\omega)$) and average over the noise, resulting in:

$$\begin{aligned} \langle |h(k, \omega)|^2 \rangle \left[|k|^6 \gamma_{\text{cw}}^2 + (\omega \zeta_{\text{eff}} + k|k| \nu_{\text{odd}})^2 \right] \\ = \langle \tilde{\chi}^{\text{iso}}(k, \omega) \tilde{\chi}^{\text{iso}}(-k, -\omega) \rangle + \langle \tilde{\chi}^{\text{aniso}}(k, \omega) \tilde{\chi}^{\text{aniso}}(-k, -\omega) \rangle. \end{aligned} \quad (\text{B.75})$$

Substituting the noise correlations [Eqs. (B.60g) and (B.60h)] into Eq. (B.75) results in:

$$\langle |h(k, \omega)|^2 \rangle \left[|k|^6 \gamma_{\text{cw}}^2 + (\omega \zeta_{\text{eff}} + k|k| \nu_{\text{odd}})^2 \right] = 2|k|k_B T^{\text{act}} \zeta_{\text{eff}} L \Gamma + \zeta_{\text{eff}} L \mathcal{A}(k), \quad (\text{B.76})$$

and in the limit $k\ell_o \ll 1$ where $\mathcal{A}$ is negligible the power spectrum reduces to:

$$\langle |h(k, \omega)|^2 \rangle = \frac{2|k|k_B T^{\text{act}} \zeta_{\text{eff}} L \Gamma}{|k|^6 \gamma_{\text{cw}}^2 + (\omega \zeta_{\text{eff}} + k|k| \nu_{\text{odd}})^2} \quad (\text{B.77})$$

The fluctuation spectra can be recovered via:

$$\begin{aligned}\langle |h(k)|^2 \rangle &= \frac{1}{2\pi\Gamma} \int_{-\infty}^{\infty} d\omega \langle |h(k, \omega)|^2 \rangle \\ &= \frac{1}{2\pi\Gamma} \int_{-\infty}^{\infty} d\omega \frac{2|k|k_B T^{\text{act}} L \Gamma}{\zeta_{\text{eff}} \left[\frac{|k|^6 \gamma_{\text{cw}}^2}{\zeta_{\text{eff}}^2} + \left(\omega + \frac{k|k|\nu_{\text{odd}}}{\zeta_{\text{eff}}} \right)^2 \right]}.\end{aligned}\quad (\text{B.78})$$

The above can be straightforwardly integrated by changing variables to $\omega' = \omega + k|k|\nu_{\text{odd}}/\zeta_{\text{eff}}$, and noting that:

$$\int_{-\infty}^{\infty} dx \frac{1}{a^2 + x^2} = \frac{\pi}{a}.\quad (\text{B.79})$$

The resulting expression for the fluctuation spectrum is then:

$$\langle |h(k)|^2 \rangle = \frac{|k|k_B T^{\text{act}} L}{\zeta_{\text{eff}} \pi} \frac{\pi}{\frac{\gamma_{\text{cw}} |k|^3}{\zeta_{\text{eff}}}} = \frac{k_B T^{\text{act}} L}{\gamma_{\text{cw}} k^2}.\quad (\text{B.80})$$

This is consistent with the fluctuation spectrum found in the previous section.

We now wish to determine whether the linearized interfacial dynamics exhibit surface area minimizing statistics in the steady-state. We do so by solving for the Fokker-Planck equation corresponding to Eq. (B.60a). Breaking Eq. (B.60a) into real and imaginary parts, recognizing that the noise variance is real, and following the arguments of Zwanzig [6] results in:

$$\frac{\partial P[\mathbf{h}]}{\partial t} = \frac{\partial}{\partial \mathbf{h}} \cdot \left[\begin{pmatrix} k^2 \gamma_{\text{cw}} & k \nu_{\text{odd}} \\ -k \nu_{\text{odd}} & k^2 \gamma_{\text{cw}} \end{pmatrix} \cdot \begin{pmatrix} \text{Re}(h) \\ \text{Im}(h) \end{pmatrix} P[\mathbf{h}] + \begin{pmatrix} \left(L k_B T^{\text{act}} + \frac{\mathcal{A}}{2|k|} \right) \frac{\partial P}{\partial \text{Re}(h)} \\ 0 \end{pmatrix} \right],\quad (\text{B.81})$$

where we are expressing $\mathbf{h} \equiv [\text{Re}(h), \text{Im}(h)]$. We now demand that the *steady-state* solution has no probabilistic flux at the boundaries, so our coupled PDE can be reduced to a system of equations, one of which being:

$$\left(-k \nu_{\text{odd}} \text{Re}(h) + k^2 \gamma_{\text{cw}} \text{Im}(h) \right) P[\mathbf{h}] = 0.\quad (\text{B.82})$$

This implies that at steady-state,

$$\text{Im}(h) = \frac{\nu_{\text{odd}} \text{Re}(h)}{k \gamma_{\text{cw}}}. \quad (\text{B.83})$$

We can substitute in this steady-state relation to the remaining equation, which is now an ordinary differential equation:

$$\left(k^2 \gamma_{\text{cw}} + \frac{\nu_{\text{odd}}^2}{\gamma_{\text{cw}}} \right) \text{Re}(h) P[\mathbf{h}] + \left(L k_B T^{\text{act}} + \frac{\mathcal{A}}{2|k|} \right) \frac{\partial P[\mathbf{h}]}{\partial \text{Re}(h)} = 0 \quad (\text{B.84})$$

The solution of this differential equation is:

$$P[\mathbf{h}] \sim \exp \left[- \frac{\frac{1}{2} \left(k^2 \gamma_{\text{cw}} + \frac{\nu_{\text{odd}}^2}{\gamma_{\text{cw}}} \right) \text{Re}(h)^2}{L k_B T^{\text{act}} + \frac{\mathcal{A}}{2|k|}} \right], \quad (\text{B.85})$$

and substituting in the steady-state relationship between the imaginary and real components of h allows one to find:

$$P[\mathbf{h}] \sim \exp \left[- \frac{k^2 \gamma_{\text{cw}} |\mathbf{h}|^2}{2 L k_B T^{\text{act}} + \frac{\mathcal{A}}{|k|}} \right]. \quad (\text{B.86})$$

In the limit $k \ell_o \ll 1$, where $\mathcal{A}$ was found to be negligible [42], the distribution takes on the simple Boltzmann form:

$$P[\mathbf{h}] \sim \exp \left[- \frac{k^2 \gamma_{\text{cw}} |\mathbf{h}|^2}{2 L k_B T^{\text{act}}} \right], \quad (\text{B.87})$$

recovering a surface-area minimizing Boltzmann distribution even in the presence of chirality.

B.3 Brownian Dynamics Simulations

Brownian dynamics simulations of over 10^5 cABPs were conducted at each combination of the parameters $\chi \in \{0.00, 0.25, 0.50, 0.75\}$ and $\ell_o/d \in \{75, 100, 125\}$. We choose $\mathbf{F}_{ij}$ to correspond to that of polydisperse hard disks [124, 161] with diameters set by a Gaussian distribution with average d and standard deviation $0.1d$ such that no hexatic phases are present and ρ can be taken as the sole order parameter of the phase separation. Hard-disk statistics were approximated by using a polydisperse WCA potential [161] with a fixed stiffness of $\mathcal{S} \equiv \epsilon/\zeta U_o d_{\text{LJ}} = 50$, where ϵ and d_{LJ} are the Lennard-Jones energy scale and (mean) diameter, respectively. The effective exclusion diameter is $d \equiv 2^{1/6} d_{\text{LJ}}$. Upon reaching a steady state of MIPS, we run simulations for an additional minimum duration of $6.6 \times 10^4 d/U_o$. Dynamical quantities were measured from simulations with durations of $2.4 \times 10^5 d/U_o$. All simulations were executed using the HOOMD-Blue software package [65].

The instantaneous shape of the interface $h(y)$ was measured using the algorithm developed by Patch *et al.* [25] every $4.45d/U_o$. $\tilde{h}(k)$ was then calculated from the discrete Fourier transform of $h(y)$, and the average quantity $\langle |\tilde{h}(k)|^2 \rangle$ is reported in Fig. 3.3(a). The average tangential flux profiles reported in Fig 3.3(b) were calculated by measuring the average density profiles and multiplying by the average velocity profile. We note that velocity is an ill-defined concept for particles undergoing Brownian dynamics, we therefore measured the y displacement of particles over a finite period of time, i.e. $(\mathbf{r}_i(t_2) - \mathbf{r}_i(t_1)) \cdot \mathbf{e}_y / (t_2 - t_1)$, similar to the method employed by Ref. [46]. A constant time separation of $t_2 - t_1 = 4.45d/U_o$ was chosen.

B.4 Media

Each video listed below displays the instantaneous interface determined from a Brownian dynamics simulation of cABP phase separation at a specified activity and chirality. The semi-transparent magenta region corresponds to the liquid phase

while the uncolored region corresponds to the gas phase. In each video, the instantaneous interface was computed using a cubic grid of points with a uniform spacing of $0.89 d_{\text{hs}}$ and a coarse-graining length of $\xi/d_{\text{hs}} = 1.78$. In each video, the instantaneous interface was computed using the algorithm developed by Patch *et al.* [25]. This method first employs a clustering algorithm to determine the particles within the liquid phase, then sorts each liquid particle into bins by its y coordinate. The x coordinate of the three particles with the highest x coordinate in each bin were then averaged, giving the location of the right interface corresponding to the y value associated with the bin. The x coordinate of the three particles with the lowest x coordinate in each bin were then averaged, giving the location of the left interface corresponding to the y value associated with the bin. Each frame is separated by a duration of $4.45 d_{\text{hs}}/U_o$ and the videos are played at a rate of 100 frames per second. All videos are publicly available at the following URL:
<https://berkeley.box.com/s/7befhar83g2nnl3ci8sdt7rkc6hb3aqh>

- [interface_75LR_0chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 75$ and a chirality of $\chi = 0$.
- [interface_75LR_025chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 75$ and a chirality of $\chi = 0.25$.
- [interface_75LR_05chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 75$ and a chirality of $\chi = 0.5$.
- [interface_75LR_075chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 75$ and a chirality of $\chi = 0.75$.

- [interface_100LR_0chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 100$ and a chirality of $\chi = 0$.
- [interface_100LR_025chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 100$ and a chirality of $\chi = 0.25$.
- [interface_100LR_05chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 100$ and a chirality of $\chi = 0.5$.
- [interface_100LR_075chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 100$ and a chirality of $\chi = 0.75$.
- [interface_125LR_0chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 125$ and a chirality of $\chi = 0$.
- [interface_125LR_025chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 125$ and a chirality of $\chi = 0.25$.
- [interface_125LR_05chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 125$ and a chirality of $\chi = 0.5$.
- [interface_125LR_075chi.mp4]:
Instantaneous interface dynamics with an activity of $\ell_o/d = 125$ and a chirality of $\chi = 0.75$.

Appendix C

Appendix: Active Nucleation

The contents of Appendix C will closely follow that of the Supplemental Information of Ref. [108]. We will present the details of the mechanical derivation of the profile of the critical nucleus, the stochastic dynamics of a critical nucleus, the equilibrium limit of both of these theories, and also present the framework for understanding active coarsening.

C.1 Mechanics of the Critical Nucleus

We aim to describe the structure of a critical nucleus with density ρ^{in} in a system that has been prepared at a supersaturation density ρ^{sat} . We assume that the critical nucleus is sufficiently small compared to the system size such that the density outside the nucleus is well approximated by the supersaturation density ρ^{sat} . In addition, we approximate the geometry of the critical nucleus as a perfect sphere with radius R_C .

When the critical nucleus is an unstable fixed point, a nucleus smaller than the critical nucleus will experience a flux normal to its interface such that the nucleus shrinks. A nucleus larger than the critical nucleus will experience *the opposite* flux normal to the interface such that the nucleus grows. Therefore, in order to solve for the critical nucleus, we seek states which have zero flux normal to the interface. Even if the critical nucleus is a stable fixed point, as recently found

in some active systems [39], similar arguments may be used to still conclude the critical nucleus will be flux-free. Two flux-free states, the macroscopically phase separated and metastable homogeneous states, are already known. The critical nucleus will correspond to a state with a *finite-size* minority phase and zero flux normal to the interface. The evolution of the density flux $\mathbf{J} = \rho(\mathbf{r}, t)\mathbf{u}(\mathbf{r}, t)$, where $\mathbf{u}$ is the number-averaged velocity of particles, can be expressed as a balance of linear momentum:

$$\frac{\partial (m\mathbf{J})}{\partial t} = -\nabla \cdot \left(\frac{m\mathbf{J}\mathbf{J}}{\rho} \right) + \nabla \cdot \boldsymbol{\sigma} + \mathbf{b}, \quad (\text{C.1})$$

where m is the mass of each particle, $\boldsymbol{\sigma}$ is the stress tensor and $\mathbf{b}$ are the body forces acting on the particles. Additionally, we assume that $\boldsymbol{\sigma}$ and $\mathbf{b}$ can be written as local gradient expansions, which is typically valid when interactions are short-ranged. For an *overdamped* system with spherical symmetry and zero flux in the radial direction, Eq. (C.1) reduces to $\nabla \cdot \boldsymbol{\sigma} + \mathbf{b} = \mathbf{0}$. Exact microscopic expressions for both $\boldsymbol{\sigma}$ and $\mathbf{b}$ can be determined through an Irving-Kirkwood-Noll procedure [120–122]. These microscopic expressions can include terms that appear to be non-local, requiring integrals over all space to, for instance, compute the stress at a particular point in space arising from particle interactions. However, these terms are often well-approximated by a local gradient expansion, the coefficients of which can be systematically determined [123]. We thus focus on systems in which $\boldsymbol{\sigma}$ and $\mathbf{b}$ can be fully expressed as locally. For spatially isotropic systems, $\boldsymbol{\sigma}$ can be expanded to second order in density gradients as:

$$\boldsymbol{\sigma} = [\alpha_0(\rho)\mathbf{I} + \alpha_1(\rho)\nabla^2\rho + \alpha_2(\rho)|\nabla\rho|^2]\mathbf{I} + \alpha_3(\rho)\nabla\rho\nabla\rho + \alpha_4(\rho)\nabla\nabla\rho, \quad (\text{C.2})$$

and $\mathbf{b}$ can be expanded to third order in density gradients as:

$$\mathbf{b} = \beta_0(\rho)\nabla\rho + \beta_1(\rho)\nabla\rho\nabla^2\rho + \beta_2(\rho)\nabla|\nabla\rho|^2 + \beta_3(\rho)\nabla\nabla^2\rho + \beta_4(\rho)|\nabla\rho|^2\nabla\rho. \quad (\text{C.3})$$

Up to a divergence-free term, we may absorb the body force into a stress-like contribution such that:

$$\nabla \cdot \boldsymbol{\sigma} + \mathbf{b} = \nabla \cdot [\boldsymbol{\sigma} + \mathbf{F}] \equiv \nabla \cdot \boldsymbol{\Sigma} = \mathbf{0}, \quad (\text{C.4a})$$

$$\mathbf{F} = \frac{1}{4\pi} \int d\mathbf{r}' \mathbf{b}(\mathbf{r}') \frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3}, \quad (\text{C.4b})$$

where we have used the Green's function identity $\nabla \cdot ((\mathbf{r} - \mathbf{r}')/|\mathbf{r} - \mathbf{r}'|^3) = 4\pi\delta(\mathbf{r} - \mathbf{r}')$ in three dimensions. Details on evaluating the integral Eq. (C.4b) are provided later in this Appendix. In general, $\mathbf{F}$ will contain terms which may be expressed as *local* density gradient expansions as well as *non-local* terms which depend on spatial integrals over the entire density field. Due to spherical symmetry, Σ has only two independent components and is given by:

$$\Sigma = \begin{bmatrix} \Sigma_{rr} & 0 & 0 \\ 0 & \Sigma_{tt} & 0 \\ 0 & 0 & \Sigma_{tt} \end{bmatrix} = \Sigma_{rr} \mathbf{e}_r \otimes \mathbf{e}_r + \Sigma_{tt} (\mathbf{I} - \mathbf{e}_r \otimes \mathbf{e}_r). \quad (\text{C.5})$$

We now find the flux-free steady states by demanding the dynamic stress tensor has zero divergence, and we expand the stress tensor to second order in density gradients:

$$\nabla \cdot \Sigma = \mathbf{0}, \quad (\text{C.6a})$$

$$\Sigma_{rr} = -\mathcal{P}(\rho) + \kappa_1(\rho) \left(\frac{\partial}{\partial r} + \frac{2}{r} \right) \frac{\partial \rho}{\partial r} + (\kappa_2(\rho) + \kappa_3(\rho)) \left(\frac{\partial \rho}{\partial r} \right)^2 + \kappa_4(\rho) \frac{\partial^2 \rho}{\partial r^2} + \mathcal{K}, \quad (\text{C.6b})$$

$$\Sigma_{tt} = -\mathcal{P}(\rho) + \kappa_1(\rho) \left(\frac{\partial}{\partial r} + \frac{2}{r} \right) \frac{\partial \rho}{\partial r} + \kappa_2(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{K}, \quad (\text{C.6c})$$

$$\mathcal{P} = -\alpha_0(\rho) - \int \beta_0(\rho) d\rho, \quad (\text{C.6d})$$

$$\kappa_1(\rho) = \alpha_1(\rho) + \beta_3(\rho), \quad (\text{C.6e})$$

$$\kappa_2(\rho) = \alpha_2(\rho) + \frac{1}{2} \frac{\partial \beta_3(\rho)}{\partial \rho} + \beta_2(\rho) - \frac{1}{2} \beta_1(\rho), \quad (\text{C.6f})$$

$$\kappa_3(\rho) = \alpha_3(\rho) + \beta_1(\rho) - \frac{\partial \beta_3}{\partial \rho}, \quad (\text{C.6g})$$

$$\kappa_4(\rho) = \alpha_4(\rho), \quad (\text{C.6h})$$

$$\mathcal{K} = \int_r^\infty \left[-\beta_4(\rho) + \frac{\partial \beta_2}{\partial \rho} + \frac{1}{2} \frac{\partial \beta_1}{\partial \rho} - \frac{1}{2} \frac{\partial^2 \beta_3}{\partial \rho^2} \right] \left(\frac{\partial \rho}{\partial r'} \right)^3 dr'. \quad (\text{C.6i})$$

Here, $\mathcal{P}(\rho)$ is the bulk isotropic contribution to Σ , which is the dynamic pressure, and $\mathcal{K}(r)$ is the non-local terms obtained from evaluating Eq. (C.4b). We note that $\mathcal{K}$ is zero when evaluated outside of the nucleus but finite when evaluated inside of the nucleus. In spherical coordinates, the divergence of Σ is given by:

$$0 = \frac{\partial \Sigma_{rr}}{\partial r} - \frac{2}{r} \mathcal{G}(\rho), \quad (\text{C.7})$$

where we have defined the “excess stress”:

$$\mathcal{G}(\rho) = \Sigma_{tt} - \Sigma_{rr}. \quad (\text{C.8})$$

The definite integral of $\mathcal{G}(\rho)$ across the interface is precisely the mechanical surface found by Kirkwood and Buff [26]:

$$\gamma_{\text{mech}} = \int_0^\infty [\Sigma_{tt} - \Sigma_{rr}] dr. \quad (\text{C.9})$$

For active particles with hard-sphere interactions, this mechanical surface tension was found to be strongly *negative* [24].

Let us consider the definite integral of Eq. (C.7) from $r = 0$ to ∞ :

$$\mathcal{P}(\rho^{\text{in}}) - \mathcal{P}(\rho^{\text{sat}}) - \mathcal{K}|_{r=0} = \int_0^\infty \left[\frac{2}{r} \mathcal{G}(\rho) \right] dr. \quad (\text{C.10})$$

Within a region of homogeneous density, Σ becomes isotropic. Therefore $\mathcal{G}$ is only non-zero in the vicinity of the interface. If the width of the interfacial region is small compared to the radius of the critical nucleus, then we may approximate Eq. (C.10) as:

$$\mathcal{P}(\rho^{\text{in}}) - \mathcal{P}(\rho^{\text{sat}}) - \mathcal{K}|_{r=0} = \frac{2\gamma_{\text{mech}}}{R_C} + O\left(\frac{1}{R_C^2}\right), \quad (\text{C.11})$$

which demonstrates that the Laplace pressure difference holds in active systems *only without* non-local stress components and is broken by systems *with* non-local stress components. A derivation of an active Laplace pressure difference was discussed in Ref. [19], however this work did not consider the contribution of non-local stress terms.

Suppose we had taken the *antiderivative* of Eq. (C.7) rather than the definite integral. Doing so results in:

$$\begin{aligned} \Sigma_{rr}(r) = -\mathcal{P}(\rho) + \kappa_1(\rho) \left[\frac{\partial}{\partial r} + \frac{2}{r} \right] \frac{\partial \rho}{\partial r} + (\kappa_2(\rho) + \kappa_3(\rho)) \left(\frac{\partial \rho}{\partial r} \right)^2 \\ + \kappa_4(\rho) \frac{\partial^2 \rho}{\partial r^2} + \mathcal{K} = W(r) + C, \end{aligned} \quad (\text{C.12})$$

where C is a constant of integration and we have defined W as:

$$W(r) = \int_0^r \left[\frac{2\mathcal{G}}{r} \right] dr. \quad (\text{C.13})$$

While we do not know the value of $W(r)$ for any arbitrary r , we have already approximated $W(\infty)$ as $2\gamma_{\text{mech}}/R_C$ when R_C is much larger than the interfacial width. In addition, L'Hôpital's rule can be used to show that the integrand of $W(r)$ is zero at $r = 0$, which implies that $W(0) = 0$. In order to determine the precise value of C , we recall that the density as $r \rightarrow \infty$ is ρ^{sat} :

$$\Sigma_{rr}(\infty) = W(\infty) + C = \frac{2\gamma_{\text{mech}}}{R_C} + C = -\mathcal{P}(\rho^{\text{sat}}) \quad (\text{C.14})$$

Therefore

$$C = -\mathcal{P}(\rho^{\text{sat}}) - \frac{2\gamma_{\text{mech}}}{R_C} \quad (\text{C.15})$$

We are then left with the equation:

$$\begin{aligned} \kappa_1(\rho) \left[\frac{\partial}{\partial r} + \frac{2}{r} \right] \frac{\partial \rho}{\partial r} + (\kappa_2(\rho) + \kappa_3(\rho)) \left(\frac{\partial \rho}{\partial r} \right)^2 + \kappa_4(\rho) \frac{\partial^2 \rho}{\partial r^2} + \mathcal{K} \\ = W(r) - \frac{2\gamma_{\text{mech}}}{R_C} + (\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})). \end{aligned} \quad (\text{C.16})$$

We now introduce the generalized Maxwell construction pseudovariable [20, 21, 71] $\mathcal{E}$ defined by the differential equation:

$$(\kappa_1(\rho) + \kappa_4(\rho)) \frac{\partial^2 \mathcal{E}}{\partial \rho^2} = \left(2(\kappa_2(\rho) + \kappa_3(\rho)) - \frac{\partial}{\partial \rho} (\kappa_1(\rho) + \kappa_4(\rho)) \right) \frac{\partial \mathcal{E}}{\partial \rho} \quad (\text{C.17})$$

For passive systems, $\{\kappa_i(\rho)\}$ are dictated by the Korteweg expansion [57], resulting in $\mathcal{E} \sim 1/\rho$. This pseudovariable was found to eliminate the need to evaluate interfacial terms when solving for the macroscopic binodals, thus providing coexistence criteria in terms of bulk equations of state. In our case multiplying by the

derivative of the pseudovisible with respect to r and integrating from $r = 0$ to $r = \infty$ will eliminate some, but not all, of the interfacial terms, resulting in:

$$\begin{aligned} \int_0^\infty \left[\frac{2\kappa_1(\rho)}{r} \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{K} \frac{\partial \mathcal{E}}{\partial \rho} \frac{\partial \rho}{\partial r} \right] dr &= \int_0^\infty \frac{\partial \mathcal{E}}{\partial r} W(r) dr - \frac{2\gamma_{\text{mech}}}{R_C} (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}}) \\ &+ \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] d\mathcal{E}. \end{aligned} \quad (\text{C.18})$$

We now integrate the first integral on the right hand side of Eq. (C.18) by parts to find:

$$\int_0^\infty \frac{\partial \mathcal{E}}{\partial r} W(r) dr = \mathcal{E}^{\text{sat}} \frac{2\gamma_{\text{mech}}}{R_C} - \int_0^\infty \frac{2}{r} \mathcal{E} \mathcal{G} dr. \quad (\text{C.19})$$

We also integrate the second integral on the left hand side of Eq. (C.18) by parts to find:

$$\int_0^\infty \mathcal{K} \frac{\partial \mathcal{E}}{\partial r} dr = -\mathcal{E}^{\text{in}} \mathcal{K}|_{r=0} - \int_0^\infty \frac{\partial \mathcal{K}}{\partial r} \mathcal{E} dr. \quad (\text{C.20})$$

Substitution of Eqs. (C.19) and (C.20) into Eq. (C.18) results in:

$$\begin{aligned} \int_0^\infty \left[\frac{2\kappa_1(\rho)}{r} \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 - \frac{\partial \mathcal{K}}{\partial r} \mathcal{E} \right] dr &= \mathcal{E}^{\text{in}} \mathcal{K}|_{r=0} - \int_0^\infty \frac{2}{r} \mathcal{E} \mathcal{G} dr \\ &+ \frac{2\gamma_{\text{mech}}}{R_C} \mathcal{E}^{\text{in}} + \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] d\mathcal{E}. \end{aligned} \quad (\text{C.21})$$

Finally, we apply our approximation that R_C is much larger than the interfacial width to find:

$$\begin{aligned} \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] d\mathcal{E} &+ \int_0^\infty (\mathcal{E}^{\text{in}} - \mathcal{E}) \frac{\partial \mathcal{K}}{\partial r} dr \\ &= \frac{2}{R_C} \int_0^\infty \left[\kappa_1(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{G} (\mathcal{E} - \mathcal{E}^{\text{in}}) \right], \end{aligned} \quad (\text{C.22})$$

where by making this approximation we have introduced error $O(R_C^{-2})$. We now define the “Ostwald” tension [39] as:

$$\gamma_{\text{ost}} = \frac{\rho^{\text{sat}} - \rho^{\text{in}}}{\rho^{\text{sat}} (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}})} \left[\int_0^\infty \left(\kappa_1(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{G}(\mathcal{E} - \mathcal{E}^{\text{in}}) \right) dr \right], \quad (\text{C.23})$$

where γ_{ost} has the same units of mechanical surface tension. We show in Section A.1 that γ_{ost} becomes identical to γ_{mech} in a passive system where $\mathcal{E} \sim 1/\rho$. Using the definition of the Ostwald tension, Eq. (C.22) can be rewritten as:

$$\begin{aligned} \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] d\mathcal{E} + \int_0^\infty (\mathcal{E}^{\text{in}} - \mathcal{E}) \frac{\partial \mathcal{K}}{\partial r} dr \\ = \frac{2\gamma_{\text{ost}} \rho^{\text{sat}} (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}})}{R_C \rho^{\text{sat}} - \rho^{\text{in}}} + O\left(\frac{1}{R_C^2}\right). \end{aligned} \quad (\text{C.24})$$

In addition, we restate the Laplace pressure difference found above:

$$\mathcal{P}(\rho^{\text{in}}) - \mathcal{P}(\rho^{\text{sat}}) - \mathcal{K}|_{r=0} = \frac{2\gamma_{\text{mech}}}{R_C} + O\left(\frac{1}{R_C^2}\right). \quad (\text{C.25})$$

Any flux-free state will satisfy Eqs. (C.25) and (C.24) simultaneously. The solution with $R_C = \infty$ corresponds to macroscopic phase separation and reduces to the phase coexistence criteria found in Ref. [21]. If a system is prepared at a saturation density ρ^{sat} not equal to the binodal, simultaneous solution of Eqs. (C.25) Eq. (C.22) allows for the calculation of the critical nucleus density ρ^{in} and *finite* critical radius R_C .

Simultaneous solution of Eqs. (C.25) and (C.22) involves a serious complication. The right hand sides of both equation can only be evaluated if the density profile of the critical nucleus is already known, which in turn depends on R_C and ρ^{in} . Therefore a *self-consistent* approach must be employed, which we outline in later in this Appendix.

We now apply the above framework to active Brownian particles defined identically to Eqs. (A.1). By following a similar procedure to that discussed in Ref. [21], one can find an expression for the flux $\mathbf{J}$ of ABPs, which is dependent on the polar order $\mathbf{m} \equiv \langle \sum_i \mathbf{q}_i \delta(\mathbf{r} - \mathbf{r}') \rangle$. The flux of the polar order is then found to be dependent on the nematic order $\mathbf{Q} \equiv \langle \sum_i \mathbf{q}_i \mathbf{q}_i \delta(\mathbf{r} - \mathbf{r}_i) \rangle$. Thus, each orientational moment of the density field has dynamics dependent on the field one moment higher. Thus, finding a form for the ABP flux dependent solely on the density requires one to truncate this hierarchy of equations with some closure relation. By setting $\mathbf{B} \equiv \langle \sum_i \mathbf{q}_i \mathbf{q}_i \mathbf{q}_i \delta(\mathbf{r} - \mathbf{r}_i) \rangle$ isotropic and demanding that $\mathbf{J} \cdot \mathbf{e}_r = 0$, we are able to solve for the form of $\mathcal{P}(\rho)$ and $\{\kappa_i(\rho)\}$ for ABPs:

$$\mathcal{P}(\rho) = p_C(\rho) + \frac{\rho \zeta U_o \ell_o \bar{U}(\rho)}{d(d-1)}, \quad (\text{C.26a})$$

$$\kappa_1(\rho) = \frac{3\ell_o^2}{2d(d-1)(d+2)} \left(\bar{U}(\rho) \right)^2 \frac{\partial p_C}{\partial \rho}, \quad (\text{C.26b})$$

$$\kappa_2(\rho) = 0, \quad (\text{C.26c})$$

$$\kappa_3(\rho) = \frac{3\ell_o^2}{2d(d-1)(d+2)} \bar{U}(\rho) \frac{\partial}{\partial \rho} \left[\bar{U}(\rho) \frac{\partial p_C}{\partial \rho} \right], \quad (\text{C.26d})$$

$$\kappa_4(\rho) = 0, \quad (\text{C.26e})$$

$$\mathcal{K} = 0. \quad (\text{C.26f})$$

Here $p_C(\rho)$ is the *conservative* pressure arising from particle interactions, $\bar{U}(\rho)$ is the dimensionless effective active speed, and $d(> 1)$ is the system dimensionality. Note that this form is identical to that found in Appendix A.1 but without any noise terms. In Eqs. (C.26) we have ignored the gradient contributions of the interaction potential to Σ , which is justified when $\ell_o \gg 1$ [19, 29, 64]. Given our expressions for $\{\kappa_i(\rho)\}$, we can solve for the generalized Maxwell construction pseudovalue as $\mathcal{E} \sim p_C(\rho)$. The closures used to derive these forms of $\mathcal{P}(\rho)$ and $\{\kappa_i(\rho)\}$ imply that finite polar in the radial direction and nematic ordering in the tangential direction exists at interfaces in states free of radial flux. We note that the theory derived in this appendix is valid for small supersaturations, i.e. the overall density

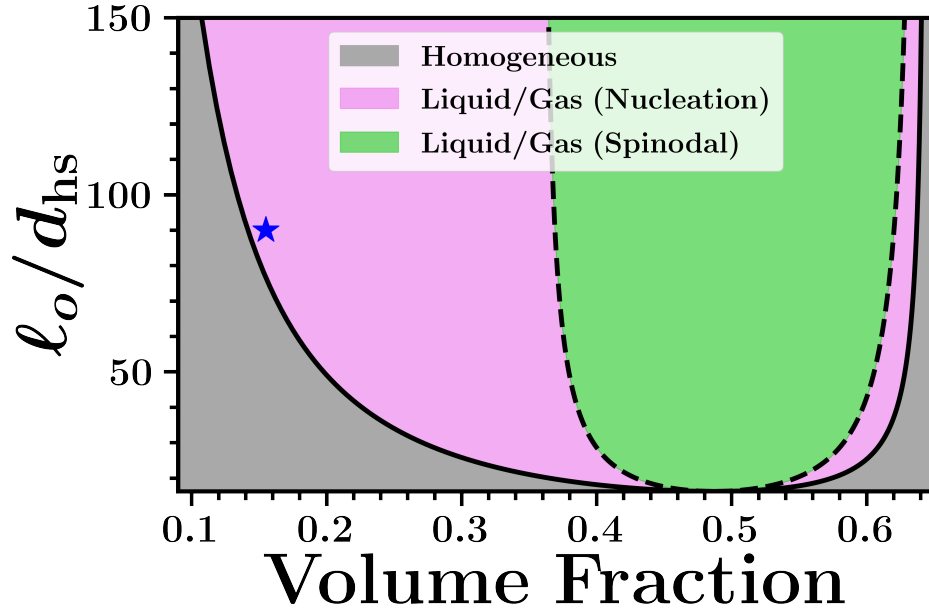


Figure C.1: Mechanically constructed phase diagram as calculated by Ref. [21]. Dashed line indicates spinodals while solid line indicates binodals. Blue star included as an example point where supersaturation is small.

of the system is close to the binodals. For reference, we include the phase diagram of ABPs as solved for by Ref. [21] in Fig. C.1 and include a blue star in the figure representing a point near the binodal where our theory may hold.

In order to numerically solve for the quantities derived in this supplemental material, we use the equations of state reported Ref. [21] for athermal active hard spheres. The two contributions to the bulk dynamic pressure were found to be well

described by:

$$\begin{aligned} \frac{p_{\text{act}}}{\zeta U_0/(d_{\text{hs}}^2)} &= \phi \left(\frac{\ell_0}{d_{\text{hs}}} \right) \frac{1}{\pi} \bar{U} \\ &= \phi \left(\frac{\ell_0}{\mathcal{D}} \right) \frac{1}{\pi} \left[1 + \left(1 - \exp \left[-2^{7/6} \left(\frac{\ell_0}{d_{\text{hs}}} \right) \right] \right) \frac{\phi}{1 - \phi/\phi_{\text{max}}} \right]^{-1} \end{aligned} \quad (\text{C.27a})$$

$$\frac{p_{\text{C}}}{\zeta U_0/(d_{\text{hs}}^2)} = 6 \times 2^{-7/6} \frac{1}{\pi} \frac{\phi^2}{\sqrt{1 - \phi/\phi_{\text{max}}}}, \quad (\text{C.27b})$$

where p_{act} is the active pressure, $\phi = \rho \pi d_{\text{hs}}^3/6$ is the volume fraction, and $\phi_{\text{max}} = 0.645$ is the maximum volume fraction of disordered hard spheres. The active pressure above is consistent with a dimensionless active speed given by:

$$\bar{U} = \left[1 + \left(1 - \exp \left[-2^{7/6} \left(\frac{\ell_0}{d_{\text{hs}}} \right) \right] \right) \frac{\phi}{1 - \phi/\phi_{\text{max}}} \right]^{-1}. \quad (\text{C.28})$$

Although these equations were empirically fit to simulations conducted in the homogeneous region of the phase diagram, we assume that they remain valid in the phase separated region.

Before outlining the self-consistent scheme to solve Eqs. (C.25) and (C.22) simultaneously, we describe the numerical construction of the density profile of the critical nucleus. Solving for this density profile is a key step of the self-consistent scheme we will later outline.

While constructing a profile, we will assume that R , ρ^{in} , and γ_{mech} are already known. Going from Eq. (C.16) to Eq. (C.18) involved integrating from $r = 0$ to $r = \infty$. Suppose we had instead integrated to some arbitrary radius between 0 and

∞ . Doing so would have resulted in:

$$\begin{aligned} \frac{1}{2} \frac{\partial \mathcal{E}}{\partial \rho} \kappa_1(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 \Big|_0^r &= \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}(r)} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{in}})] d\mathcal{E} \\ &\quad - \frac{2}{R_C} \left[\int_0^r \left(\kappa_1(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{E} \mathcal{G} \right) dr - \mathcal{E}(r) \int_0^r \mathcal{G} dr \right]. \end{aligned} \quad (\text{C.29})$$

We now convert the integrals to be over ρ instead of $\mathcal{E}$ or r :

$$\begin{aligned} \frac{1}{2} \frac{\partial \mathcal{E}}{\partial \rho} \kappa_1(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 &= \int_{\rho^{\text{in}}}^{\rho(r)} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{in}})] \frac{\partial \mathcal{E}}{\partial \rho} d\rho \\ &\quad - \frac{2}{R_C} \left[\int_{\rho^{\text{in}}}^{\rho(r)} \left(\kappa_1(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right) + \mathcal{E} \mathcal{G} \left(\frac{\partial \rho}{\partial r} \right)^{-1} \right) d\rho \right. \\ &\quad \left. - \mathcal{E}(\rho) \int_{\rho^{\text{in}}}^{\rho(r)} \mathcal{G} \left(\frac{\partial \rho}{\partial r} \right)^{-1} d\rho \right], \end{aligned} \quad (\text{C.30})$$

where we have used the radial symmetry condition $\partial \rho / \partial r = 0$ at $r = 0$. We can then isolate the density gradient by rearranging and taking the square root of both sides:

$$\begin{aligned} \frac{\partial \rho}{\partial r} &= \pm f(\rho) = \pm \left(\frac{2}{\kappa_1(\rho)} \left(\frac{\partial \mathcal{E}}{\partial \rho} \right)^{-1} \int_{\rho^{\text{in}}}^{\rho(r)} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{in}})] \frac{\partial \mathcal{E}}{\partial \rho} d\rho \right. \\ &\quad - \frac{2}{R_C} \left[\int_{\rho^{\text{in}}}^{\rho(r)} \left(\kappa_1(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right) + \mathcal{E} \mathcal{G} \left(\frac{\partial \rho}{\partial r} \right)^{-1} \right) d\rho \right. \\ &\quad \left. \left. - \mathcal{E}(\rho) \int_{\rho^{\text{in}}}^{\rho(r)} \mathcal{G} \left(\frac{\partial \rho}{\partial r} \right)^{-1} d\rho \right] \right)^{1/2}. \end{aligned} \quad (\text{C.31})$$

Equation (C.31) maps any value of density between ρ^{in} and ρ^{sat} to its derivative in the r direction. Using ρ^{in} as an initial value, this mapping between density values

and r derivatives allows for the construction of another mapping between density values and r coordinate in the density profile:

$$r(\rho) = \int_{\rho^{\text{in}}}^{\rho} \frac{1}{f(\rho')} d\rho'. \quad (\text{C.32})$$

We identify this mapping as $\varphi_R(r)$, the average density profile of the critical nucleus.

The above procedure which requires R_C , γ_{mech} , and ρ^{in} can be used with the following self-consistent scheme for simultaneous solution of Eqs. (C.25) and (C.22):

1. Begin with an initial guess for the density profile, $\rho(r)$.
2. Using $\rho(r)$, evaluate γ_{mech} from Eq. (C.9).
3. Using a given value of ρ^{sat} , $\rho(r)$, and γ_{mech} , find the optimal value of R_C and ρ^{in} that solve Eqs. (C.25) and (C.22).
4. If on the first iteration of the scheme, record the Laplace pressure difference, $2\gamma_{\text{mech}}/R_C$.
5. Solve for a new density profile, $\rho^*(r)$ using the γ_{mech} and the newly calculated R_C and ρ^{in} .
6. Using $\rho^*(r)$, update γ_{mech} from Eq. (C.9).
7. Record the updated value of the Laplace pressure difference, $2\gamma_{\text{mech}}/R_C$.
8. If the new value of the Laplace pressure is within tolerance of the old one, the procedure has converged and set $\varphi_R(r) = \rho^*(r)$. Return the current values of R_C , ρ^{in} , and the newest density profile. Otherwise, repeat the process starting from Step 1 using $\rho^*(r)$ as the updated $\rho(r)$.

The above numerical scheme was used to calculate the data plotted in Figs. 1 and 2 of the main text. The density profile corresponding to macroscopic phase separation was used as the initial guess of the above numerical scheme for the closest value of ρ^{sat} to the binodal, and the density profile obtained there was used as the initial guess for the next closest value of ρ^{sat} to the binodal, etc.

We now discuss the connection of the above coexistence theory to the recently identified non-equilibrium chemical potential. As recently proposed in Ref. [64], we define the nonequilibrium chemical potential g (equivalent to that defined by Solon *et al.* [19]) through a generalized Gibbs-Duhem relation:

$$dg = \mathcal{E}d\mathcal{P}. \quad (\text{C.33})$$

This definition ensures that g will be equal between two macroscopic coexisting phases, a coexistence criteria that is identical to the a generalized equal-area Maxwell construction on $\mathcal{P}$ with respect to $\mathcal{E}$. The product rule results in:

$$dg = d(\mathcal{P}\mathcal{E}) - \mathcal{P}d\mathcal{E}. \quad (\text{C.34})$$

We may then integrate Eq. (C.34) from the inside of the nucleus to the outside to find:

$$g^{\text{sat}} - g^{\text{in}} = \mathcal{P}^{\text{sat}}\mathcal{E}^{\text{sat}} - \mathcal{P}^{\text{in}}\mathcal{E}^{\text{in}} - \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} \mathcal{P}d\mathcal{E}. \quad (\text{C.35})$$

Substitution of Eq. (C.11) into Eq. (C.35) results in:

$$g^{\text{sat}} - g^{\text{in}} = -\frac{\mathcal{E}^{\text{in}}2\gamma_{\text{mech}}}{R_C} - \mathcal{E}^{\text{in}}\mathcal{K}\Big|_{r=0} - \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P} - \mathcal{P}^{\text{sat}}] d\mathcal{E}. \quad (\text{C.36})$$

Substitution of Eq. (C.24) into Eq. (C.36) results in:

$$g^{\text{sat}} - g^{\text{in}} = -\Delta g = -\frac{2}{R_C} \left[\mathcal{E}^{\text{in}}\gamma_{\text{mech}} + \frac{\gamma_{\text{ost}}\rho^{\text{sat}}\Delta\mathcal{E}}{\Delta\rho} \right] + \int_0^\infty (2\mathcal{E}^{\text{in}} - \mathcal{E}) \frac{\partial\mathcal{K}}{\partial r} dr. \quad (\text{C.37})$$

In general, the chemical potential within and outside the droplet will be different. However, as we demonstrate later in this Appendix, this difference vanishes for

a passive system. We may also define a pseudopotential as $\Phi \equiv \int \mathcal{P} d\mathcal{E}$ as the antiderivative of the pressure with respect to $\mathcal{E}$, allowing indefinite integration of Eq. (C.34) to result in:

$$g = \mathcal{P}\mathcal{E} - \Phi. \quad (\text{C.38})$$

Defining Φ allows us to express the critical radius from Eq. (C.24) as:

$$R_C = \frac{2\gamma_{\text{ost}}\rho^{\text{sat}}\Delta\mathcal{E}}{(-\Delta\Phi + \Delta\mathcal{E}\mathcal{P}^{\text{sat}})\Delta\rho}, \quad (\text{C.39})$$

so long as Σ can be expressed fully locally. This form for R_C will become convenient when discussing the effective nucleation barrier read off from the stochastic dynamics of nucleation.

We now turn our attention to when the terms in the stress tensor can be written fully locally. Earlier we introduced the dynamic stress tensor $\Sigma = \sigma + \mathbf{F}$, where σ is the conservative stress tensor due to interaction forces and ideal gas pressure while $\mathbf{F}$ is defined as the Coulomb integral of the body force:

$$\mathbf{F} = \frac{1}{4\pi} \int d\mathbf{r}' \mathbf{b}(\mathbf{r}') \frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3}. \quad (\text{C.40})$$

It is straightforward to show that:

$$\frac{\partial}{\partial \mathbf{r}'} \frac{1}{|\mathbf{r} - \mathbf{r}'|} = -\frac{\mathbf{r} - \mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|^3}. \quad (\text{C.41})$$

Then, following integration by parts, Eq. (C.40) becomes:

$$\mathbf{F} = \frac{1}{4\pi} \int d\mathbf{r}' \frac{\partial}{\partial \mathbf{r}'} \mathbf{b}(\mathbf{r}') \frac{1}{|\mathbf{r} - \mathbf{r}'|}. \quad (\text{C.42})$$

Note that $|\mathbf{r} - \mathbf{r}'|^{-1}$ is the Green's function of the Laplace operator in three dimensions. Therefore solving for $\mathbf{F}$ is equivalent to finding the inverse Laplacian of $\nabla \mathbf{b}$.

Under conditions of spherical symmetry this can be solved for as:

$$\begin{aligned} \mathbf{F} = & - \int_r^\infty dr_2 \frac{1}{r_2^2} \int_0^{r_2} dr_1 r_1^2 \frac{\partial}{\partial r_1} [\mathbf{b}_r] \mathbf{e}_r \mathbf{e}_r + \int_r^\infty dr_2 \frac{1}{r_2^2} \int_0^{r_2} dr_1 r_1 (\mathbf{b}_r) \mathbf{e}_\theta \mathbf{e}_\theta \\ & + \int_r^\infty dr_2 \frac{1}{r_2^2} \int_0^{r_2} dr_1 r_1 (\mathbf{b}_r) \mathbf{e}_\phi \mathbf{e}_\phi \end{aligned} \quad (\text{C.43})$$

While the divergence of $\mathbf{F}$ is equal to $\mathbf{b}$, we are not guaranteed that all components of $\mathbf{F}$ are *local*. Depending on the form of $\mathbf{b}$ one may find components of $\mathbf{F}$ that depend on integrals over the entire spatial profile, conceptually similar to the non-local chemical potential found in Ref. [39]. In order to obey the required symmetries of the flux, an expansion of $\mathbf{b}$ in gradients of the density may only contain odd gradients in the density. Any first order term will have the form $b_o(\rho) \nabla \rho$, which is trivially the divergence of $[\int b_o(\rho) d\rho] \mathbf{I}$. Thus first order terms in $\mathbf{b}$ will always result in bulk isotropic terms of $\mathbf{F}$, which can be expressed locally. However, the third order terms in $\mathbf{b}$ will result in second order terms of $\mathbf{F}$ that may or may not have non-local contributions. The exact form of these stress components in spherical coordinates may be computed by substituting the third order terms of an expansion of $\mathbf{b}$ into Eq. (C.43). For a spatially isotropic system, the third order gradient expansion of $\mathbf{b}$ is:

$$\mathbf{b} = \beta_0(\rho) \nabla \rho + \beta_1(\rho) \nabla \rho \nabla^2 \rho + \beta_2(\rho) \nabla |\nabla \rho|^2 + \beta_3(\rho) \nabla \nabla^2 \rho + \beta_4(\rho) |\nabla \rho|^2 \nabla \rho \quad (\text{C.44})$$

When the density field and the forces are spherically symmetric, i.e. $\nabla \rho$ is parallel to $\mathbf{e}_r$, $\mathbf{F}$ can be expressed as:

$$\begin{aligned} \mathbf{F} = & \left[\int \beta_0(\rho) d\rho + \beta_3(\rho) \nabla^2 \rho + \left(\frac{1}{2} \frac{\partial \beta_3}{\partial \rho} + \beta_2(\rho) - \frac{1}{2} \beta_1(\rho) \right) |\nabla \rho|^2 + \mathcal{K}(r) \right] \mathbf{I} \\ & + \left(\beta_1(\rho) - \frac{\partial \beta_3}{\partial \rho} \right) \nabla \rho \nabla \rho, \end{aligned} \quad (\text{C.45})$$

where $\mathcal{K}$ is a non-local term defined as:

$$\mathcal{K} = \int_r^\infty \left[-\beta_4(\rho) + \frac{\partial \beta_2}{\partial \rho} + \frac{1}{2} \frac{\partial \beta_1}{\partial \rho} - \frac{1}{2} \frac{\partial^2 \beta_3}{\partial \rho^2} \right] \left(\frac{\partial \rho}{\partial r'} \right)^3 dr'. \quad (\text{C.46})$$

By taking the divergence of $\mathbf{F}$ one can verify that $\mathbf{b}$ is recovered. We note that $\mathbf{F}$ is not unique; it may be altered by divergence-free terms.

In certain cases, such as ABPs, the body force can naturally be found as the divergence of a stress tensor, and in these cases no non-local terms appear in $\mathbf{F}$ [21, 29]. The condition for $\mathbf{F}$ to be fully local is:

$$\beta_4(\rho) = \frac{\partial \beta_2}{\partial \rho} + \frac{1}{2} \frac{\partial \beta_1}{\partial \rho} - \frac{1}{2} \frac{\partial^2 \beta_3}{\partial \rho^2}. \quad (\text{C.47})$$

C.2 Connection to Active Model B+

With the development of our mechanical perspective, we can now revisit the work of Tjhung *et al.* [39] which derive the criteria for the critical nucleus for model AMB+ (see Eqs. 16-22 in Ref. [39]) and determine if there is a mechanical interpretation for their criteria. As frequently noted by the authors of Ref. [39], while AMB+ is a general phenomenological description of active systems, it remains unclear if the quantities that emerge from this perspective, such as the pseudotension and pseudopressure, are connected to the actual mechanical tension and pressure, respectively. Here, we will formally map our criteria to that of Ref. [39]. In doing so, we demonstrate that the mapping is not unique, and therefore the interpretation of pseudotension and pseudopressure are indeed arbitrary.

We make a few modifications to the version of model AMB+ presented in Ref. [39] to facilitate comparison to our work. These include the consideration of a nonconstant mobility (the mobility is often presumed constant to reduce the stochastic contribution of the flux from multiplicative to additive) and we will not specify a particular form (e.g., the chemical potential stemming from the canonical quartic free energy) of the “bulk” AMB+ chemical potential, μ_o below. We also replace the linearly scaled (by the critical density) density (ϕ in Ref. [39]) with the physical number density, ρ . Finally, as we consider nucleation far from the critical point, we did not limit our perspective to small density departures from the critical density. As a result, our flux expression is $O(\nabla^3)$ while that of AMB+ (as presented in Ref. [39]) is $O(\nabla^3 \rho^2)$. The restriction to quadratic density in this

form of AMB+ leads to the absence of some terms allowable by symmetry but higher order in density, such as those considered in Ref. [47]. We nevertheless consider the version of AMB+ without the additional terms in order to recover the criteria obtained in Ref. [39].

We begin with a summary of the derivation of finite-size coexistence criteria presented in Ref. [39]. We adopt all notation used in Ref. [39], except we replace the AMB+ parameter ζ with $\mathcal{Z}$ in order to avoid confusion with the microscopic drag and replace the density derivative of the AMB+ effective free energy $f'(\rho)$ with μ_o for additional clarity.

The deterministic component of the flux in AMB+ is given by:

$$\begin{aligned} \mathbf{J}/M[\rho] = & -\frac{\partial\mu_o}{\partial\rho}\nabla\rho + K(\rho)\nabla\nabla^2\rho + \frac{\partial K}{\partial\rho}\nabla\rho\nabla^2\rho + \frac{1}{2}\frac{\partial K}{\partial\rho}\nabla|\nabla\rho|^2 \\ & + \frac{1}{2}\frac{\partial^2 K(\rho)}{\partial\rho^2}\nabla\rho|\nabla\rho|^2 - \lambda(\rho)\nabla|\nabla\rho|^2 - \frac{\partial\lambda}{\partial\rho}\nabla\rho|\nabla\rho|^2 + \mathcal{Z}(\rho)\nabla\rho\nabla^2\rho. \end{aligned} \quad (\text{C.48})$$

For a spherically symmetric system, the radial flux can be rewritten as:

$$\frac{\mathbf{J} \cdot \mathbf{e}_r}{M[\rho]} = -\frac{\partial\mu}{\partial r}, \quad (\text{C.49})$$

where the effective chemical potential is given by:

$$\begin{aligned} \mu = \mu_o - K(\rho)\frac{\partial^2\rho}{\partial r^2} - \frac{2}{r}K(\rho)\frac{\partial\rho}{\partial r} + \left(\lambda - \frac{1}{2}\frac{\partial K}{\partial\rho} - \frac{1}{2}\mathcal{Z}(\rho)\right)\left(\frac{\partial\rho}{\partial r}\right)^2 \\ - \frac{1}{2}\int_r^\infty \frac{\partial\mathcal{Z}}{\partial\rho}\left(\frac{\partial\rho}{\partial r'}\right)^3 dr' + \int_r^\infty \frac{2\mathcal{Z}}{r'}\left(\frac{\partial\rho}{\partial r'}\right)^2 dr', \end{aligned} \quad (\text{C.50})$$

Again, a key difference between this expression and that found in Ref. [39] is that we are allowing for density dependence on all coefficients and retaining terms in the flux at the order of ∇^3 . The finite size coexistence criteria can be found by setting the radial flux to zero. This implies that μ is a constant, which means one

coexistence criteria can be immediately read as:

$$\mu_0(\rho^{\text{sat}}) = \mu_0(\rho^{\text{in}}) - \frac{1}{2} \int_r^\infty \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r'} \right)^3 dr' + \frac{2}{R_C} \int_r^\infty \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r'} \right)^2 dr'. \quad (\text{C.51})$$

Therefore the chemical potential is constant everywhere, with a jump in the bulk component of the chemical potential occurring at the interface. The second coexistence criteria can be found by introducing the pseudovvariable which satisfies the following differential equation:

$$\frac{\partial^2 \psi}{\partial \rho^2} = \frac{\mathcal{Z} - 2\lambda}{K} \frac{\partial \psi}{\partial \rho}. \quad (\text{C.52})$$

We now multiply Eq. (C.50) by $\partial_r \psi$ and integrate across all space to find:

$$\begin{aligned} \mu_o(\rho^{\text{sat}}) (\psi^{\text{sat}} - \psi^{\text{in}}) - \int_0^\infty \mu_o(\rho) \frac{\partial \psi}{\partial r} dr &= - \int_0^\infty \frac{2}{r} K(\rho) \frac{\partial \psi}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 \\ &- \frac{1}{2} \int_0^\infty \frac{\partial \psi}{\partial r} \int_r^\infty \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r'} \right)^3 dr dr' + 2 \int_0^\infty \frac{\partial \psi}{\partial r} \int_r^\infty \frac{\mathcal{Z}(\rho)}{r'} \left(\frac{\partial \rho}{\partial r'} \right)^2 dr' dr. \end{aligned} \quad (\text{C.53})$$

Integration by parts and algebraic manipulation of Eq. (C.53) results in:

$$\int_{\psi^{\text{in}}}^{\psi^{\text{sat}}} [\mu_0 - \mu_0^{\text{sat}}] d\psi + \frac{1}{2} \int_0^\infty (\psi^{\text{in}} - \psi) \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r'} \right)^3 dr = \frac{2\sigma}{R_C}, \quad (\text{C.54})$$

where the pseudotension σ is defined as:

$$\begin{aligned} \sigma &= \int_0^\infty K(\rho) \frac{\partial \psi}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 dr \\ &- \int_0^\infty \mathcal{Z}(\rho) \psi(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr + \psi^{\text{in}} \int_0^\infty \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr. \end{aligned} \quad (\text{C.55})$$

Together, Eqs. (C.51) and (C.54) comprise the finite-size coexistence criteria for AMB+. The *mechanical* finite-size coexistence criteria derived in this work were found by setting the radial component of the divergence of the dynamic stress tensor to zero. No constitutive model for flux was specifically invoked. However, we can map AMB+ to an equivalent mechanical model in which we assume that the flux is proportional to a body force, i.e. $\mathbf{J} \propto \mathbf{b}$. We can expand this body force $\mathbf{b}$ using Eq. (C.44), which then allows us to map the expansion coefficients $\{\beta_i\}$ to AMB+ coefficients as:

$$\beta_0(\rho) = -M[\rho] \frac{\partial \mu_o}{\partial \rho}, \quad (\text{C.56a})$$

$$\beta_1(\rho) = M[\rho] \mathcal{Z}(\rho) + M[\rho] \frac{\partial K}{\partial \rho}, \quad (\text{C.56b})$$

$$\beta_2(\rho) = -M[\rho] \lambda(\rho) + \frac{1}{2} M[\rho] \frac{\partial K}{\partial \rho}, \quad (\text{C.56c})$$

$$\beta_3(\rho) = M[\rho] K(\rho), \quad (\text{C.56d})$$

$$\beta_4(\rho) = -M[\rho] \frac{\partial \lambda}{\partial \rho} + \frac{1}{2} M[\rho] \frac{\partial^2 K}{\partial \rho^2}. \quad (\text{C.56e})$$

One can then use Eq. (C.45) to identify the stress expansion coefficients $\{\kappa_i\}$:

$$\mathcal{P} = \int M[\rho] \frac{\partial \mu_o}{\partial \rho} d\rho, \quad (\text{C.57a})$$

$$\kappa_1(\rho) = M[\rho] K(\rho), \quad (\text{C.57b})$$

$$\kappa_2(\rho) = -M[\rho] \lambda(\rho) + \frac{1}{2} M[\rho] \frac{\partial K}{\partial \rho} + \frac{1}{2} \frac{\partial M}{\partial \rho} K(\rho) - \frac{1}{2} M[\rho] \mathcal{Z}(\rho), \quad (\text{C.57c})$$

$$\kappa_3(\rho) = M[\rho] \mathcal{Z}(\rho) - K(\rho) \frac{\partial M}{\partial \rho}, \quad (\text{C.57d})$$

$$\kappa_4(\rho) = 0, \quad (\text{C.57e})$$

$$\mathcal{K} = \frac{1}{2} \int_r^\infty \left[-\frac{\partial^2 M}{\partial \rho^2} - \frac{2\lambda(\rho) - \mathcal{Z}(\rho) + \frac{\partial K}{\partial \rho}}{K(\rho)} \frac{\partial M}{\partial \rho} + \frac{\frac{\partial \mathcal{Z}}{\partial \rho}}{K(\rho)} M[\rho] \right] K(\rho) \left(\frac{\partial \rho}{\partial r} \right)^3 dr. \quad (\text{C.57f})$$

We can now appreciate that $M[\rho]$ is a degree of freedom in this mapping, and depending on the choice of $M[\rho]$ the physical interpretation of quantities such as μ_o , ψ , and σ may change. If one insists that Σ be written fully locally, one can solve for a mobility $M[\rho]$ that satisfies this demand by solving the differential equation:

$$-\frac{\partial^2 M}{\partial \rho^2} - \frac{2\lambda(\rho) - \mathcal{Z}(\rho) + \frac{\partial K}{\partial \rho}}{K(\rho)} \frac{\partial M}{\partial \rho} + \frac{\frac{\partial \mathcal{Z}}{\partial \rho}}{K(\rho)} M[\rho] = 0. \quad (\text{C.58})$$

Solving this equation is difficult without further information regarding K , λ , and ζ . In the remainder of this Section, we will discuss the physical interpretations of AMB+ quantities (e.g. chemical potential, pseudotension) and coexistence criteria for particular choices of $M[\rho]$.

One choice for $M[\rho]$ is to set as proportional to some constant, i.e. $M[\rho] \sim 1$. For this choice, one can use Eqs. (C.57) to find:

$$\mathcal{P} \sim \mu_0, \quad (\text{C.59a})$$

$$\kappa_1(\rho) \sim K(\rho), \quad (\text{C.59b})$$

$$\kappa_2(\rho) \sim -\lambda(\rho) + \frac{1}{2} \frac{\partial K}{\partial \rho} - \frac{1}{2} \mathcal{Z}(\rho), \quad (\text{C.59c})$$

$$\kappa_3(\rho) \sim \mathcal{Z}(\rho), \quad (\text{C.59d})$$

$$\kappa_4(\rho) = 0, \quad (\text{C.59e})$$

$$\mathcal{K} \sim \frac{1}{2} \int_r^\infty \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr. \quad (\text{C.59f})$$

We can now see that when $M[\rho] \sim 1$, the dynamic pressure is analogous to the AMB+ chemical potential. We now ask how to relate $\mathcal{E}$ with ψ . For convenience, we restate the definition of $\mathcal{E}$ given by Eq. (C.17):

$$\frac{\partial^2 \mathcal{E}}{\partial \rho^2} = \frac{2(\kappa_2(\rho) + \kappa_3(\rho)) - \frac{\partial}{\partial \rho} (\kappa_1(\rho) + \kappa_4(\rho))}{\kappa_1(\rho) + \kappa_4(\rho)} \frac{\partial \mathcal{E}}{\partial \rho}. \quad (\text{C.60})$$

Substitution of Eqs. (C.59) into Eq. (C.17) results in:

$$\frac{\partial^2 \mathcal{E}}{\partial \rho^2} = -\frac{2\lambda - \mathcal{Z}}{K} \frac{\partial \mathcal{E}}{\partial \rho}. \quad (\text{C.61})$$

This result, combined with Eq. (C.52), implies that $\psi \sim \mathcal{E}$ when $M[\rho] \sim 1$. We can now compare the mechanically derived coexistence criteria to that of AMB+. The Laplace pressure difference found in this work is:

$$\mathcal{P}^{\text{in}} - \mathcal{P}^{\text{sat}} = \mathcal{K} \Big|_{r=0} + \frac{2\gamma_{\text{mech}}}{R_C}. \quad (\text{C.62})$$

The mechanical surface tension under a $M[\rho] \sim 1$ mapping is equal to:

$$\gamma_{\text{mech}} = \int_0^\infty \left[-\kappa_3(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 - \kappa_4(\rho) \frac{\partial^2 \rho}{\partial r^2} \right] dr = - \int_0^\infty \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr. \quad (\text{C.63})$$

Therefore our mechanically derived Laplace pressure difference is:

$$\mathcal{P}^{\text{in}} - \mathcal{P}^{\text{sat}} = \frac{1}{2} \int_0^r \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr - \frac{2}{R_C} \int_0^\infty \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr, \quad (\text{C.64})$$

which is identical to Eq. (C.51) because $\mathcal{P} \sim \mu_0$. The second coexistence criteria found by this work is given by:

$$\begin{aligned} \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P} - \mathcal{P}^{\text{sat}}] d\mathcal{E} + \int_0^\infty (\mathcal{E}^{\text{in}} - \mathcal{E}^{\text{out}}) \left(\frac{\partial \mathcal{K}}{\partial r} \right) dr \\ = \frac{2}{R_C} \int_0^\infty \left(\kappa_1(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{G}(\mathcal{E} - \mathcal{E}^{\text{in}}) \right) dr. \end{aligned} \quad (\text{C.65})$$

Substitution of the relations to AMB+ coefficients in our constant mobility mapping results in:

$$\begin{aligned} \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P} - \mathcal{P}^{\text{sat}}] d\mathcal{E} - \mathcal{E}^{\text{in}} \frac{1}{2} \int_0^\infty \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr + \frac{1}{2} \int_0^\infty \mathcal{E} \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr \\ = \frac{2}{R_C} \int_0^\infty \left(K(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{E}^{\text{in}} \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 - \mathcal{E} \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 \right) dr. \end{aligned} \quad (\text{C.66})$$

Noting that under this mapping $\mathcal{P} \sim \mu_0$ and $\psi \sim \mathcal{E}$, we can see that the right hand side of the above equation reduces to $2\sigma/R_C$ and the second AMB+ coexistence criteria Eq. (C.54) is recovered.

Another usefeul choice for $M[\rho]$ is to set it to be proportional to the inverse of $\mathcal{E}$, i.e. $M[\rho] \sim 1/\mathcal{E}$. For this choice, one can use Eqs. (C.57) to find:

$$\mathcal{P} = \int \frac{1}{\mathcal{E}} \frac{\partial \mu_o}{\partial \rho} d\rho, \quad (\text{C.67a})$$

$$\kappa_1(\rho) = \frac{1}{\mathcal{E}} K(\rho), \quad (\text{C.67b})$$

$$\kappa_2(\rho) = -\frac{1}{\mathcal{E}} \lambda(\rho) + \frac{1}{2} \frac{1}{\mathcal{E}} \frac{\partial K}{\partial \rho} - \frac{1}{2} \frac{1}{\mathcal{E}^2} \frac{\partial \mathcal{E}}{\partial \rho} K(\rho) - \frac{1}{2} \frac{1}{\mathcal{E}} \mathcal{Z}(\rho), \quad (\text{C.67c})$$

$$\kappa_3(\rho) = \frac{1}{\mathcal{E}} \mathcal{Z}(\rho) + \frac{1}{\mathcal{E}^2} \frac{\partial \mathcal{E}}{\partial \rho} K(\rho), \quad (\text{C.67d})$$

$$\kappa_4(\rho) = 0, \quad (\text{C.67e})$$

$$\mathcal{K} \sim \frac{1}{2} \int_r^\infty \frac{1}{\mathcal{E}} \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr. \quad (\text{C.67f})$$

From Eqs. (C.67), one can immediately see that $\mathcal{E} d\mathcal{P} = d\mu_0$, which implies that the effective chemical potential g identified by this work is proportional to the bulk AMB+ chemical potential μ_0 when $M[\rho] \sim 1/\mathcal{E}$. Further, one can substitute the mapping Eqs. (C.67) into the pseudovariabe definitions Eq. (C.17) and Eq. (C.52) to find that $\mathcal{E} \sim 1/\psi$. We now consider the coexistence criteria under this $M[\rho] \sim 1/\mathcal{E}$ mapping. We start with the mechanical Maxwell construction:

$$\begin{aligned} \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P} - \mathcal{P}^{\text{sat}}] d\mathcal{E} = & -\frac{1}{2} \int_0^\infty \left(\mathcal{E}^{\text{in}} - \mathcal{E} \right) \frac{1}{\mathcal{E}} \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr \\ & + \frac{2}{R_C} \int_0^\infty \left(\kappa_1(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{G} \left(\mathcal{E} - \mathcal{E}^{\text{in}} \right) \right) dr. \end{aligned} \quad (\text{C.68})$$

Substitution of Eq. (C.36) results in:

$$\begin{aligned}
g^{\text{in}} - g^{\text{out}} + \frac{2}{R_C} \mathcal{E}^{\text{in}} \int_0^\infty \left[\frac{1}{\mathcal{E}} \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 + K(\rho) \frac{1}{\mathcal{E}^2} \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 \right] dr \\
- \frac{1}{2} \mathcal{E}^{\text{in}} \int_0^\infty \frac{1}{\mathcal{E}} \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr = -\frac{1}{2} \int_0^\infty \left(\mathcal{E}^{\text{in}} - \mathcal{E} \right) \frac{1}{\mathcal{E}} \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr \\
+ \frac{2}{R_C} \int_0^\infty \left(\kappa_1(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^2 + \mathcal{G}(\mathcal{E} - \mathcal{E}^{\text{in}}) \right) dr. \quad (\text{C.69})
\end{aligned}$$

Substitution of Eqs. (C.67) results in:

$$g^{\text{in}} - g^{\text{out}} = \frac{1}{2} \int_0^\infty \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr - \frac{2}{R_C} \int_0^\infty \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr \quad (\text{C.70})$$

Therefore, the AMB+ coexistence criteria Eq. (C.51) is recovered (up to a $(\partial_r \rho)^3$ contribution). We now consider the mechanically derived Laplace pressure difference, with Eqs. (C.67) substituted:

$$\begin{aligned}
\mathcal{P}^{\text{in}} - \mathcal{P}^{\text{sat}} = \frac{1}{2} \int_0^\infty \frac{1}{\mathcal{E}} \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr \\
- \frac{2}{R_C} \int_0^\infty \left[\frac{1}{\mathcal{E}} \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 + \frac{1}{\mathcal{E}^2} \frac{\partial \mathcal{E}}{\partial \rho} K(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 \right] dr. \quad (\text{C.71})
\end{aligned}$$

Recall the generalized Gibbs-Duhem relation $dg = \mathcal{E}d\mathcal{P}$. Under the choice $M[\rho] \sim 1/\mathcal{E}$, we know that $\psi \sim 1/\mathcal{E}$ and $g \sim \mu_0$, which implies that $\psi d\mu_o = d\mathcal{P} = d(\mu_o \psi) - \mu_o d\psi$. Therefore:

$$\mathcal{P}^{\text{in}} - \mathcal{P}^{\text{sat}} = \mu_0^{\text{in}} \psi^{\text{in}} - \mu_0^{\text{sat}} \psi^{\text{sat}} + \int_{\psi^{\text{in}}}^{\psi^{\text{sat}}} \mu_0 d\psi. \quad (\text{C.72})$$

From Eq. (C.51) this can be rewritten as:

$$\begin{aligned} \mathcal{P}^{\text{in}} - \mathcal{P}^{\text{sat}} = & \frac{1}{2} \psi^{\text{in}} \int_0^\infty \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr \\ & - \psi^{\text{in}} \frac{2}{R_C} \int_0^\infty \mathcal{Z}(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr + \int_{\psi^{\text{in}}}^{\psi^{\text{sat}}} [\mu_0 - \mu_o^{\text{sat}}] d\psi. \end{aligned} \quad (\text{C.73})$$

We can then substitute Eq. (C.73) into the Laplace pressure difference to find that:

$$\int_{\psi^{\text{in}}}^{\psi^{\text{sat}}} [\mu_0 - \mu_o^{\text{sat}}] d\psi + \frac{1}{2} \int_0^\infty (\psi^{\text{in}} - \psi) \frac{\partial \mathcal{Z}}{\partial \rho} \left(\frac{\partial \rho}{\partial r} \right)^3 dr = \frac{2\sigma}{R_C}, \quad (\text{C.74})$$

which recovers the second coexistence criteria Eq. (C.54).

We emphasize that choosing $M[\rho]$ to be constant or proportional to $1/\mathcal{E}$ facilitates a clear physical interpretation for AMB+, but will in general lead to nonlocal terms. Choosing $M[\rho]$ such that the mapping is fully local may lead to difficulty in physically interpreting AMB+ quantities. For example, consider the values of $\{\kappa_i\}$ found for ABPs in Eqs. (C.26). Substitution of these expressions into Eqs. (C.57) results in constraining $\mathcal{K} = 0$, which then leads to a solution for $M[\rho]$ as:

$$M[\rho] \sim \exp \left[- \int \frac{\left[\frac{\partial}{\partial \rho} \left(\bar{U} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \right) \right]}{\bar{U}^2 \frac{\partial^2 p_C}{\partial \rho^2}} d\rho \right]. \quad (\text{C.75})$$

This solution for $M[\rho]$ leads to us identifying μ_0 as:

$$\mu_0 \sim \int \exp \left[\int \frac{\left[\frac{\partial}{\partial \rho} \left(\bar{U} \frac{\partial}{\partial \rho} \left[\bar{U} \frac{\partial p_C}{\partial \rho} \right] \right) \right]}{\bar{U}^2 \frac{\partial^2 p_C}{\partial \rho^2}} d\rho \right] d\mathcal{P}, \quad (\text{C.76})$$

which has a less clear physical interpretation than when $M[\rho]$ is constant or proportional to $1/\mathcal{E}$. This implies that, despite the generic mapping between mechanics and AMB+ presented here, directly comparing quantities such as pressure, chemical potential, or surface tension within our model of ABP transport to AMB+ is non-trivial.

C.3 Dynamics of an Active Nucleus

We aim to derive the Langevin equation for the radius of a nucleus of active Brownian particles in the spherically symmetric geometry described in the previous section. We assume the dynamics of the nucleus to proceed quasistatically: any given nucleus with radius R (not necessarily equal to R_C) will be in a local mechanical balance with its surroundings. This implies that the density immediately outside of the nucleus $\rho^{\text{out}} \neq \rho^{\text{sat}}$, with diffusive transport in the majority phase driven by this inequality. Then, as diffusion will cause ρ^{out} to change in time, the local mechanical balance will demand R to change in time as well. The starting point to derive the dynamics described above is the fluctuating hydrodynamics which we presented in Appendix A.1. Substitution of Eqs. (A.34e) and Eq. (A.34b) into Eq. (A.34a) results in:

$$\zeta \frac{\partial \rho}{\partial t} = -\nabla^2 \Sigma_{rr} + \frac{2}{r} \frac{\partial \mathcal{G}}{\partial r} + \frac{2}{r^2} \mathcal{G} - \zeta \nabla \cdot \boldsymbol{\eta}^{\text{act}}. \quad (\text{C.77})$$

From here, we proceed in a similar fashion as Cates and Nardini [78]. We recently established a surface-area minimizing principle for liquid-gas interfaces of ABPs resulting from a positive capillary-wave tension, γ_{cw} , which resists fluctuations to the area-minimizing interfacial shape. The strength of the fluctuations are set by $k_B T^{\text{act}}$ [42], while the ratio of the capillary tension to $k_B T^{\text{act}}$ – the active interfacial stiffness – controls the amplitude of long-wavelength interfacial fluctuations. A fluctuating nucleus can be well-approximated as a sphere when the average radius R satisfies $(R^2 \gamma_{\text{cw}} / k_B T^{\text{act}})^{1/2} \gg 1$. Therefore we introduce the radially symmetric ansatz connecting the density field and the radius of a nucleus R at time t , which was proposed by Bray [97]:

$$\rho = \varphi_R [r - R(t)], \quad (\text{C.78})$$

where φ_R is the density profile calculated in the previous section. Substitution of Eq. (C.78) into Eq. (C.77) results in:

$$-\zeta \varphi'_R \frac{\partial R}{\partial t} = -\nabla^2 \Sigma_{rr} + \frac{2}{r} \frac{\partial \mathcal{G}}{\partial r} + \frac{2}{r^2} \mathcal{G} - \zeta \nabla \cdot \boldsymbol{\eta}^{\text{act}}, \quad (\text{C.79})$$

where φ'_R is the r derivative of φ_R . We now define the inverse Laplace operator as:

$$\nabla^{-2}f(r) = - \int_r^\infty \int_0^{r_2} dr_2 dr_1 \left(\frac{r_1}{r_2} \right)^2 f(r_1), \quad (\text{C.80})$$

where $f(r)$ is a function that vanishes in the limit of $r \rightarrow \infty$. We apply the inverse Laplace operator to both sides of Eq. (C.79). The effect of the inverse Laplace operator on the second term on the left hand side is:

$$\nabla^{-2} \left[\frac{2}{r} \frac{\partial \mathcal{G}}{\partial r} \right] = - \int_r^\infty dr_2 \frac{1}{r_2^2} \int_0^{r_2} dr_1 2r_1 \frac{\partial \mathcal{G}}{\partial r_1}. \quad (\text{C.81})$$

We now integrate Eq. (C.81) by parts while choosing:

$$\begin{aligned} u &= \int_0^{r_2} dr_1 2r_1 \frac{\partial \mathcal{G}}{\partial r_1}, & du &= 2r_2 \frac{\partial \mathcal{G}}{\partial r_2} dr_2, \\ dv &= -\frac{1}{r_2^2} dr_2, & v &= \frac{1}{r_2}. \end{aligned}$$

Then Eq. (C.81) can equivalently be expressed as

$$\nabla^{-2} \left[\frac{2}{r} \frac{\partial \mathcal{G}}{\partial r} \right] = -\frac{2}{r} \int_0^r dr_1 r_1 \frac{\partial \mathcal{G}}{\partial r_1} + 2\mathcal{G}(r). \quad (\text{C.82})$$

A second integration by parts results in:

$$\begin{aligned} \nabla^{-2} \left[\frac{2}{r} \frac{\partial \mathcal{G}}{\partial r} \right] &= -\frac{2}{r} \left(r\mathcal{G}(r) - \int_0^r \mathcal{G} dr_1 \right) + 2\mathcal{G}(r), \\ &= \frac{2}{r} \int_0^r \mathcal{G} dr_1. \end{aligned} \quad (\text{C.83})$$

Applying a similar procedure to the other terms in Eq. (C.79) results in:

$$\nabla^{-2} \left[\frac{2}{r^2} \mathcal{G} \right] = -\frac{2}{r} \int_0^r \mathcal{G} dr_1 - 2 \int_r^\infty \frac{\mathcal{G}}{r_1} dr_1, \quad (\text{C.84})$$

$$\nabla^{-2} \left[-\zeta \phi'_R \frac{\partial R}{\partial t} \right] = \zeta R \frac{\partial R}{\partial t} (\rho^{\text{sat}} - \rho^{\text{in}}) + O\left(\frac{1}{R}\right), \quad (\text{C.85})$$

$$\nabla^{-2} [-\nabla^2 \Sigma_{rr}] = -\Sigma_{rr} - \mathcal{P}(\rho^{\text{sat}}), \quad (\text{C.86})$$

where the $\mathcal{P}(\rho^{\text{sat}})$ in Eq. (C.86) comes from the boundary term of Σ_{rr} as $r \rightarrow \infty$. From the above equations one can see that applying the inverse Laplace operator to both sides of Eq. (C.79) results in:

$$\zeta R \frac{\partial R}{\partial t} \Delta \rho + O\left(\frac{1}{R}\right) = \Sigma_{rr} + \mathcal{P}(\rho^{\text{sat}}) + \int_r^\infty dr_1 \frac{2}{r_1} \mathcal{G} - \nabla^{-2} [\zeta \nabla \cdot \boldsymbol{\eta}^{\text{act}}], \quad (\text{C.87})$$

where we have defined the difference between an arbitrary property a at the interior and exterior of the bubble/droplet as $\Delta a \equiv a^{\text{in}} - a^{\text{sat}}$. We now multiply both sides of the equation by $\partial \mathcal{E} / \partial r$ and integrate from $r = 0$ to $r = \infty$:

$$\begin{aligned} -\zeta R \frac{\partial R}{\partial t} \Delta \rho \Delta \mathcal{E} + O\left(\frac{1}{R}\right) &= \int_0^\infty \Sigma_{rr} \frac{\partial \mathcal{E}}{\partial r} dr + \mathcal{P}(\rho^{\text{sat}}) (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}}) \\ &+ \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \int_r^\infty dr_1 \frac{2\mathcal{G}}{r_1} - \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \nabla^{-2} [\zeta \nabla \cdot \boldsymbol{\eta}^{\text{act}}]. \end{aligned} \quad (\text{C.88})$$

The double integral can be integrating by parts with:

$$\begin{aligned} u &= \int_r^\infty \frac{2\mathcal{G}}{r_1} dr_1, \quad du = -\frac{2\mathcal{G}}{r} dr, \\ dv &= \frac{\partial \mathcal{E}}{\partial r} dr, \quad v = \mathcal{E} \end{aligned}$$

$$\begin{aligned} -\zeta R \frac{\partial R}{\partial t} \Delta \rho \Delta \mathcal{E} + O\left(\frac{1}{R}\right) &= \int_0^\infty \Sigma_{rr} \frac{\partial \mathcal{E}}{\partial r} dr + \mathcal{P}(\rho^{\text{sat}}) (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}}) - \frac{2\gamma_{\text{mech}}}{R} \mathcal{E}^{\text{in}} \\ &+ \frac{2}{R} \int_0^\infty \mathcal{E} \mathcal{G} dr - \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \nabla^{-2} [\zeta \nabla \cdot \boldsymbol{\eta}^{\text{act}}]. \end{aligned} \quad (\text{C.89})$$

Now consider the first integral on the right hand side. Substituting in the constitutive expression for Σ_{rr} from Eq. (C.6) and integrating by parts results in

$$\int_0^\infty \Sigma_{rr} \frac{\partial \mathcal{E}}{\partial r} dr = - \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} d\mathcal{E} \mathcal{P}(\rho) + \int_0^\infty \frac{2}{r} a(\rho) \frac{\partial \mathcal{E}}{\partial \rho} (\phi'_R)^2. \quad (\text{C.90})$$

Substitution of Eq. (C.90) into Eq. (C.89) gives:

$$\begin{aligned} -\zeta R \frac{\partial R}{\partial t} \Delta \rho \Delta \mathcal{E} + O\left(\frac{1}{R}\right) &= - \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} d\mathcal{E} \mathcal{P}(\rho) + \mathcal{P}(\rho^{\text{sat}}) (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}}) - \frac{2\gamma_{\text{mech}}}{R} \mathcal{E}^{\text{in}} \\ &\quad + \frac{2}{R} \int_0^\infty \left(\mathcal{E} \mathcal{G} + a(\rho) \frac{\partial \mathcal{E}}{\partial \rho} \left(\frac{\partial \phi_R}{\partial r} \right)^2 \right) dr \\ &\quad - \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \nabla^{-2} [\zeta \nabla \cdot \eta^{\text{act}}]. \end{aligned} \quad (\text{C.91})$$

From Eq. (C.23) we can rewrite Eq. (C.91) as:

$$\begin{aligned} -\zeta R \frac{\partial R}{\partial t} \Delta \rho \Delta \mathcal{E} + O\left(\frac{1}{R}\right) &= - \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} d\mathcal{E} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] + \frac{2\gamma_{\text{ost}} \rho^{\text{sat}} \Delta \mathcal{E}}{R \Delta \rho} \\ &\quad - \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \nabla^{-2} [\zeta \nabla \cdot \eta^{\text{act}}]. \end{aligned} \quad (\text{C.92})$$

Recognizing that the first integral on the right-hand-side of Eq. (C.92) is a constant, we can rearrange:

$$\zeta R \frac{\partial R}{\partial t} \Delta \rho \Delta \mathcal{E} + O\left(\frac{1}{R}\right) = \frac{2\gamma_{\text{ost}} \rho^{\text{sat}} \Delta \mathcal{E}}{\Delta \rho} \left[\frac{1}{R_C} - \frac{1}{R} \right] + \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \nabla^{-2} [\zeta \nabla \cdot \eta^{\text{act}}], \quad (\text{C.93})$$

where the critical radius is defined as:

$$R_C = \frac{2\gamma_{\text{ost}} \rho^{\text{sat}} \Delta \mathcal{E}}{\Delta \rho \int_{\mathcal{E}^{\text{in}}}^{\mathcal{E}^{\text{sat}}} [\mathcal{P}(\rho) - \mathcal{P}(\rho^{\text{sat}})] d\mathcal{E}}, \quad (\text{C.94})$$

which is identical to the critical radius found in the previous section [see Eq. (C.24)], highlighting the consistency of the two approaches.

The final evolution equation for the radial growth can be compactly written as:

$$\zeta_{\text{eff}} \frac{\partial R}{\partial t} = F^D(R) + \tilde{\chi}(t), \quad (\text{C.95})$$

where we have defined an effective drag force:

$$\zeta_{\text{eff}} = \frac{4\pi (\rho^{\text{sat}} - \rho^{\text{in}})^2 R^3 \zeta}{\rho^{\text{sat}}}, \quad (\text{C.96})$$

as well as a deterministic force:

$$F^D(R) = 8\pi R^2 \gamma_{\text{ost}} \left[\frac{1}{R_C} - \frac{1}{R} \right]. \quad (\text{C.97})$$

$\tilde{\chi}(t)$ represents the stochastic force with:

$$\tilde{\chi}(t) = \frac{\zeta_{\text{eff}}}{R \Delta \rho \Delta \mathcal{E}} \nabla^{-2} [\nabla \cdot \boldsymbol{\eta}^{\text{act}}]. \quad (\text{C.98})$$

We next derive the statistics of $\tilde{\chi}(t)$ finding that it has zero mean and a variance given by:

$$\langle \tilde{\chi}(t) \tilde{\chi}(t') \rangle = 2k_B T^{\text{act}} \zeta_{\text{eff}} \delta(t - t'). \quad (\text{C.99})$$

From Eqs. (C.95) and (C.99) we can see that the dynamics of the nucleus obey the fluctuation dissipation theorem. In addition, the deterministic term can be integrated to define an effective potential governing nucleation as further discussed later in this section. We now seek to derive the statistics of $\tilde{\chi}(t)$ [Eq. (C.99)]. We start by rewriting the statistics of $\boldsymbol{\eta}^{\text{act}}$ to be compatible with our spherical symmetry conditions:

$$\langle \boldsymbol{\eta}^{\text{act}}(\mathbf{r}, t) \boldsymbol{\eta}^{\text{act}}(\mathbf{r}', t') \rangle = 2 \frac{k_B T^{\text{act}}}{4\pi \zeta} \left(\rho \mathbf{I} - \frac{d}{d-1} \mathbf{Q}' \right) \delta(t - t') \left(\frac{\delta(r - r')}{r^2} \right). \quad (\text{C.100})$$

As found in Ref. [42], $\mathbf{Q}'$ is given by:

$$\mathbf{Q}' = -\frac{d-1}{\ell_o^2 D_R \zeta \bar{U}(\rho)} \left[\kappa_1(\rho) \nabla^2 \rho \mathbf{I} + \kappa_3(\rho) \nabla \rho \nabla \rho \right]. \quad (\text{C.101})$$

Thus, $\boldsymbol{\eta}^{\text{act}}$ may have anisotropic contributions to its correlator, particularly at interfaces where gradients in the density are large. This anisotropy is due to $\boldsymbol{\eta}^{\text{act}}$'s microscopic roots in the aggregate of individual $\boldsymbol{\Omega}_i \times \mathbf{q}_i$. In regions of space with nematic alignment, the result of the cross product of an isotropic noise with each orientation $\mathbf{q}_i$ will be biased towards directions orthogonal to that alignment. Before finding the statistics of $\tilde{\chi}$, we first find the statistics of $\xi_R(\mathbf{r}, t)$, which we define as

$$\xi_R(\mathbf{r}, t) = -\nabla \cdot \boldsymbol{\eta}^{\text{act}}. \quad (\text{C.102})$$

We split the correlations of ξ_R into three contributions,

$$\begin{aligned} \langle \xi_R(\mathbf{r}, t) \xi_R(\mathbf{r}', t') \rangle &= \langle \xi_R(\mathbf{r}, t) \xi_R(\mathbf{r}', t') \rangle^{\text{iso}} + \langle \xi_R(\mathbf{r}, t) \xi_R(\mathbf{r}', t') \rangle_1^{\text{aniso}} \\ &\quad + \langle \xi_R(\mathbf{r}, t) \xi_R(\mathbf{r}', t') \rangle_2^{\text{aniso}}, \end{aligned} \quad (\text{C.103})$$

where the contribution labeled iso originates from the component of the $\boldsymbol{\eta}^{\text{act}}$ correlator proportional to $\rho \mathbf{I}$ while the contributions labeled aniso originates from the component of the $\boldsymbol{\eta}^{\text{act}}$ correlator proportional to $\mathbf{Q}'$. Each contribution is given by:

$$\langle \xi_R(\mathbf{r}, t) \xi_R(\mathbf{r}', t') \rangle^{\text{iso}} = -\frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \left(\frac{\partial}{\partial r} + \frac{2}{r} \right) \left[\rho \frac{\partial}{\partial r} \left(\frac{\delta(r-r')}{r^2} \right) \right], \quad (\text{C.104})$$

$$\begin{aligned} \langle \xi_R(\mathbf{r}, t) \xi_R(\mathbf{r}', t') \rangle_1^{\text{aniso}} &= -\frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \\ &\times \left(\frac{\partial}{\partial r} + \frac{2}{r} \right) \left[\left(\frac{1}{\ell_o^2 D_R \zeta \bar{U}} \left[\kappa_1(\rho) \left(\frac{\partial}{\partial r} + \frac{2}{r} \right) \frac{\partial \rho}{\partial r} - \kappa_3(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 \right] \right) \frac{\partial}{\partial r} \left(\frac{\delta(r-r')}{r^2} \right) \right], \end{aligned} \quad (\text{C.105})$$

and

$$\begin{aligned} \langle \xi_R(\mathbf{r}, t) \xi_R(\mathbf{r}', t') \rangle_2^{\text{aniso}} &= \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t - t') \\ &\quad \left(\frac{\partial}{\partial r} + \frac{2}{r} \right) \left[\left(\frac{1}{r'} \frac{1}{\ell_o^2 D_R \zeta \bar{U}} \left[\kappa_3(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 \right] \right) \frac{\delta(r - r')}{r^2} \right]. \end{aligned} \quad (\text{C.106})$$

We now define $\chi(t)$ as:

$$\chi(t) = - \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \int_r^\infty dr_2 \frac{1}{r_2^2} \int_0^{r_2} r_1^2 \xi_R(\mathbf{r}_1, t). \quad (\text{C.107})$$

The correlations of $\chi(t)$ are then given by

$$\begin{aligned} \langle \chi(t) \chi(t') \rangle &= \\ &\quad \left\langle \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \int_r^\infty dr_2 \int_0^{r_2} \xi_R(\mathbf{r}_1, t) \int_0^\infty dr' \frac{\partial \mathcal{E}}{\partial r'} \right. \\ &\quad \left. \times \int_r^\infty dr_4 \int_0^{r_4} \left(\frac{r_1 r_3}{r_2 r_4} \right)^2 \xi_R(\mathbf{r}_3, t') \right\rangle, \end{aligned} \quad (\text{C.108})$$

which can be rewritten as:

$$\begin{aligned} \langle \chi(t) \chi(t') \rangle &= \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \int_r^\infty \int_{r'}^\infty \int_0^{r_2} \int_0^{r_4} dr_1 dr_2 dr_3 dr_4 \\ &\quad \times \left(\frac{r_1 r_3}{r_2 r_4} \right)^2 \langle \xi_R(\mathbf{r}_1, t) \xi_R(\mathbf{r}_3, t') \rangle. \end{aligned} \quad (\text{C.109})$$

Now we similarly divide $\chi(t)$ into contributions:

$$\langle \chi(t) \chi(t') \rangle = \langle \chi(t) \chi(t') \rangle^{\text{iso}} + \langle \chi(t) \chi(t') \rangle_1^{\text{aniso}} + \langle \chi(t) \chi(t') \rangle_2^{\text{aniso}}, \quad (\text{C.110})$$

where

$$\begin{aligned} \langle \chi(t) \chi(t') \rangle^{\text{iso}} &= \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\ &\quad \times \int_r^\infty \int_{r'}^\infty \int_0^{r_2} \int_0^{r_4} dr_1 dr_2 dr_3 dr_4 \left(\frac{r_1 r_3}{r_2 r_4} \right)^2 \langle \xi_R(\mathbf{r}_1, t) \xi_R(\mathbf{r}_3, t') \rangle^{\text{iso}}, \end{aligned} \quad (\text{C.111})$$

$$\begin{aligned} \langle \chi(t)\chi(t') \rangle_1^{\text{aniso}} &= \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\ &\times \int_r^\infty \int_{r'}^\infty \int_0^{r_2} \int_0^{r_4} dr_1 dr_2 dr_3 dr_4 \left(\frac{r_1 r_3}{r_2 r_4} \right)^2 \langle \xi_R(\mathbf{r}_1, t) \xi_R(\mathbf{r}_3, t') \rangle_1^{\text{aniso}}, \end{aligned} \quad (\text{C.112})$$

and

$$\begin{aligned} \langle \chi(t)\chi(t') \rangle_2^{\text{aniso}} &= \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\ &\times \int_r^\infty \int_{r'}^\infty \int_0^{r_2} \int_0^{r_4} dr_1 dr_2 dr_3 dr_4 \left(\frac{r_1 r_3}{r_2 r_4} \right)^2 \langle \xi_R(\mathbf{r}_1, t) \xi_R(\mathbf{r}_3, t') \rangle_2^{\text{aniso}}. \end{aligned} \quad (\text{C.113})$$

We will first solve for $\langle \chi(t)\chi(t') \rangle^{\text{iso}}$. Substitution of Eq. (C.104) into Eq. (C.111) results in:

$$\begin{aligned} \langle \chi(t)\chi(t') \rangle^{\text{iso}} &= \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\ &\times \int_r^\infty \int_{r'}^\infty \int_0^{r_2} \int_0^{r_4} dr_1 dr_2 dr_3 dr_4 \left(\frac{r_1 r_3}{r_2 r_4} \right)^2 \\ &\times \left(\frac{\partial}{\partial r_1} + \frac{2}{r_1} \right) \left[\rho(r_1) \frac{\partial}{\partial r_1} \left(\frac{\delta(r_1 - r_3)}{r_1^2} \right) \right]. \end{aligned} \quad (\text{C.114})$$

We now distribute the divergence in r_1 to find:

$$\begin{aligned} \langle \chi(t)\chi(t') \rangle^{\text{iso}} &= \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\ &\times \int_r^\infty \int_{r'}^\infty \int_0^{r_2} \int_0^{r_4} dr_1 dr_2 dr_3 dr_4 \left(\frac{r_1 r_3}{r_2 r_4} \right)^2 \frac{\partial}{\partial r_1} \left[\rho(r_1) \frac{\partial}{\partial r_1} \left(\frac{\delta(r_1 - r_3)}{r_1^2} \right) \right] \\ &+ \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\ &\times \int_r^\infty \int_{r'}^\infty \int_0^{r_2} \int_0^{r_4} dr_1 dr_2 dr_3 dr_4 \left(\frac{r_3}{r_2 r_4} \right)^2 2r_1 \left[\rho(r_1) \frac{\partial}{\partial r_1} \left(\frac{\delta(r_1 - r_3)}{r_1^2} \right) \right]. \end{aligned} \quad (\text{C.115})$$

We now integrate the top integral by parts, which results in a boundary term as well as an integral which cancels out with the bottom integral. We are then left with:

$$\begin{aligned}
\langle \chi(t) \chi(t') \rangle^{\text{iso}} = & -\frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
& \times \int_r^\infty \int_{r'}^\infty \int_0^{r_4} dr_2 dr_3 dr_4 \\
& \times \left(\frac{r_1 r_3}{r_2 r_4} \right)^2 \left(\rho(r_1) \frac{\partial}{\partial r_1} \left(\frac{\delta(r_1 - r_3)}{r_1^2} \right) \right) \bigg|_{r_1=0}^{r_1=r_2} \quad (\text{C.116})
\end{aligned}$$

Evaluation of the limits must be done with care, particularly for the $r_1 = 0$ term. Before evaluating these limits, we rearrange such that there are no derivatives in r_1 :

$$\begin{aligned}
\langle \chi(t) \chi(t') \rangle^{\text{iso}} = & \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
& \times \int_r^\infty \int_{r'}^\infty \int_0^{r_4} dr_2 dr_3 dr_4 \left(\frac{r_3}{r_2 r_4} \right)^2 \rho(r_1) \frac{2}{r_1} \delta(r_1 - r_3) \bigg|_{r_1=0}^{r_1=r_2} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
& \times \int_r^\infty \int_{r'}^\infty \int_0^{r_4} dr_2 dr_3 dr_4 \left(\frac{r_3}{r_2 r_4} \right)^2 \left(\rho(r_1) \frac{\partial}{\partial r_3} \delta(r_1 - r_3) \right) \bigg|_{r_1=0}^{r_1=r_2}. \quad (\text{C.117})
\end{aligned}$$

We now evaluate the limits, noting that the $r_1 \rightarrow 0$ limits will vanish:

$$\begin{aligned}
\langle \chi(t)\chi(t') \rangle^{\text{iso}} &= \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
&\quad \times \int_r^\infty \int_{r'}^\infty \int_0^{r_4} dr_2 dr_3 dr_4 \left(\frac{r_3}{r_2 r_4} \right)^2 \rho(r_2) \frac{2}{r_2} \delta(r_3 - r_2) \\
&\quad + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
&\quad \times \int_r^\infty \int_{r'}^\infty \int_0^{r_4} dr_2 dr_3 dr_4 \left(\frac{r_3}{r_2 r_4} \right)^2 \left(\rho(r_2) \frac{\partial}{\partial r_3} \delta(r_2 - r_3) \right).
\end{aligned} \tag{C.118}$$

We now integrate the first integral with respect to r_3 . Because the integral with respect to r_3 goes from $r_3 = 0$ to $r_3 = r_4$, the integral will be zero if $r_4 < r_2$. However, if $r_4 > r_2$, we can apply the sifting property of the delta function. We can handle both cases by applying the sifting function while applying up a factor of $H(r_4 - r_2)$,

$$\begin{aligned}
\langle \chi(t)\chi(t') \rangle^{\text{iso}} &= \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
&\quad \times \int_r^\infty \int_{r'}^\infty dr_2 dr_4 \left(\frac{1}{r_4} \right)^2 \rho(r_2) \frac{2}{r_2} H(r_4 - r_2) \\
&\quad + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
&\quad \times \int_r^\infty \int_{r'}^\infty \int_0^{r_4} dr_2 dr_3 dr_4 \left(\frac{r_3}{r_2 r_4} \right)^2 \left(\rho(r_2) \frac{\partial}{\partial r_3} \delta(r_2 - r_3) \right),
\end{aligned} \tag{C.119}$$

where H is the Heaviside step function. We can now integrate the bottom integral

by parts which results in:

$$\begin{aligned}
\langle \chi(t)\chi(t') \rangle^{\text{iso}} &= \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
&\quad \times \int_r^\infty \int_{r'}^\infty dr_2 dr_4 \left(\frac{1}{r_4} \right)^2 \rho(r_2) \frac{2}{r_2} H(r_4 - r_2) \\
&\quad + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
&\quad \times \int_r^\infty \int_{r'}^\infty dr_2 dr_4 \left(\frac{r_3}{r_2 r_4} \right)^2 (\rho(r_2) \delta(r_2 - r_3)) \Bigg|_{r_3=0}^{r_3=r_4} \\
&\quad - \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
&\quad \times \int_r^\infty \int_{r'}^\infty \int_0^{r_4} dr_2 dr_3 dr_4 \left(\frac{1}{r_2 r_4} \right)^2 (2r_3 \rho(r_2) \delta(r_2 - r_3)) .
\end{aligned} \tag{C.120}$$

Evaluate the integral in r_3 of the bottom integral leads to a term which cancels with the top integral, resulting in:

$$\begin{aligned}
\langle \chi(t)\chi(t') \rangle^{\text{iso}} &= \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \\
&\quad \times \int_r^\infty \int_{r'}^\infty dr_2 dr_4 \left(\frac{r_3}{r_2 r_4} \right)^2 (\rho(r_2) \delta(r_2 - r_3)) \Bigg|_{r_3=0}^{r_3=r_4} .
\end{aligned} \tag{C.121}$$

We can now evaluate the limits,

$$\begin{aligned}
\langle \chi(t)\chi(t') \rangle^{\text{iso}} &= \\
&= \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \int_r^\infty dr_2 \left(\frac{1}{r_2} \right)^2 \rho(r_2) \int_{r'}^\infty dr_4 \delta(r_2 - r_4) .
\end{aligned} \tag{C.122}$$

Evaluating the integral in r_4 now depends on r' . If $r' < r_2$ we may apply the sifting property of the delta function. If $r' > r_2$ the integral vanishes. Therefore we apply the sifting property and gain a factor of the Heaviside step function:

$$\langle \chi(t) \chi(t') \rangle^{\text{iso}} = \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t - t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \int_r^\infty dr_2 \left(\frac{1}{r_2} \right)^2 \rho(r_2) H(r_2 - r'). \quad (\text{C.123})$$

If $r < r'$ then the integral will be zero until r_2 reaches r' . Then we may absorb the step function into the limits of integration:

$$\langle \chi(t) \chi(t') \rangle^{\text{iso}} = \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t - t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \int_{\max(r, r')}^\infty dr_2 \left(\frac{1}{r_2} \right)^2 \rho(r_2). \quad (\text{C.124})$$

Integration by parts results in:

$$\begin{aligned} \langle \chi(t) \chi(t') \rangle^{\text{iso}} = & \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t - t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \frac{\rho(\max(r, r'))}{\max(r, r')} \\ & + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t - t') \int_0^\infty \int_0^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \int_{\max(r, r')}^\infty dr_2 \left(\frac{1}{r_2} \right) \frac{\partial \rho}{\partial r_2}. \end{aligned} \quad (\text{C.125})$$

The $\max(r, r')$ functions are cumbersome to deal with as is. We therefore divide

the integrals over r' into sections where $r' < r$ or $r' > r$:

$$\begin{aligned}
\langle \chi(t) \chi(t') \rangle^{\text{iso}} = & \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^r dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \frac{\rho(r)}{r} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_r^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \frac{\rho(r')}{r'} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^r dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \int_r^\infty dr_2 \left(\frac{1}{r_2} \right) \frac{\partial \rho}{\partial r_2} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_r^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \int_{r'}^\infty dr_2 \left(\frac{1}{r_2} \right) \frac{\partial \rho}{\partial r_2}.
\end{aligned} \tag{C.126}$$

We now integrate the last line by parts in r' , directly evaluate the integral over r' in the second line, and combine like terms. This results in:

$$\begin{aligned}
\langle \chi(t) \chi(t') \rangle^{\text{iso}} = & \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_0^r dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \frac{\rho(r)}{r} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty \int_r^\infty dr dr' \frac{\partial \mathcal{E}}{\partial r} \frac{\partial \mathcal{E}}{\partial r'} \frac{\rho(r')}{r'} \\
& - \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \int_r^\infty dr_2 \left(\frac{1}{r_2} \right) \frac{\partial \rho}{\partial r_2} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \int_r^\infty dr' \mathcal{E} \left(\frac{1}{r'} \right) \frac{\partial \rho}{\partial r'}.
\end{aligned} \tag{C.127}$$

We now integrate the second integral of the first line by parts and directly evaluate

the r' integral in the first term of the first line, which results in:

$$\begin{aligned}
\langle \chi(t) \chi(t') \rangle^{\text{iso}} = & \frac{4k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \mathcal{E} \frac{\rho(r)}{r} \\
& - \frac{4k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \frac{\rho(r)}{r} \\
& - \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \int_r^\infty dr_2 \left(\frac{1}{r_2} \right) \frac{\partial \rho}{\partial r_2} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \int_r^\infty dr' \mathcal{E} \left(\frac{1}{r'} \right) \frac{\partial \rho}{\partial r'}. \quad (\text{C.128})
\end{aligned}$$

We now integrate both the second and third line by parts, then combine like terms, which results in:

$$\begin{aligned}
\langle \chi(t) \chi(t') \rangle^{\text{iso}} = & \frac{4k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \mathcal{E} \frac{\rho(r)}{r} \\
& - \frac{4k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \frac{\rho(r)}{r} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \left(\mathcal{E}^{\text{in}} \right)^2 \int_0^\infty dr \left(\frac{1}{r} \right) \frac{\partial \rho}{\partial r} \\
& - \frac{4k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \mathcal{E} \left(\frac{1}{r} \right) \frac{\partial \rho}{\partial r} \\
& + \frac{2k_B T^{\text{act}}}{4\pi\zeta} \delta(t-t') \int_0^\infty dr (\mathcal{E}(r))^2 \left(\frac{1}{r} \right) \frac{\partial \rho}{\partial r}. \quad (\text{C.129})
\end{aligned}$$

Now, as was assumed before, we note that each of the above integrals is non-zero only near the interface. Then if R is much larger than the width of the interface,

we may rewrite each integral as:

$$\begin{aligned}
\langle \chi(t)\chi(t') \rangle^{\text{iso}} = & \frac{4k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \mathcal{E} \rho(r) \\
& - \frac{4k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \rho(r) \\
& + \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \left(\mathcal{E}^{\text{in}} \right)^2 \int_0^\infty dr \frac{\partial \rho}{\partial r} \\
& - \frac{4k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \mathcal{E} \frac{\partial \rho}{\partial r} \\
& + \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \int_0^\infty dr (\mathcal{E}(r))^2 \frac{\partial \rho}{\partial r} + O(1/R^2). \quad (\text{C.130})
\end{aligned}$$

The first integral may be rewritten as:

$$\begin{aligned}
\langle \chi(t)\chi(t') \rangle^{\text{iso}} = & \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \int_0^\infty dr \frac{\partial}{\partial r} [(\mathcal{E})^2] \rho(r) \\
& - \frac{4k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \frac{\partial \mathcal{E}}{\partial r} \rho(r) \\
& + \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \left(\mathcal{E}^{\text{in}} \right)^2 \int_0^\infty dr \frac{\partial \rho}{\partial r} \\
& - \frac{4k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \mathcal{E} \frac{\partial \rho}{\partial r} \\
& + \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \int_0^\infty dr (\mathcal{E}(r))^2 \frac{\partial \rho}{\partial r} + O(1/R^2). \quad (\text{C.131})
\end{aligned}$$

Then the above integrals may be compactly rewritten as:

$$\begin{aligned} \langle \chi(t)\chi(t') \rangle^{\text{iso}} = & \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \int_0^\infty dr \frac{\partial}{\partial r} [(\mathcal{E})^2 \rho(r)] \\ & - \frac{4k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \mathcal{E}^{\text{in}} \int_0^\infty dr \frac{\partial}{\partial r} [\mathcal{E}\rho] \\ & + \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') (\mathcal{E}^{\text{in}})^2 \int_0^\infty dr \frac{\partial \rho}{\partial r} + O(1/R^2). \end{aligned} \quad (\text{C.132})$$

All integrals may now be evaluated directly and simplify to:

$$\langle \chi(t)\chi(t') \rangle^{\text{iso}} = \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \rho^{\text{sat}} (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}})^2 + O(1/R^2). \quad (\text{C.133})$$

$\langle \chi(t)\chi(t') \rangle_1^{\text{aniso}}$ and $\langle \chi(t)\chi(t') \rangle_2^{\text{aniso}}$ can be solved for in a similar manner as $\langle \chi(t)\chi(t') \rangle^{\text{iso}}$, and we find that $\langle \chi(t)\chi(t') \rangle_1^{\text{aniso}}$ is zero while $\langle \chi(t)\chi(t') \rangle_2^{\text{aniso}}$ is:

$$\begin{aligned} \langle \chi(t)\chi(t') \rangle_2^{\text{aniso}} = & \frac{4k_B T^{\text{act}}}{4\pi D_R \zeta \ell_o^2 R^2} \Delta \mathcal{E} \delta(t-t') \left[\int_0^\infty dr \frac{\kappa_3(\rho)}{\bar{U}(\rho)} \left(\frac{\partial \rho}{\partial r} \right)^2 (\mathcal{E}^{\text{in}} - \mathcal{E}) \right] + O\left(\frac{1}{R^3}\right), \end{aligned} \quad (\text{C.134})$$

which implies that the most significant contributions to the anisotropic noise correlations are $O(1/R^2)$ or smaller. Therefore,

$$\langle \chi(t)\chi(t') \rangle = \frac{2k_B T^{\text{act}}}{4R\pi\zeta} \delta(t-t') \rho^{\text{sat}} (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}})^2 + O(1/R^2). \quad (\text{C.135})$$

We now define $\tilde{\chi}$ as:

$$\tilde{\chi}(t) = -\frac{\zeta_{\text{eff}}}{R (\rho^{\text{sat}} - \rho^{\text{in}}) (\mathcal{E}^{\text{sat}} - \mathcal{E}^{\text{in}})} \chi(t), \quad (\text{C.136})$$

which will have zero mean and correlations:

$$\langle \tilde{\chi}(t)\tilde{\chi}(t') \rangle = 2k_B T^{\text{act}} \zeta_{\text{eff}} \delta(t-t'). \quad (\text{C.137})$$

Cates and Nardini [78] have argued that the nucleation of active matter must have a defineable potential barrier because Eqs. (C.95) and (C.99) satisfy an effective fluctuation dissipation theorem. This nucleation potential is defined such that $F^D(R) = -\partial U(R)/\partial R$:

$$U(R) = \int dR 8\pi R^2 \gamma_{\text{ost}} \left[\frac{1}{R} - \frac{1}{R_C} \right], \quad (\text{C.138})$$

where $F^D(R)$ is defined by Eq. (C.97). In general, evaluating this integral analytically is difficult because γ_{ost} is implicitly a function of R . Cates and Nardini proposed *neglecting* the R dependence of γ_{ost} and treating it as a constant:

$$U(R) = 8\pi \gamma_{\text{ost}} \left[\frac{R^2}{2} - \frac{R^3}{3R_C} \right]. \quad (\text{C.139})$$

We verify the validity of this approximation by evaluating the *integrand* of $U(R)$ both numerically and via the approximation proposed by Cates and Nardini. We follow the same numerical scheme as discussed in the previous section, except now R is taken as an input and the two unknown values solved for are ρ^{in} and ρ^{out} . The result of calculating $-\partial U(R)/\partial R$ both from our numerical scheme and the approximation of Cates and Nardini is plotted in Fig. C.2.

The difference between ignoring and accounting for the R dependence of γ_{ost} is marginal for both bubbles and droplets, as shown by Fig. C.2. Therefore evaluating coefficients at $R = R_C$ and treating them as constant is a robust approximation, justifying the use of Eq. (C.139). We can then define a potential barrier as $U(R_C)$, which is given by:

$$U(R_C) = \frac{4}{3}\pi \gamma_{\text{ost}} R_C^2. \quad (\text{C.140})$$

We use Eq. (C.140) to calculate the nucleation barriers presented in the main text.

We may also substitute R_C as defined by Eq. (C.39) into Eq. (C.139) to find a clean separation between terms proportional to surface area and volume:

$$U(R) = 4\pi \gamma_{\text{ost}} R^2 - \frac{4}{3}\pi R^3 \mathcal{V}, \quad (\text{C.141})$$

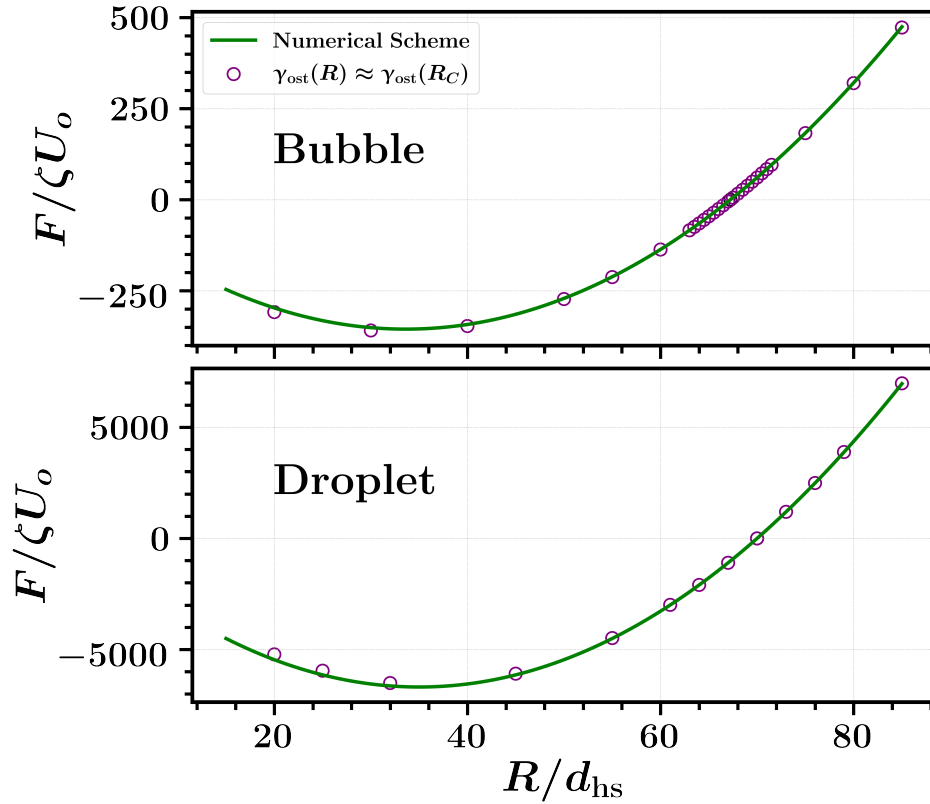


Figure C.2: Deterministic force as defined by the negative derivative in R of Eq. (C.138) as evaluated via the numerical scheme described above or treating coefficients as constant in R . The bubble forces were evaluated for a system at $\ell_o/d_{hs} = 70.7$ and $R_C/d_{hs} = 67$. The droplet forces were evaluated for a system at $\ell_o/d_{hs} = 70.7$ and $R_C/d_{hs} = 70$.

where $\mathcal{V}$ has units of energy per volume and has form:

$$\mathcal{V} = \frac{\mathcal{P}(\rho^{\text{sat}})\Delta\rho}{\rho^{\text{sat}}} - \frac{\Delta\Phi\Delta\rho}{\rho^{\text{sat}}\Delta\mathcal{E}}. \quad (\text{C.142})$$

C.4 Becker-Döring Nucleation Rate Calculation

The existence of an effective potential and the establishment of nuclei obeying an effective detailed balance allows for one to extend equilibrium Becker-Döring theory [32, 45, 112] to active particles in order to predict the rate of droplet

nucleation. The nucleation event is characterized by the formation of a cluster with a critical number of particles, so we would like to determine the rate at which such critical clusters form. We will denote the number of clusters with ℓ particles per unit volume as c_ℓ . Additionally, we will assume that clusters are formed and destroyed by *monomer* attachment and detachment. In other words, a size $(\ell + 1)$ cluster is formed by the merging of a size ℓ cluster and a size 1 cluster. The opposite process would be a size ℓ cluster being destroyed by splitting into a size $\ell - 1$ cluster and a size 1 cluster. The *flux*, J_ℓ , in cluster-size space of ℓ clusters is then given by the difference between the rate at which droplets containing ℓ particles are formed and the rate at which they are destroyed:

$$J_\ell = c_{\ell-1}S_{\ell-1}\beta_{\ell-1} - c_\ell S_\ell \alpha_\ell; \ell \geq 2. \quad (\text{C.143})$$

Here S_ℓ is the surface area of an ℓ cluster, β_ℓ is the flux of single particles arriving on the surface of the cluster, and α_ℓ is the flux of single particles leaving the surface of the cluster. The evolution of c_ℓ is then given by:

$$\frac{\partial c_\ell}{\partial t} = J_\ell - J_{\ell+1}. \quad (\text{C.144})$$

We now need to distinguish between two useful limits. One limit, which we call *steady state* requires that all $\partial_t c_\ell = 0$ but the individual J_ℓ may remain finite. The other limit, which we call *quasi-equilibrium* requires that all $\partial_t c_\ell = 0$ but additionally requires that all individual $J_\ell = 0$. The effective detailed balance established for nucleation dynamics implies that the dynamics which govern quasi-equilibrium must also lead to quasi-equilibrium, as Eq. (C.95) obeys an effective time-reversal symmetry. This implies that the values of β_ℓ and α_ℓ which are valid at quasi-equilibrium must also be valid in the steady-state. Cates and Nardini [78] established that, to linear approximation, the principle of detailed balance is recovered by nucleating clusters in active system even if no such principle exists microscopically. Then α_ℓ is:

$$\alpha_\ell = \frac{c_{\ell-1}^{\text{eq}} S_{\ell-1} \beta_{\ell-1}}{c_\ell^{\text{eq}} S_\ell}, \quad (\text{C.145})$$

and the flux J_ℓ at *steady-state* may be expressed as:

$$J_\ell = c_{\ell-1}S_{\ell-1}\beta_{\ell-1} - c_\ell S_\ell \frac{c_{\ell-1}^{\text{eq}} S_{\ell-1} \beta_{\ell-1}}{c_\ell^{\text{eq}} S_\ell}. \quad (\text{C.146})$$

This flux may be rewritten as:

$$J_\ell = \beta_{\ell-1} S_{\ell-1} c_{\ell-1}^{\text{eq}} \left[\frac{c_{\ell-1}}{c_{\ell-1}^{\text{eq}}} - \frac{c_\ell}{c_\ell^{\text{eq}}} \right]. \quad (\text{C.147})$$

Note that in the steady state all J_ℓ with $\ell \geq 2$ are equal to $J_{\ell+1}$. Then we can label $J_\ell = \mathcal{R}$ and write:

$$\frac{\mathcal{R}}{\beta_{\ell-1} S_{\ell-1} c_{\ell-1}^{\text{eq}}} = \left[\frac{c_{\ell-1}}{c_{\ell-1}^{\text{eq}}} - \frac{c_\ell}{c_\ell^{\text{eq}}} \right]. \quad (\text{C.148})$$

We now sum both sides of the equation from $\ell = 2$ to $\ell = \Lambda + 1$, where Λ is some very large number (larger than the critical nucleus size). After summing, the right hand side of Eq. (C.148) will vanish except for the first term where $\ell = 2$ and the last term where $\ell = \Lambda + 1$:

$$\mathcal{R} = \left[\frac{c_1}{c_1^{\text{eq}}} - \frac{c_{\Lambda+1}}{c_{\Lambda+1}^{\text{eq}}} \right] \left[\sum_{\ell=1}^{\Lambda} \frac{1}{\beta_\ell S_\ell c_\ell^{\text{eq}}} \right]^{-1}. \quad (\text{C.149})$$

We now argue that since the bulk metastable phase (a dilute gas) exists overwhelmingly as single particles, so the quasi-equilibrium and steady-state concentration of single particles are indistinguishable and $c_1/c_1^{\text{eq}} \approx 1$. Additionally, Λ is larger than the size of the critical nucleus, so $c_{\Lambda+1}$ should be zero in the metastable steady state but *finite* at quasi-equilibrium. Then Eq. (C.149) reduces to:

$$\mathcal{R} = \left[\sum_{\ell=1}^{\Lambda} \frac{1}{\beta_\ell S_\ell c_\ell^{\text{eq}}} \right]^{-1}. \quad (\text{C.150})$$

Using the effective nucleation potential [78], we know the relative probability of observing a nucleus with radius R , and (given a spherical geometry), we therefore we have an expression for c_ℓ^{eq} as:

$$c_\ell^{\text{eq}} = \rho^{\text{sat}} \exp \left[(k_B T^{\text{act}})^{-1} U(R_\ell) \right], \quad (\text{C.151})$$

where $U(R_\ell)$ is the effective potential evaluated at a radius corresponding to a sphere of ℓ particles. Switching from a summation to an integral, we can then approximate the nucleation rate as:

$$\mathcal{R} = \rho^{\text{sat}} \left[\int_1^\Lambda d\ell \frac{1}{\beta_\ell S_\ell} \exp \left[\frac{U(R_\ell)}{k_B T^{\text{act}}} \right] \right]^{-1}. \quad (\text{C.152})$$

We now would like to make the connection between radius and cluster size more explicit:

$$\ell = \frac{4}{3} \pi R^3 \rho^{\text{extin}}. \quad (\text{C.153})$$

Alternatively, one may rearrange Eq. (C.153) and obtain an expression for the radius in terms of size:

$$R = \left[\frac{3\ell}{4\pi\rho^{\text{in}}} \right]^{1/3}. \quad (\text{C.154})$$

We now consider the derived form of the nucleation barrier, Eq. (C.141), and express it in terms of cluster size:

$$U(\ell) = 4\pi\gamma_{\text{ost}} \left[\frac{3\ell}{4\pi\rho^{\text{in}}} \right]^{2/3} - \frac{\ell\mathcal{V}}{\rho^{\text{in}}}. \quad (\text{C.155})$$

From this form the critical cluster size ℓ^* can be solved for by extremizing $U(\ell)$, resulting in:

$$\ell^* = \frac{32}{3} \pi \rho^{\text{in}} \left(\frac{\gamma_{\text{ost}}}{\mathcal{V}} \right)^3. \quad (\text{C.156})$$

We now Taylor expand the the barrier about this maximum to second order,

$$U(\ell) = U(\ell^*) - \frac{1}{2} \eta (\ell - \ell^*)^2, \quad (\text{C.157})$$

where we have defined η as the second derivative of the barrier with respect to ℓ and evaluated at ℓ^* . Explicitly, this is solved for as:

$$\eta = \frac{1}{32\pi (\rho^{\text{in}})^2} \frac{\mathcal{V}^4}{\gamma_{\text{ost}}^3}. \quad (\text{C.158})$$

We now consider the integral from Eq. (C.152). Substitution of Eq. (C.157) into this integral results in:

$$\int_1^\Lambda d\ell \frac{1}{\beta_\ell S_\ell} \exp \left[\frac{U(R_\ell)}{k_B T^{\text{act}}} \right] = \int_1^\Lambda d\ell \frac{1}{\beta_\ell S_\ell} \exp \left[\frac{U(\ell^*)}{k_B T^{\text{act}}} \right] \exp \left[-\frac{\eta(\ell - \ell^*)^2}{2k_B T^{\text{act}}} \right]. \quad (\text{C.159})$$

The exponential of the barrier can be pulled out of the integral, which leads to:

$$\int_1^\Lambda d\ell \frac{1}{\beta_\ell S_\ell} \exp \left[\frac{U(R_\ell)}{k_B T^{\text{act}}} \right] = \exp \left[\frac{U(\ell^*)}{k_B T^{\text{act}}} \right] \int_1^\Lambda d\ell \frac{1}{\beta_\ell S_\ell} \exp \left[-\frac{\eta(\ell - \ell^*)^2}{2k_B T^{\text{act}}} \right]. \quad (\text{C.160})$$

We now change integration variables from ℓ to $\delta\ell = \ell - \ell^*$:

$$\int_1^\Lambda d\ell \frac{1}{\beta_\ell S_\ell} \exp \left[\frac{U(R_\ell)}{k_B T^{\text{act}}} \right] = \exp \left[\frac{U(\ell^*)}{k_B T^{\text{act}}} \right] \int_{1-\ell^*}^{\Lambda-\ell^*} d\delta\ell \frac{1}{\beta_{\ell^*} S_{\ell^*}} \exp \left[-\frac{\eta \delta\ell^2}{2k_B T^{\text{act}}} \right]. \quad (\text{C.161})$$

The integral will be dominated by value at which it is peaked, which is where $\delta\ell = 0$, as long as $1/\eta$ is large. Then we approximate β_ℓ, S_ℓ as their values when $\ell = \ell^*$, resulting in:

$$\begin{aligned} \int_1^\Lambda d\ell \frac{1}{\beta_\ell S_\ell} \exp \left[\frac{U(R_\ell)}{k_B T^{\text{act}}} \right] = \\ \frac{1}{\beta_{\ell^*} 4\pi \left(\frac{3\ell^*}{4\pi\rho^{\text{in}}} \right)^{2/3}} \exp \left[\frac{U(\ell^*)}{k_B T^{\text{act}}} \right] \int_{-\infty}^{\infty} d\delta\ell \exp \left[-\frac{\eta \delta\ell^2}{2k_B T^{\text{act}}} \right]. \end{aligned} \quad (\text{C.162})$$

Finally, we can evaluate the Gaussian integral:

$$\int_1^\Lambda d\ell \frac{1}{\beta_\ell S_\ell} \exp \left[\frac{U(R_\ell)}{k_B T^{\text{act}}} \right] = \frac{1}{\beta_{\ell^*} 4\pi \left(\frac{3\ell^*}{4\pi\rho^{\text{in}}} \right)^{2/3}} \exp \left[\frac{U(\ell^*)}{k_B T^{\text{act}}} \right] \sqrt{\frac{2\pi k_B T^{\text{act}}}{\eta}}. \quad (\text{C.163})$$

Substitution of Eq. (C.163) into Eq. (C.152) results in an expression for the rate of nucleation:

$$\mathcal{R} = \rho^{\text{sat}} \beta_{\ell^*} 4\pi \left(\frac{3\ell^*}{4\pi\rho^{\text{in}}} \right)^{2/3} \sqrt{\frac{\eta}{2\pi k_B T^{\text{act}}}} \exp \left[-\frac{U(\ell^*)}{k_B T^{\text{act}}} \right]. \quad (\text{C.164})$$

Everything in Eq. (C.164) can now be evaluated *except* for the rate of single particle attachment on the critical nucleus β_{ℓ^*} , for which we use the simple kinetic model proposed Redner [76, 110]. β_ℓ is the rate of absorption of particles onto the surface of a cluster. This rate should be given by the density of the gas phase times the speed at which the particles swim times the probability that a given particle will absorb onto the surface:

$$\beta_\ell = \rho^{\text{sat}} U_o P^{\text{abs}}, \quad (\text{C.165})$$

where P^{abs} is the probability a particle absorbs onto the surface. Assuming that the surface of the nucleus has an inward pointing normal vector $\mathbf{n}$, we model P^{abs} as *zero* if a particle points away from the interface and proportional to $\mathbf{n} \cdot \mathbf{q}$ if pointing in the hemisphere towards interface. In other words P^{abs} , is given by:

$$P^{\text{abs}} = \begin{cases} \mathbf{n} \cdot \mathbf{q} & \mathbf{n} \cdot \mathbf{q} > 0 \\ 0 & \mathbf{n} \cdot \mathbf{q} < 0 \end{cases} \quad (\text{C.166})$$

We pick a point on the sphere with unit normal vector $-\mathbf{e}_x$, and then represent the orientation of a given particle as:

$$\mathbf{q} = \sin \theta \cos \phi \mathbf{e}_x + \sin \theta \sin \phi \mathbf{e}_y + \cos \theta \mathbf{e}_z. \quad (\text{C.167})$$

Then, assuming that the orientations of the relevant particles are uniformly distributed, the *average* of β_ℓ can be expressed as:

$$\langle \beta_\ell \rangle = \rho^{\text{sat}} U_o \frac{1}{4\pi} \int_0^{2\pi} \int_0^\pi \sin \theta P^{\text{abs}} d\theta d\phi. \quad (\text{C.168})$$

P^{abs} is only nonzero when ϕ is between 0 and $\pi/2$ as well as when ϕ is between $3\pi/2$ and 2π , for all values of θ . Then Eq. (C.168) reduces to:

$$\begin{aligned} \langle \beta_\ell \rangle &= \rho^{\text{sat}} U_o \frac{1}{2\pi^2} \left[\int_0^{\pi/2} \int_0^\pi \sin^2 \theta \cos \phi d\theta d\phi + \int_{3\pi/2}^{2\pi} \int_0^\pi \sin^2 \theta \cos \phi d\theta d\phi \right] \\ &= \frac{\rho^{\text{sat}} U_o}{4}. \end{aligned} \quad (\text{C.169})$$

Substitution of Eq. (C.169) into Eq. (C.164) results in:

$$\mathcal{R} = (\rho^{\text{sat}})^2 \pi U_o \left(\frac{3\ell^*}{4\pi\rho^{\text{in}}} \right)^{2/3} \sqrt{\frac{\eta}{2\pi k_B T^{\text{act}}}} \exp \left[-\frac{U(\ell^*)}{k_B T^{\text{act}}} \right]. \quad (\text{C.170})$$

The expected time to observe a nucleation event is given by $\tau_{\text{nuc}} = (\mathcal{R}V)^{-1}$, where V is the total volume of the system. We calculate $\mathcal{R}$ for ABP droplets and plot the results in Fig. C.3. Following straightforward manipulations, it can be shown that η is alternatively expressed as:

$$\eta = \frac{\gamma_{\text{ost}}}{2\pi(\rho^{\text{in}})^2 R_C^4}. \quad (\text{C.171})$$

Substitution of Eq. (C.171) into Eq. (C.170) results in:

$$\mathcal{R} = (\rho^{\text{sat}})^2 \pi U_o \sqrt{\frac{\gamma_{\text{ost}}}{2\pi(\rho^{\text{in}})^2 k_B T^{\text{act}}}} \exp \left[-\frac{U(\ell^*)}{k_B T^{\text{act}}} \right]. \quad (\text{C.172})$$

$\mathcal{R}$ is calculated for ABP droplets and the results are plotted in Fig. C.3. At constant ν^{sat} , while $\gamma_{\text{ost}}/k_B T^{\text{act}}$ scales roughly linearly with run length, the absolute value of ρ^{sat} also decreases. At constant ℓ_o , ρ^{sat} increases linearly with ν^{sat} . These dependencies on activity and saturation within the prefactor are far weaker than the exponential dependence of a barrier which depends strongly on both activity and saturation. Thus the qualitative behavior of Fig. C.3 is dominated by the strength of the nucleation barrier.

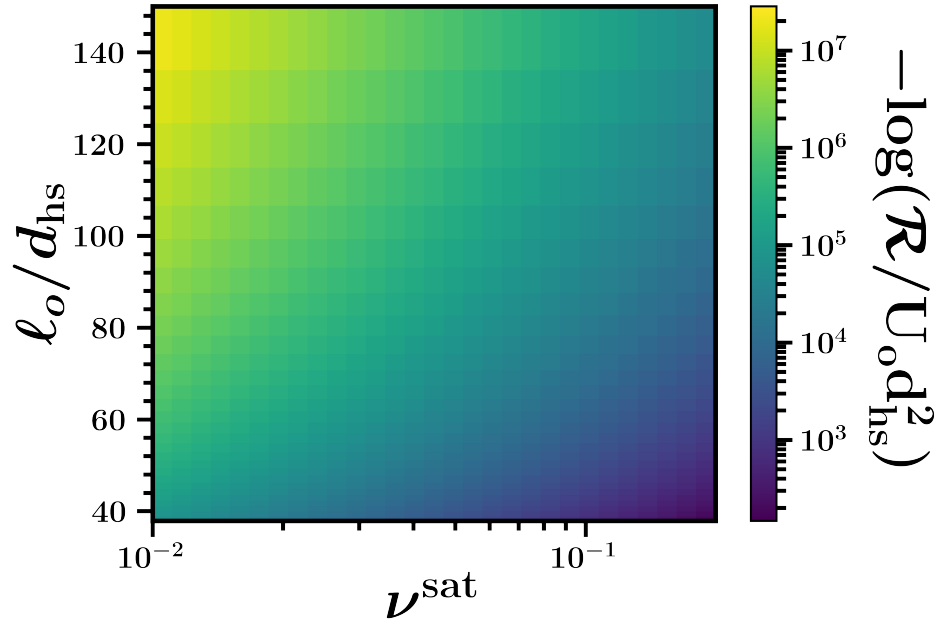


Figure C.3: Nucleation rate of droplets as a function of activity and supersaturation, as predicted by Eq. (C.170)

C.5 Thermodynamics of Classical Nucleation

We consider a nucleus of radius R and density ρ^{in} coexisting with a parent phase of density ρ^{out} . The system has an overall density of ρ^{sat} and volume V . The Helmholtz free energy of this system is then given by:

$$\mathcal{F} = \frac{4}{3}\pi R^3 f(\rho^{\text{in}}) + \left(V - \frac{4}{3}\pi R^3\right) f(\rho^{\text{out}}) + 4\pi R^2 \gamma, \quad (\text{C.173})$$

where γ is the surface tension and $f(\rho)$ is a bulk Helmholtz free energy density of a system with uniform density ρ . The total number of particles in the system is conserved with:

$$\rho^{\text{in}} \frac{4}{3}\pi R^3 + \rho^{\text{out}} \left(V - \frac{4}{3}\pi R^3\right) = \rho^{\text{sat}} V. \quad (\text{C.174})$$

Our free energy has dependencies on three adjustable internal variables, ρ^{in} , ρ^{out} , and R . The properties associated with a critical nucleus can be found by performing a constrained extremization with respect to ρ^{in} , ρ^{out} , and R and looking

for solutions where the radius is finite and not macroscopic. We construct the following Lagrangian:

$$\mathcal{W} = \mathcal{F}(\rho^{\text{in}}, \rho^{\text{out}}, R) - \lambda \left(\rho^{\text{in}} \frac{4}{3} \pi R^3 + \rho^{\text{out}} \left(V - \frac{4}{3} \pi R^3 \right) - \rho^{\text{sat}} V \right), \quad (\text{C.175})$$

where λ is a Lagrange multiplier. We extremize $\mathcal{W}$ by setting its partial derivatives with respect to ρ^{in} , ρ^{out} , λ , and R (and evaluating these variables at their unknown critical values) to zero:

$$0 = \frac{\partial \mathcal{W}}{\partial \rho^{\text{in}}} \Big|_{\rho^{\text{in}} = \rho^{\text{in}*}} = \frac{\partial f}{\partial \rho} \Big|_{\rho = \rho^{\text{in}*}} - \lambda^*, \quad (\text{C.176a})$$

$$0 = \frac{\partial \mathcal{W}}{\partial \rho^{\text{out}}} \Big|_{\rho^{\text{out}} = \rho^{\text{out}*}} = \frac{\partial f}{\partial \rho} \Big|_{\rho = \rho^{\text{out}*}} - \lambda^*, \quad (\text{C.176b})$$

$$0 = \frac{\partial \mathcal{W}}{\partial R} \Big|_{R=R_C} = f(\rho^{\text{in}}) - f(\rho^{\text{out}}) - \lambda^* (\rho^{\text{in}} - \rho^{\text{out}}) + \frac{2\gamma}{R_C}, \quad (\text{C.176c})$$

$$0 = \frac{\partial \mathcal{W}}{\partial \lambda} \Big|_{\lambda=\lambda^*} = (\rho^{\text{in}} - \rho^{\text{out}}) \frac{4}{3} \pi R_C^3 + (\rho^{\text{out}} - \rho^{\text{sat}}) V. \quad (\text{C.176d})$$

Here $\rho^{\text{in}*}$, $\rho^{\text{out}*}$, λ^* , and R_C denote the extremized values of ρ^{in} , ρ^{out} , λ , and R , respectively. Defining the chemical potential as $\mu(\rho) \equiv \partial f / \partial \rho$ allows us to appreciate that the extremized value of the Lagrange multiplier, λ^* , is simply the coexistence chemical potential with $\lambda^* \equiv \mu^{\text{coexist}} = \mu(\rho^{\text{in}*}) = \mu(\rho^{\text{out}*})$ by combining Eqs. (C.176a) and (C.176b). By defining the thermodynamic pressure, $p(\rho) \equiv -f(\rho) + \rho\mu(\rho)$, we can express the coexistence criteria with the following three equations:

$$\mu(\rho^{\text{in}*}) = \mu(\rho^{\text{out}*}), \quad (\text{C.177a})$$

$$p(\rho^{\text{in}*}) = p(\rho^{\text{out}*}) + \frac{2\gamma}{R_C}, \quad (\text{C.177b})$$

$$\rho^{\text{sat}} V = \rho^{\text{in}*} \frac{4}{3} \pi R_C^3 + \rho^{\text{out}*} \left(V - \frac{4}{3} \pi R_C^3 \right). \quad (\text{C.177c})$$

Equations (C.177) allow for the determination of the unknown densities and critical radius.

As we are interested in solutions in which the radius takes on large but non-macroscopic values, we consider the limit of $R^3 \ll V$. In this limit, the particle conservation equation [Eq. (C.174)] reduces to $\rho^{\text{out}} \approx \rho^{\text{sat}}$ and the nucleus in contact with a infinite particle reservoir with chemical potential $\mu(\rho^{\text{sat}}) \equiv \mu^{\text{sat}}$. The Lagrangian then takes the following form:

$$\mathcal{W} \approx \frac{4}{3}\pi R^3 \left(w(\rho^{\text{in}}) - w(\rho^{\text{sat}}) \right) + 4\pi R^2 \gamma, \quad (\text{C.178})$$

where $w(\rho) \equiv f(\rho) - \mu^{\text{sat}}\rho$ is the grand potential energy density (also equivalent to the negative pressure, $-p$) of a system with density ρ in contact with a particle reservoir with chemical potential μ^{sat} . We can plainly identify that the simplified Lagrangian contains two familiar terms in classical nucleation theory: a *negative volumetric term* proportional to the grand potential energy density difference between the nucleus and the parent phase and *positive areal term* proportional to the surface tension. The volumetric driving term defined in the main text [i.e., $\mathcal{W} = -\frac{4}{3}\pi R^3 \mathcal{V} + 4\pi R^2 \gamma$] has several equivalent forms $\mathcal{V} \equiv -\Delta w \equiv \Delta p \equiv -\Delta f + \mu^{\text{sat}}\Delta\rho$ where we have again used the notation of $\Delta a = a(\rho^{\text{in}}) - a(\rho^{\text{sat}})$. Extremization of $\mathcal{W}$ with respect to R and ρ^{in} results in the following coexistence criteria:

$$\mu(\rho^{\text{in}*}) = \mu^{\text{sat}}, \quad (\text{C.179a})$$

$$p(\rho^{\text{in}*}) = p(\rho^{\text{sat}}) + \frac{2\gamma}{R_C}. \quad (\text{C.179b})$$

Note that these criteria could also have been obtained by directly taking the limit of $R_C \ll V$ in Eqs. (C.177). The value of the Lagrangian evaluated at the critical radius and interior density is the *nucleation barrier*, $F^\dagger \equiv \mathcal{W}(\rho^{\text{in}} = \rho^{\text{in}*}, R = R_C)$. Finally, Eqs. (C.179) can be trivially rearranged to isolate R_C with:

$$R_C = \frac{2\gamma}{w(\rho^{\text{sat}}) - w(\rho^{\text{in}*})}. \quad (\text{C.179c})$$

We note that in the main text and for most of this SI we denote $\rho^{\text{in}*}$ simply as ρ^{in} for notational convenience. The stability of the solutions to Eqs. (C.179) can be determined by computing the Hessian matrix of $\mathcal{W}$ and evaluating it at the solutions. A positive surface energy and volumetric driving force for growth will generally result in the critical nucleus representing a unstable fixed point.

We now reformulate the above thermodynamic perspective to a microscopically grounded mechanical perspective. We consider particles which follow an overdamped Langevin equation with an additional translational Brownian stochastic noise that satisfies the fluctuation-dissipation theorem. The overdamped Langevin equation for the i th particle of this system is:

$$\dot{\mathbf{r}}_i = \frac{1}{\zeta} \sum_{j \neq i}^N \mathbf{F}_{ij} + \boldsymbol{\eta}_i(t), \quad (\text{C.180})$$

where $\boldsymbol{\eta}_i$ is the translational Brownian noise with zero average and variance $\langle \boldsymbol{\eta}_i(t) \boldsymbol{\eta}_j(t') \rangle = 2(k_B T / \zeta) \delta(t - t') \delta_{ij} \mathbf{I}$. For this system to be passive, we require that the interparticle forces $\mathbf{F}_{ij}$ are reciprocal, i.e. $\mathbf{F}_{ij} = -\mathbf{F}_{ji}$. We assume that the interparticle forces, overall system density, and temperature are such that the system is phase separated. Following the Irving-Kirkwood-Noll [120, 122] procedure for a system described by Eq. (C.180) results in the following density field dynamics at the mean-field level:

$$\frac{\partial \rho}{\partial t} = -\nabla \cdot \mathbf{J}, \quad (\text{C.181a})$$

$$\mathbf{J} = \frac{1}{\zeta} \nabla \cdot (\boldsymbol{\sigma}^C + \boldsymbol{\sigma}^B), \quad (\text{C.181b})$$

$$\boldsymbol{\sigma}^C = \left[-p_C(\rho) + \rho \kappa(\rho) \nabla^2 \rho + \frac{\left(\kappa(\rho) + \rho \frac{\partial \kappa}{\partial \rho} \right)}{2} |\nabla \rho|^2 \right] \mathbf{I} - \kappa(\rho) \nabla \rho \nabla \rho, \quad (\text{C.181c})$$

$$\boldsymbol{\sigma}^B = -k_B T \rho \mathbf{I}, \quad (\text{C.181d})$$

where $\boldsymbol{\sigma}^B$ is the “ideal gas” stress generated by the stochastic translational Brownian noise (absent in our treatment of athermal active matter) and we define $p(\rho) =$

$p_C(\rho) + k_B T \rho$. We also note that in this model of equilibrium dynamics the stress tensor is fully local.

Within these passive dynamics we may identify $\kappa_1(\rho) = \rho\kappa(\rho)$, $\kappa_2(\rho) = 1/2 (\rho\partial\kappa/\partial\rho + \kappa(\rho))$, $\kappa_3(\rho) = -\kappa(\rho)$, and $\kappa_4(\rho) = 0$. This result was first identified by Korteweg [57], and these relations have been demonstrated as equivalent to assuming variational dynamics of some Ginzburg-Landau free energy functional [162]. In this case Eq. (C.17) reduces to:

$$\frac{\partial^2 \mathcal{E}}{\partial \rho^2} = -\frac{1}{\rho} \frac{\partial \mathcal{E}}{\partial \rho} \quad (\text{C.182})$$

In this case, (within inconsequential constants of integration,) the identity of the pseudovariation becomes $\mathcal{E} \sim 1/\rho = v$, the molecular volume. By substituting Eq. (C.181) into Eq. (C.9), we may simplify the mechanical surface tension to:

$$\gamma_{\text{mech}} = \int_0^\infty dr \mathcal{G}(\rho) = \int_0^\infty dr \kappa(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2. \quad (\text{C.183})$$

By substituting $\mathcal{E} = 1/\rho$ into Eq. (C.23), we may simplify the Ostwald tension to:

$$\begin{aligned} \gamma_{\text{ost}} &= \frac{v^{\text{sat}} (\rho^{\text{sat}} - \rho^{\text{in}})}{(v^{\text{sat}} - v^{\text{in}})} \left[\int_0^\infty \left(-\rho\kappa(\rho) \frac{1}{\rho^2} \left(\frac{\partial \rho}{\partial r} \right)^2 + \kappa(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 \left(\frac{1}{\rho} - v^{\text{in}} \right) \right) dr \right], \\ &= -\frac{v^{\text{sat}} v^{\text{in}} (\rho^{\text{sat}} - \rho^{\text{in}})}{(v^{\text{sat}} - v^{\text{in}})} \left[\int_0^\infty \kappa(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr \right], \\ &= -\frac{(v^{\text{in}} \rho^{\text{sat}} - 1)}{(1 - \rho^{\text{sat}} v^{\text{in}})} \left[\int_0^\infty \kappa(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr \right], \\ &= \int_0^\infty \kappa(\rho) \left(\frac{\partial \rho}{\partial r} \right)^2 dr = \gamma_{\text{mech}}, \end{aligned} \quad (\text{C.184})$$

where we have used $v = 1/\rho$. Therefore the equilibrium Ostwald tension is identical to the equilibrium mechanical surface tension.

We now discuss the equilibrium relationship between $\mathcal{E}$ and the AMB+ pseudovariable ψ in order to avoid any potential confusion. As discussed in Appendix C.2, AMB+ can be mapped to the divergence of some dynamic stress tensor Σ , with the mapping dependent on the choice of mobility $M[\rho]$. Choosing $M[\rho] = \rho$ and setting the nonequilibrium AMB+ constants $(\lambda, \mathcal{Z})$ will result in a Σ that exactly recovers the Korteweg stress tensor from Eq. (C.181). This mapping also results in $\psi \sim \rho$. One may also deduce that $\psi \sim \rho$ after noting that the choice of $M[\rho] = \rho$ is an instance of the $M[\rho] \sim 1/\mathcal{E}$ case already discussed in detail.

The above mechanical and thermodynamic perspectives are in fact equivalent. We now demonstrate that the mechanically-derived generalized critical nucleus criteria [Eqs. (C.25) and (C.24)] applied to passive systems recover the criteria obtained through the variational method described above. The pseudovariable variable for passive systems [21, 71] is simply $\mathcal{E} \sim v$ where $v = 1/\rho$ is the specific volume and the equivalence of the Ostwald and mechanical tensions $\gamma_{\text{ost}} = \gamma_{\text{mech}} \equiv \gamma$ results in the generalized criteria now taking the following form for passive systems:

$$p(\rho^{\text{in}}) - p(\rho^{\text{sat}}) = \frac{2\gamma}{R_C}, \quad (\text{C.185a})$$

$$\int_{v^{\text{in}}}^{v^{\text{sat}}} [p(v) - p(v^{\text{sat}})] dv + \frac{2\gamma v^{\text{in}}}{R_C} = 0, \quad (\text{C.185b})$$

where we have also recognized that the dynamic pressure is simply the static thermodynamic pressure. Eq. (C.185a) is the familiar Laplace pressure difference criterion that was recovered above using thermodynamics. All that is required to establish the equivalence of the criteria obtained from our mechanical approach [Eqs. (C.185)] and the thermodynamically obtained criteria [Eqs. (C.179)] is to demonstrate the equivalence of Eq. (C.185b) with equality of chemical potential. We first eliminate the surface tension from Eq. (C.185b) by using the Laplace pressure difference, resulting in:

$$\int_{v^{\text{in}}}^{v^{\text{sat}}} p(v) dv - \left(p(v^{\text{sat}})v^{\text{sat}} - p(v^{\text{in}})v^{\text{in}} \right) = 0. \quad (\text{C.186})$$

Integration by parts demonstrates that the left-hand-side of the above equation is simply $\int_{p^{\text{in}}}^{p^{\text{sat}}} v dp$, which is equivalent to $\int_{\mu^{\text{in}}}^{\mu^{\text{sat}}} d\mu$ via the Gibbs-Duhem relation, $v dp = d\mu$. We thus arrive at the expected:

$$\mu(\rho^{\text{in}}) = \mu^{\text{sat}}, \quad (\text{C.187})$$

demonstrating the equivalence of our mechanically derived criteria with those derived from thermodynamic considerations.

In Appendix C.3 we identified a general expression for the *volumetric* contribution to the effective nucleation barrier:

$$\mathcal{V} = \frac{\mathcal{P}(\rho^{\text{sat}})\Delta\rho}{\rho^{\text{sat}}} - \frac{\Delta\Phi\Delta\rho}{\rho^{\text{sat}}\Delta\mathcal{E}}. \quad (\text{C.188})$$

With $\Phi = \int p d\mathcal{E}$ and again noting $\gamma_{\text{ost}} = \gamma_{\text{mech}} \equiv \gamma$ and $\mathcal{E} \sim v$, one can straightforwardly show:

$$\mathcal{V} = \mu^{\text{sat}}\Delta\rho - \Delta f = -\Delta w. \quad (\text{C.189})$$

Therefore the mechanical theory recovers the expected thermodynamic driving force for nucleation found in Eq. (C.178).

We now discuss an example of a system where classical nucleation theory will apply. Let us consider an equilibrium system where $\kappa(\rho)$ is a constant and $p(\rho)$ is given by the van der Waals equation of state:

$$(p + \rho^2 a) \left(\frac{1}{\rho} - b \right) = k_B T, \quad (\text{C.190})$$

where a is a measure of the average attraction between molecules and b is the volume excluded by a single particle. The critical point of this system is given by:

$$p^C = \frac{a}{27b^2}, \quad k_B T^C = \frac{8a}{27b}, \quad \rho^C = \frac{1}{3b}. \quad (\text{C.191})$$

We now non-dimensionalize variables by their value at the critical point:

$$p' = \frac{p}{p^C}, \quad T' = \frac{T}{T^C}, \quad \rho' = \frac{\rho}{\rho^C}. \quad (\text{C.192})$$

Then the dimensionless van der Waals equation of state is given by:

$$p' = \frac{8T'}{\frac{3}{\rho'} - 1} - 3(\rho')^2. \quad (\text{C.193})$$

Equation (C.192) results in the following dimensionless form of κ :

$$\kappa' = \kappa \frac{(\rho^C)^{8/3}}{p^C} \quad (\text{C.194})$$

We note that this non-dimensionalized form of the van der Waals equation of state leads to two different natural energy scales: $k_B T^C$ and p^C/ρ^C . When calculating the mechanical surface tension of a phase separated Van der Waals fluid via Eq. (C.9), our above non-dimensionalization results in the latter energy scale. In order to convert it to have units of $k_B T^C$ one can simply multiply the final result by a factor of 3/8.

We now set $\kappa' = 1$ and calculate the coexisting density and radius of the critical nuclei for van der Waals fluids. This calculation was carried out similarly to those done for the active nuclei, only now with the van der Waals equation of state. The results of this calculation are plotted in Fig. C.4. In addition, we may calculate the nucleation barrier as a function of saturation and temperature using Eq. (C.140), noting that $\gamma_{\text{ost}} = \gamma_{\text{mech}}$ for this passive system. The results of this calculation are plotted in Fig. C.5.

C.6 Active Coarsening Numerics

In this Section we demonstrate Eqs. (C.95), (C.24) and (C.25) may be applied to the late-stage coarsening dynamics of Active Brownian Particles. We consider the growth of many nuclei in a supersaturated solution. However, we will also assume that the saturation is sufficiently small that the overall volume fraction of the final phase is effectively zero. In this limit the nuclei will evolve independently of one another, and we may use the single nucleus Langevin equation [Eq. (C.95)] as the

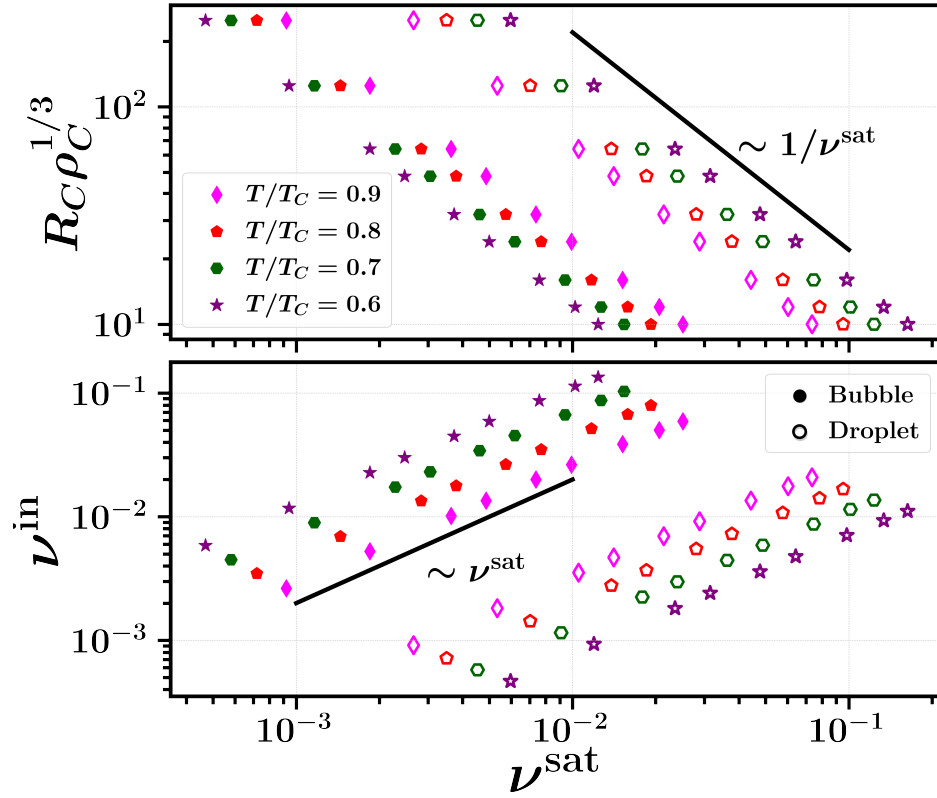


Figure C.4: Critical radius and relative change in nucleus density (ν^{in}) as a function of supersaturation (ν^{sat}).

dynamics for all nuclei. For convenience, we will rewrite Eq. (C.95) as:

$$\frac{\partial R}{\partial t} = \frac{1}{\zeta_{\text{eff}}} F^D(R) + g(R)\eta(t), \quad (\text{C.195})$$

where $\eta(t)$ is a unit Gaussian white noise with zero mean and variance $\delta(t - t')$ and $g(R)$ is defined as:

$$g(R) = \sqrt{\frac{2k_B T^{\text{act}} \rho^{\text{sat}}}{4\pi \zeta \Delta \rho^2 R^3}}. \quad (\text{C.196})$$

and $F^D(R)$ remains defined as:

$$F^D(R) = 8\pi R^2 \gamma_{\text{ost}} \left[\frac{1}{R_C} - \frac{1}{R} \right]. \quad (\text{C.197})$$

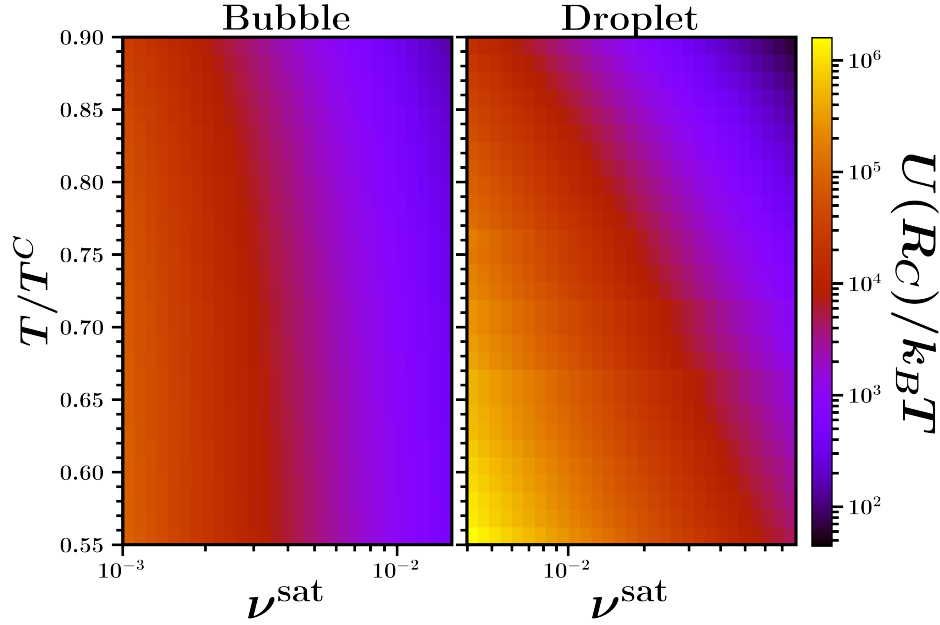


Figure C.5: Barrier $U(R_C)$ relative to thermal energy as a function of temperature and supersaturation.

Although Eq. (C.95) contains multiplicative noise, the choice of time discretization scheme (e.g. Itô, Stratonovich, etc.) is inconsequential as the drift term [125] $\frac{1}{2}g(R)g'(R)$ is $O(R^{-4})$. We now wish to solve for how the distribution of particle sizes evolves with time. This can be accomplished by first determining the Fokker-Planck equation associated with Eq. (C.195):

$$\frac{\partial P}{\partial t} = -\frac{\partial}{\partial R} \left[\frac{1}{\zeta_{\text{eff}}} F^D(R) P(R) + \frac{1}{2} \frac{\partial}{\partial R} \left(g^2(R) P(R) \right) \right]. \quad (\text{C.198})$$

From Eq. (C.196), we can see that the diffusive term will be negligible in comparison to the drift term for large R , resulting in:

$$\frac{\partial P}{\partial t} = -\frac{\partial}{\partial R} \left[\frac{1}{\zeta_{\text{eff}}} F^D(R) P(R; t) \right]. \quad (\text{C.199})$$

We can now proceed in the same fashion as Lifshitz and Slyozov [33], where it was assumed that, at long times, the asymptotic form of the distribution takes the

following form:

$$P(R; t) = \frac{1}{R_C(t)} \mathcal{N}(x), \quad (\text{C.200})$$

where we have defined the dimensionless radius $x \equiv R/R_C$ and $\mathcal{N}(x)$ is the dimensionless distribution of radii scaled by R_C . The time-dependence of $P(R; t)$ comes from that of the critical radius. Substitution of Eq. (C.200) into Eq. (C.199) results in:

$$\frac{1}{R_C^2} \frac{\partial R_C}{\partial t} \left[\mathcal{N}(x) + x \frac{d\mathcal{N}}{dx} \right] = \frac{2\rho^{\text{sat}}\gamma_{\text{ost}}}{\zeta R_C^4 \Delta\rho^2} \left[\frac{d\mathcal{N}}{dx} (x^{-1} - x^{-2}) + \mathcal{N}(x) (2x^{-3} - x^{-2}) \right] \quad (\text{C.201})$$

In order for the ansatz Eq. (C.200) to be consistent with Eq. (C.201) (i.e., $\mathcal{N}(x)$ *only depends on* x), we require:

$$R_C^2 \frac{\partial R_C}{\partial t} = \frac{2\rho^{\text{sat}}}{\zeta \Delta\rho^2} \gamma_{\text{ost}} C, \quad (\text{C.202})$$

where C is some constant. This implies that the critical radius evolves in time according to:

$$R_C(t) = \left[R_C^3(t_o) + \frac{6\rho^{\text{sat}}}{\zeta \Delta\rho^2} \gamma_{\text{ost}} C t \right]^{1/3}. \quad (\text{C.203})$$

By equating the bracketed terms in Eq. (C.201) (within a dimensionless constant) one obtains a differential equation for $\mathcal{N}(x)$ which one can solve to obtain the asymptotic LSW distribution for nucleus size [33]. Here, we are simply interested in how the average nucleus size evolves in time. Straightforward insertion of Eq. (C.200) into $\langle R \rangle = \int_0^\infty P(R; t) R dR$ reveals that the average radial dynamics are identical to those of R_C [Eq. (C.203)] within a multiplicative dimensionless constant.

We now consider a system of phase separated active particles where the nucleation of minority phase has completed and the final phase composition has been reached. In general, the system may contain many domains of minority phase. The overall volume fraction of this system will be finite and thus nuclei may interact with one another. When $\gamma_{\text{ost}} > 0$, we expect the forward Ostwald ripening

process to occur and large domains of minority phase to grow at the expense of small domains until the system reaches a final steady state with a single domain. Rather than fixing the system to be finite size and considering the influence of boundaries, we consider an infinite tiling of periodic cells. Due to the large system sizes and long timescales necessary to probe the dynamics of this process which evolves towards larger and larger domains, direct integration of Eq. (A.34) may be impractical. The problem may be greatly simplified by noting that majority phase will relax on timescales much faster than the domains of minority phase grow or shrink. Ignoring stochastic contributions, the density field dynamics (A.34) take the form:

$$\frac{\partial \rho}{\partial t} = -\frac{1}{\zeta} \nabla \cdot \nabla \cdot \Sigma. \quad (\text{C.204})$$

By restricting our perspective to *only describe the majority phase*, we may drop the time dependence from Eq. (C.204) by taking the quasi-static limit. From the perspective of the majority phase, the particles injected/adsorbed from the nuclei can be represented with finite-flux (and moving) boundary conditions at the interface between nuclei and the parent phase. Here, we take another approach and approximate majority phase particle addition/removal through spatially varying volumetric *sources and sinks*. This approach results in the quasi-static continuity equation now taking the following form:

$$0 = -\nabla \cdot \nabla \cdot \Sigma + \zeta \sum_{u,v,w} \sum_{i=1}^N S_i(\mathbf{r}), \quad (\text{C.205})$$

where we gain the source or sink term S_i in lieu of explicit consideration of the i th minority domain. Here the sum over (u, v, w) is a summation over all periodic translations of the cell. Additionally, coarse-graining the effect of the minority phase on the density field into sources/sinks renders the interface between the nuclei and parent phase beyond our level of description. We may therefore disregard the higher order gradient terms from the dynamic stress tensor resulting in:

$$0 = \nabla^2 \mathcal{P}(\mathbf{r}) + \zeta \sum_{u,v,w} \sum_{i=1}^N S_i(\mathbf{r}). \quad (\text{C.206})$$

Globally, the number of particles must remain conserved, imposing the following constraint on S_i :

$$\sum_{i=1}^N \int_V S_i(\mathbf{r}) d\mathbf{r} = 0. \quad (\text{C.207})$$

From Eq. (C.206), a mathematical analogue between the pressure field and the electrostatic potential due to N charge densities may be drawn. By approximating the minority domains as spherically symmetric we may safely collapse the “charge densities” S_i into point charges at their centers by defining $S_i(\mathbf{r}) \equiv 2\pi B_i \delta(\mathbf{r} - \mathbf{r}_i) / \zeta$. Equation (C.206) can now be expressed as:

$$0 = \nabla^2 \mathcal{P}(\mathbf{r}) + 4\pi \sum_{u,v,w} \sum_{i=1}^N B_i \delta(\mathbf{r} - \mathbf{r}_i - u\mathbf{a} - v\mathbf{b} - w\mathbf{c}), \quad (\text{C.208})$$

where $\mathbf{a}$, $\mathbf{b}$, $\mathbf{c}$ are the lattice vectors and the particle conservation constraint [Eq. (C.207)] becomes $\sum B_i = 0$. The solution to Eq. (C.208) is well-known and given by:

$$\mathcal{P}(\mathbf{r}) = \mathcal{P}_o + \sum_{u,v,w} \sum_{i=1}^N \frac{B_i}{|\mathbf{r} - \mathbf{r}_i - u\mathbf{a} - v\mathbf{b} - w\mathbf{c}|}, \quad (\text{C.209})$$

where $\mathcal{P}_o$ is the unknown pressure of the majority phase far from the nuclei. We now assume that every individual nucleus is in mechanical equilibrium with its immediate surroundings. As outlined in Section C.1, Eqs. (C.24) and (C.25) allow us to solve for the coexistence densities of a finite sized nucleus with radius R . We define $\rho^R(R)$ as a function mapping the size of a nucleus with size R to the *outside* coexistence density. Then, if $\mathbf{r}_j^s$ is a point on the surface of a nucleus of size R_j , we have:

$$\mathcal{P}(\mathbf{r}_j^s) = \mathcal{P}(\rho^R(R_j)) \quad (\text{C.210})$$

Substitution of the boundary condition [Eq. (C.210)] into Eq. (C.209) gives:

$$\mathcal{P}(\mathbf{r}_j^s) = \mathcal{P}(\rho^R(R_j)) = \mathcal{P}_o + \frac{B_j}{R_j} + \sum_{u,v,w} \sum_{i=1}^N \frac{B_i}{|\mathbf{r}_j^s - \mathbf{r}_i - u\mathbf{a} - v\mathbf{b} - w\mathbf{c}|}, \quad (\text{C.211})$$

where the sum ignores the $i = j$ term when $u, v, w = 0$. A key subtlety of Eq. (C.211) is that $\mathbf{r}_j^s$ is a point on the *surface* of nucleus j while $\mathbf{r}_i$ is the *center* of nucleus i . By expanding the summation in terms of Legendre polynomials and averaging over all possible choices of $\mathbf{r}_j^s$ on the surface of the j th nucleus, one may show that

$$\mathcal{P}(\rho^R(R_j)) = \mathcal{P}_o + \frac{B_j}{R_j} + \sum_{u,v,w} \sum_{i=1}^N \frac{B_i}{|\mathbf{r}_j - \mathbf{r}_i - u\mathbf{a} - v\mathbf{b} - w\mathbf{c}|}, \quad (\text{C.212})$$

where $\mathbf{r}_j$ is the center of nucleus j . Evaluation of the summation over an infinite periodic lattice can be accomplished via standard techniques of Ewald summation, although with the added complication that the sum ignores the $i = j$ case when $u, v, w = 0$, resulting in:

$$\begin{aligned} \mathcal{P}(\rho^R(R_j)) &= \mathcal{P}(\rho^{\text{mat}}) + B_j \left(\frac{1}{R_j} - \sqrt{\frac{1}{\eta\pi}} \right) \\ &+ \frac{4\pi}{\mathbf{a} \cdot (\mathbf{b} \times \mathbf{c})} \sum_{\mathbf{k} \neq \mathbf{0}} \sum_{i=1}^N \frac{B_i}{|\mathbf{k}|^2} \exp \left[-\eta |\mathbf{k}|^2 - i\mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_i) \right] \\ &+ \sum_{u,v,w} \sum_{i=1}^N \frac{B_i}{|\mathbf{r}_j - \mathbf{r}_i - u\mathbf{a} - v\mathbf{b} - w\mathbf{c}|} \operatorname{erfc} \left(\frac{|\mathbf{r}_j - \mathbf{r}_i - u\mathbf{a} - v\mathbf{b} - w\mathbf{c}|}{2\sqrt{\eta}} \right), \end{aligned} \quad (\text{C.214})$$

where $\mathbf{k}$ are the reciprocal lattice vectors corresponding to our periodic cell and η is free parameter that controls how quickly the summation converges in real space as opposed to reciprocal space. Equation (C.214) can be expressed as a matrix equation $\mathbf{b} = \mathbf{A} \cdot \mathbf{x}$, where

$$\mathbf{b} = \begin{bmatrix} \mathcal{P}(\rho^R(R_1)) \\ \mathcal{P}(\rho^R(R_2)) \\ \vdots \\ \mathcal{P}(\rho^R(R_N)) \\ 0 \end{bmatrix}, \quad (\text{C.215a})$$

$$\mathbf{x} = \begin{bmatrix} B_1 \\ B_2 \\ \vdots \\ B_N \\ \mathcal{P}(\rho^{\text{mat}}) \end{bmatrix}, \quad (\text{C.215b})$$

and

$$\mathbf{A} = \begin{bmatrix} \frac{1}{R_1} + D & O_{12} & O_{13} & \dots & 1 \\ O_{21} & \frac{1}{R_2} + D & O_{23} & \dots & 1 \\ O_{31} & O_{32} & \frac{1}{R_3} + D & \dots & 1 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 1 & 1 & 1 & \dots & 0 \end{bmatrix}. \quad (\text{C.215c})$$

Here D is given by:

$$D = -\sqrt{\frac{1}{\eta\pi}} + \frac{4\pi}{L_x L_y L_z} \sum_{\mathbf{k} \neq 0} \frac{1}{|\mathbf{k}|^2} \exp(-\eta|\mathbf{k}|^2) \\ + \sum_{u,v,w \neq 0} \frac{1}{(|u\mathbf{a}|^2 + |v\mathbf{b}|^2 + |w\mathbf{c}|^2)^{1/2}} \text{erfc}\left(\frac{1}{2\sqrt{\eta}} \left(|u\mathbf{a}|^2 + |v\mathbf{b}|^2 + |w\mathbf{c}|^2\right)^{1/2}\right), \quad (\text{C.216})$$

and O_{ij} is given by:

$$O_{ij} = \frac{4\pi}{\mathbf{a} \cdot (\mathbf{b} \times \mathbf{c})} \sum_{\mathbf{k} \neq 0} \frac{1}{|\mathbf{k}|^2} \exp\left(-\eta|\mathbf{k}|^2 - i\mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_i)\right) \\ + \sum_{u,v,w} \frac{1}{|\mathbf{r}_j - \mathbf{r}_i - u\mathbf{a} - v\mathbf{b} - w\mathbf{c}|} \text{erfc}\left(\frac{|\mathbf{r}_j - \mathbf{r}_i - u\mathbf{a} - v\mathbf{b} - w\mathbf{c}|}{2\sqrt{\eta}}\right). \quad (\text{C.217})$$

Given the size and locations of each nucleus in the cell, one may then solve Eqs. (C.215) in order to find all B_i and the background pressure $\mathcal{P}_o$. Let's now consider the rate of change of the radius of a particular nucleus and how it translates to the nucleus source/sink strength B_i . If nucleus i changes its radius R_i by an

amount dR_i over a time period dt , then the rate of mass injected into the majority phase by particle i is given by:

$$4\pi R_i^2 \frac{dR_i}{dt} (\rho^{\text{out}} - \rho^{\text{in}}) \quad (\text{C.218})$$

By the definition of S_i , the rate of mass injected into the majority phase by nucleus i is also equal to the integral of S_i across the entire system of volume. Then we have:

$$4\pi R_i^2 \frac{dR_i}{dt} (\rho^{\text{out}} - \rho^{\text{in}}) = \int_V d\mathbf{r} S_i \quad (\text{C.219})$$

From the definition of B_i we then have:

$$4\pi R_i^2 \frac{dR_i}{dt} (\rho^{\text{out}} - \rho^{\text{in}}) = \frac{4\pi}{\zeta} B_i \quad (\text{C.220})$$

Then the rate of change of the radius of nucleus i is given by:

$$\frac{dR_i}{dt} = \frac{B_i}{\zeta R_i^2 (\rho^{\text{out}} - \rho^{\text{in}})} \quad (\text{C.221})$$

Therefore solution of the matrix equation defined by Eqs. (C.215) allows for the extraction of the coarsening rates.

Given the periodic cell dimensions, nucleus locations, and nucleus radii, it is straightforward to set up the necessary matrix equations to find the coarsening rates and numerically integrate. We conduct numerical simulation of periodic cells of 250 nuclei radii randomly drawn from a distribution. The PACKMOL software package [132] was used to give random locations within the periodic cells such that all nuclei had a separation of at least two times the largest radius. Particle sizes were sampled such that they had an average radius of $750d_{\text{hs}}$ and a cubic cell of width $a_o = 2.1 \times 10^5 d_{\text{hs}}$, corresponding to an overall volume fraction of 2×10^{-7} . For the overall phase fractions simulated, we expect the results of LSW theory [33] to be recovered. The main predictions of LSW theory are the average nucleus size growing with the cube root of time and a steady-state distribution of

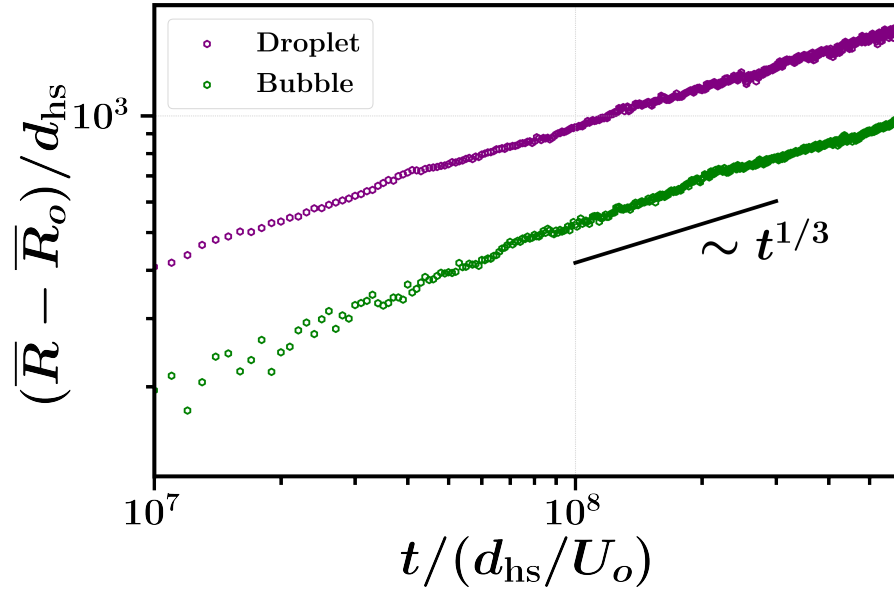


Figure C.6: Average radius growth as a function of time for nuclei initialized with a Gaussian distribution with mean $750d_{\text{hs}}$.

nucleus sizes $P(R) \propto \mathcal{N}(x)$ at long times, where x is the ratio of the nucleus radius to the average nucleus radius and $\mathcal{N}$ is proportional to:

$$\begin{cases} x^2(3+x)^{-7/3} \left(\frac{3}{2} - x\right)^{-11/3} \exp\left(-\frac{3}{3-2x}\right) & x < 3/2 \\ 0 & x \geq 3/2. \end{cases}$$

We therefore initialized all simulations with radii sampled from a Gaussian distribution as well as initial radii sampled from the distribution predicted by LSW. Simulations were integrated in time for a duration of $6 \times 10^8 d_{\text{hs}}/U_o$. Simulation results were averaged over 10 independent randomly generated initial positions and radii for both of the initial distributions. We plot the average nucleus radius cubed as a function of time in Figs. C.6 and C.7. Growth rates demonstrate excellent agreement with the scaling predicted by LSW theory. Droplets coarsen significantly faster than bubbles due to the higher γ_{ost} and therefore larger driving force for coarsening. The distribution of nucleus sizes was also measured in the final third of the simulation time (i.e. times from $4 - 6 \times 10^8$) and are given by

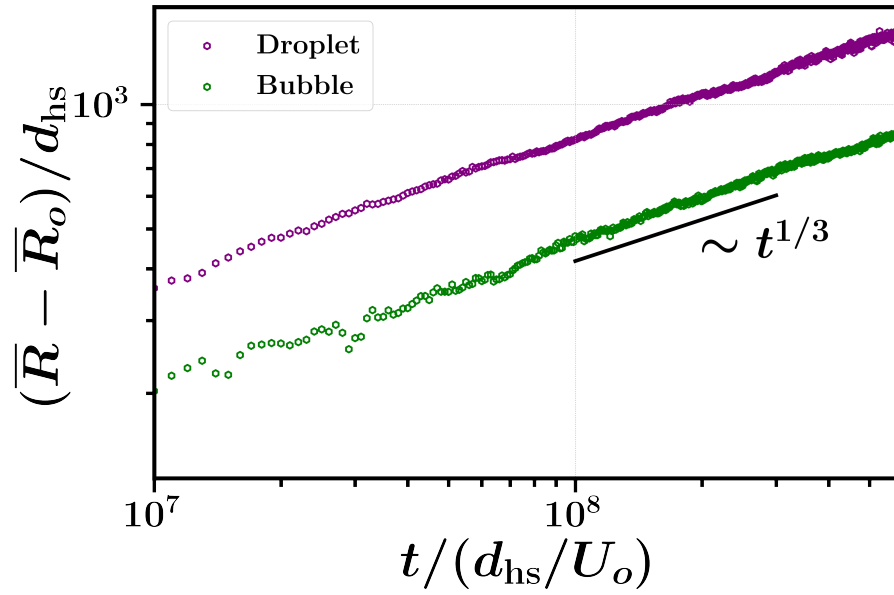


Figure C.7: Average radius growth as a function of time for nuclei initialized with an LSW distribution with mean $750d_{hs}$.

Figs. C.8 and C.9. The nuclei initialized from Gaussian distribution veer from their initial sampling significantly by the end of the simulation time while nuclei initialized from the LSW distribution stay close to the LSW distribution, although with some error due to the finite amount of sampling.

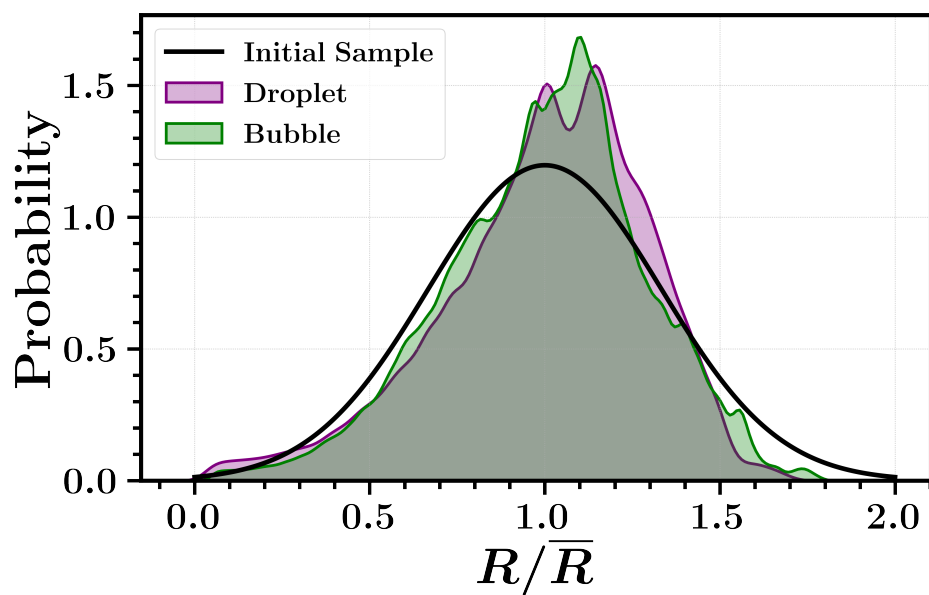


Figure C.8: Nucleus size distribution sampled from last third of simulation steps for particles initialized with Gaussian distribution.

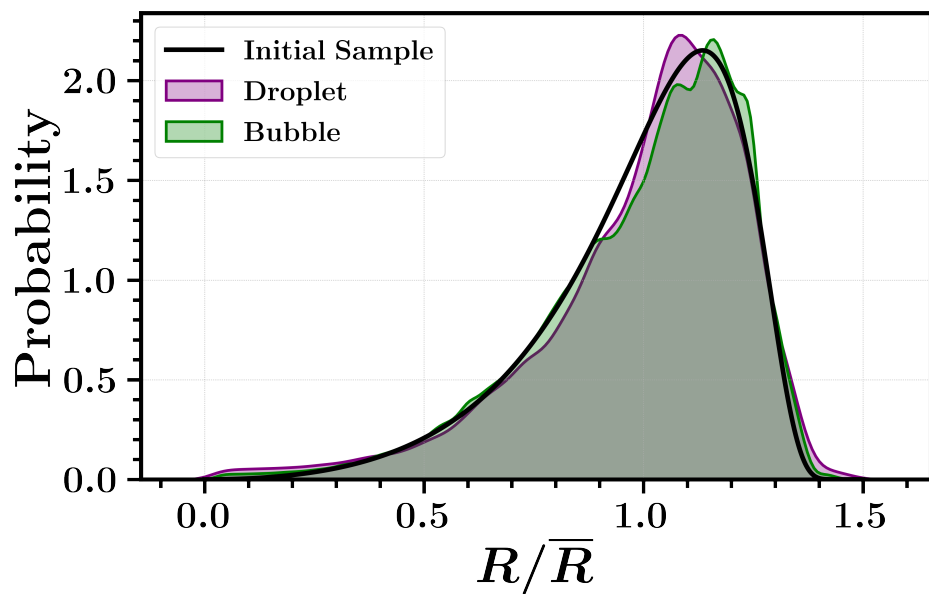


Figure C.9: Nucleus size distribution sampled from last third of simulation steps for particles initialized with LSW distribution.

Appendix D

Appendix: Thermal Noise, Hexatic Order, and Bubbles

The contents of Appendix D will closely follow that of the Supplemental Information of Ref. [139]. We will present the details of how numerical simulations and subsequent analysis were carried out, as well as several figures done to ensure the reliability of our analysis.

D.1 Simulation Details

The pairwise force between particles in Eq. (5.1) can be written as the gradient of a two-body interaction potential, i.e., $\mathbf{F}_{ij} = -\nabla V_{ij}(|\mathbf{r}_i - \mathbf{r}_j|)$, where ∇ is the gradient with respect to the vector connecting particle i and j . In all simulations we employ a radially shifted Lennard-Jones potential [161] of the form:

$$V(r_{ij})/\epsilon = \begin{cases} 4 \left[\left(\frac{\sigma}{r_{ij} - \alpha_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij} - \alpha_{ij}} \right)^6 \right] + 1, & r_{ij} \leq r_{ij}^{\text{cut}} \\ 0, & r_{ij} > r_{ij}^{\text{cut}}. \end{cases} \quad (\text{D.1})$$

where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$, ϵ is the characteristic Lennard-Jones (LJ) energy, σ is the LJ diameter, and α_{ij} is the radial shift. The potential is cut off at the minimum, ensuring purely repulsive interactions with $r_{ij}^{\text{cut}} \equiv 2^{1/6}\sigma + \alpha_{ij}$. In the absence of

a radial shift, our potential reduces to the WCA potential [124] and the cutoff distance can be interpreted as an effective hard-disk diameter (for sufficiently stiff potentials) with $r_{\text{cut}} = 2^{1/6}\sigma \equiv d$. We allow particles to differ in size through α_{ij} : a pair of particles with distinct diameters begin to feel repulsion at $r_{ij}^{\text{cut}} = \alpha_{ij} + d$ which we take to be the mean particle diameter of the pair (i.e., $d_{ij} = (d_i + d_j)/2$ where d_i is the hard-disk diameter of particle i).

For the athermal ($\mathcal{T} = 0$) simulations, particles were initialized randomly and the simulations were continued until the system reached a fully coarsened steady state with a single liquid domain and a single gas domain. In the polydisperse simulation, this was done using a square $800\sigma \times 800\sigma$ simulation box, resulting in an overall area fraction of ϕ_o of approximately 0.49. The kinetics of the monodisperse case were sluggish, and achieving a final state with a single liquid and gas domain within reasonable simulation time required using a smaller $700\sigma \times 700\sigma$ simulation box, resulting in an overall area fraction of ϕ_o of approximately 0.64. Despite the difference in overall area fraction, both athermal simulations reached qualitatively similar steady states. After reaching a steady state, statistics were then collected over a minimum time span of $5 \times 10^4 \sigma / U_o$. All thermal simulations ($\mathcal{T} > 0$) were initialized from the final simulation frame of the athermal simulations, allowed to reach a steady state over $5 \times 10^4 \sigma / U_o$, then integrated for an additional $5 \times 10^4 \sigma / U_o$ in order to collect statistics. The monodisperse simulations were conducted at thermalization coefficients of $\mathcal{T} = \{0, \frac{1}{100}, \frac{1}{27}, \frac{1}{9}, \frac{1}{3}, 1\}$ while polydisperse simulations were calculated at thermalization coefficients of $\mathcal{T} = \{0, \frac{1}{100}, \frac{1}{3}, 1\}$. In order to avoid appreciably shifting the critical point (and therefore the phase diagram, see Appendix D.2), we restrict our simulations to thermalization coefficients $\mathcal{T} \leq 1$.

D.2 Impact of Thermalization on Critical Point

In athermal ABPs, the dynamic pressure controlling the bulk coexistence criteria [21] was determined to be:

$$\mathcal{P} = p^{\text{act}}(\phi) + p^c(\phi), \quad (\text{D.2a})$$

$$a_o p^{\text{act}}(\phi) = \frac{\zeta U_o \bar{U} \ell_o \phi}{2}, \quad (\text{D.2b})$$

where ϕ is the local packing fraction, $\bar{U}$ is a dimensionless effective active speed that is between 0 and 1, and $a_o = \pi d^2/4$ is the single particle area. Here $p^c(\phi)$ is the conservative pressure due to particle collisions. Here, $d = 2^{1/6}\sigma$ corresponds to the hard particle diameter. The critical point, which represents the onset of the spinodal criteria, can be found by setting both the first and second derivative of $\mathcal{P}(\phi)$ to zero and solving for the critical packing fraction and activity (ϕ_c, ℓ_c). When translational noise is introduced, $\mathcal{P}$ is modified in a few ways. First, an *ideal gas* pressure becomes present, and is proportional to $k_B T \phi$:

$$a_o p^{\text{ig}} = \zeta D_T \phi = \frac{\mathcal{T} \sigma^2 U_o}{\ell_o} \phi, \quad (\text{D.3})$$

Additionally, the equations of state for $\bar{U}$ and $p^c(\phi)$ may change. We will assume that the equations of state do not change and remain of the form used in Refs. [18, 85]:

$$\frac{p^c d}{\zeta U_o} = \frac{8}{\pi} \phi^2 \left(1 - \frac{\phi}{\phi_o}\right)^{-1}, \quad (\text{D.4a})$$

$$\bar{U} = \left[1 + \phi \left(1 - \frac{\phi}{\phi_o}\right)^{-1}\right]^{-1}, \quad (\text{D.4b})$$

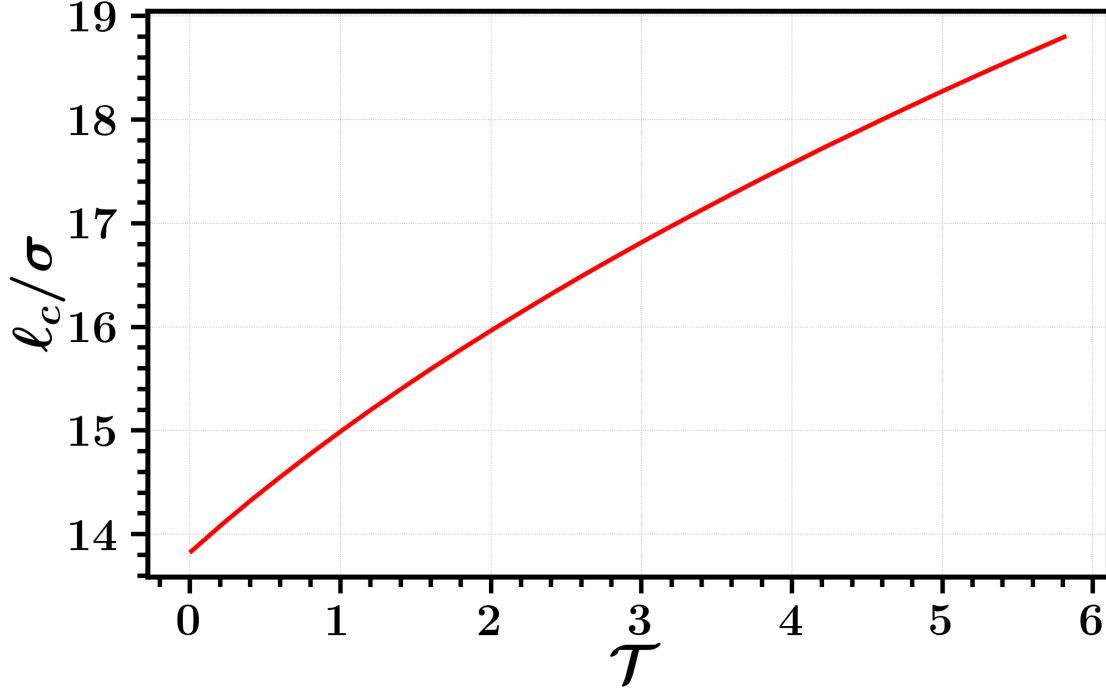


Figure D.1: Critical activity ℓ_c as a function of thermalization $\mathcal{T}$.

where $\phi_o \equiv 0.9$ approximates the close packing fraction for two dimensions. Using Eqs. (D.2), (D.3), and (D.4), we calculate ϕ_c and ℓ_c as a function of $\mathcal{T}$.

Based on the results of Figs. D.1 and D.2, we restrict our thermalization to be less than one ($\mathcal{T} < 1$) in order not to shift the critical activity by more than ten percent.

D.3 Steady State Verification

To check that our simulations have reached a steady state, we verify that measured quantities of interest indeed are time independent. In particular, we can average the measured quantities over smaller chunks of time and verify a lack of systematic drift over time in those measured quantities. We choose to do this for two quantities that were measured for all simulations: the power-law exponent ω of $n(A)$, and the bubble area fraction ϕ_B . These measurements are broken up into time

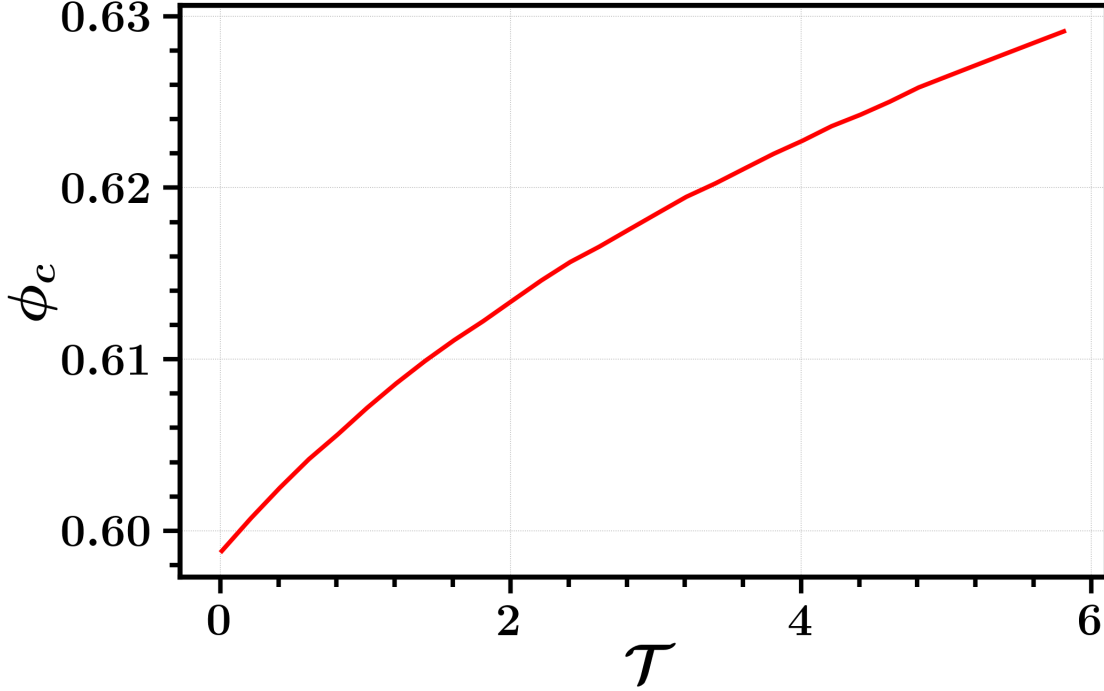


Figure D.2: Critical area fraction ϕ_c as a function of thermalization $\mathcal{T}$.

chunks of $2500 \sigma/U_o$ to average over, and plotted across the second half of the $100000 U_o/\sigma$ trajectory where statistics reported in the main text were collected. While the measured quantities could vary significantly, especially for simulations where bubbles were rarefied, no systematic drift was identified.

D.4 Analysis Details

In order to record the bubble statistics $n(A)$ and $P(A)$, an algorithm for identifying bubbles based on the method used in Ref. [35] was employed. First, the local coarse-grained area fraction $\tilde{\phi}$ calculated using the coarse-graining function:

$$\tilde{\phi}(\mathbf{r}) = \sum_{i=1}^N \pi \left(\frac{d_i}{2} \right)^2 \frac{1}{2\pi\xi^2} \exp \left[-\frac{|\mathbf{r} - \mathbf{r}_i|^2}{2\xi^2} \right], \quad (\text{D.5})$$

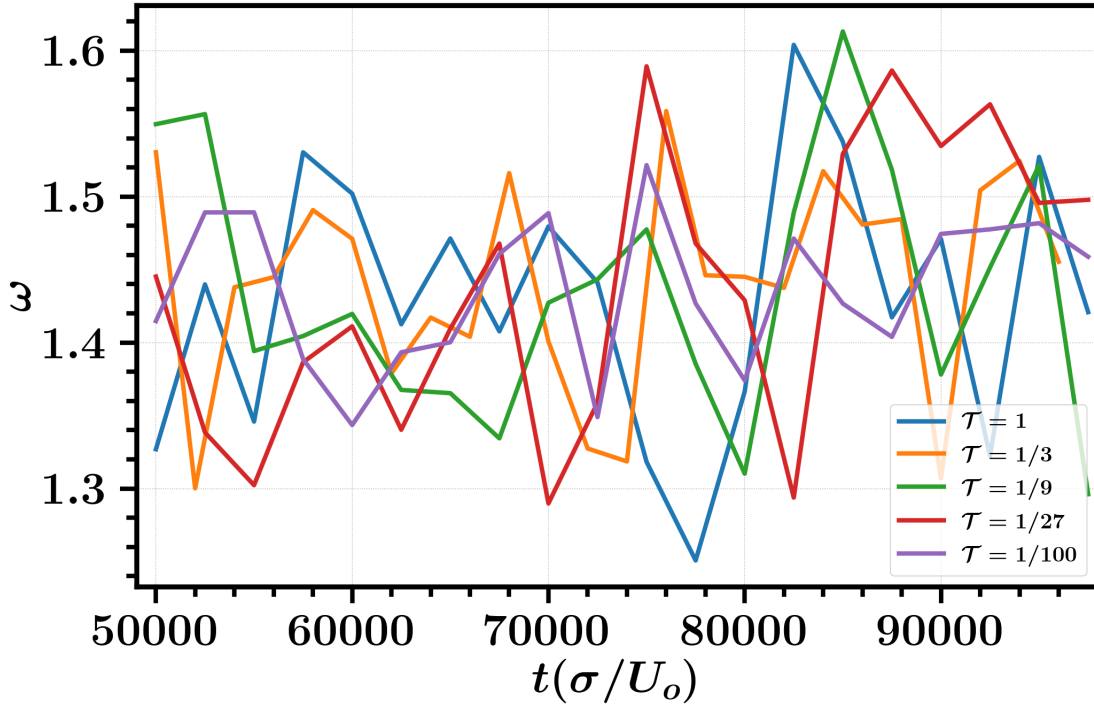


Figure D.3: Fits of bubble distribution power law exponent ω over time for thermalized monodisperse systems.

where ξ is a coarse-graining length that we set to 2.5σ for all calculations and $d_i = \sigma_i + (2^{1/6} - 1)\sigma$ is the particle diameter. $\tilde{\phi}(\mathbf{r})$ was calculated on a square grid of sample points with spacing of 3.2σ . Grid points with a value of $\tilde{\phi}$ less than some cutoff density ϕ_{cut} were labeled as gas phase. In all calculations we set $\phi_{\text{cut}} = 0.65$. The cluster analysis algorithm DBSCAN [150] was then used to identify clusters of grid points labeled as gas phase. DBSCAN uses the following rules for determining clusters:

- Two objects are considered neighbors if they are within some cutoff distance ϵ_{cut} of each other. An object is considered a “core” object if it has n_{min} neighbors.
- Clusters are defined by the connected pathways of all core objects and neighbors of core objects.

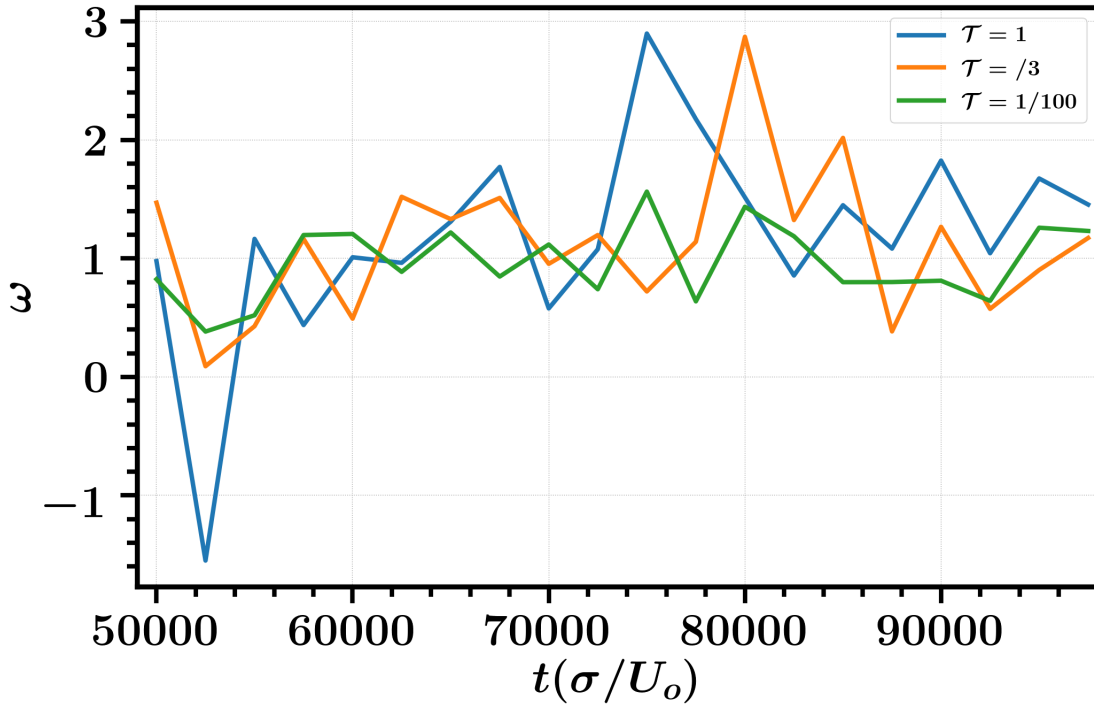


Figure D.4: Fits of bubble distribution power law exponent ω over time for thermalized polydisperse systems.

- Objects that are not core or neighbors of cores are not part of any cluster.

Bubbles were identified by using DBSCAN on all points labeled as gas phase with ϵ_{cut} set to be the grid spacing 3.2σ , and $n_{\text{min}} = 4$ corresponding to a square grid. The largest bubble/cluster identified in each frame was the bulk gas phase, which we ignore. The area of all other bubbles was calculated, and all bubbles with area larger than $A_{\text{min}} = 16\pi\sigma^2$ were added to a histogram with binwidth $\Delta A = 10\sigma^2$. $n(A)$ was calculated by normalizing this histogram by the number of frames considered and dividing by the bin width. $P(A)$ was calculated by normalizing $n(A)$ by the average number of bubbles observed, N_{bub} . Both quantities were then divided by the histogram bin spacing in order to maintain the correct units. ϕ_B was calculated by recording the total area of identified bubbles and dividing by the total area of gas phase.

We also provide $n(A)$ and $P(A)$ for all thermalized systems (not included in the

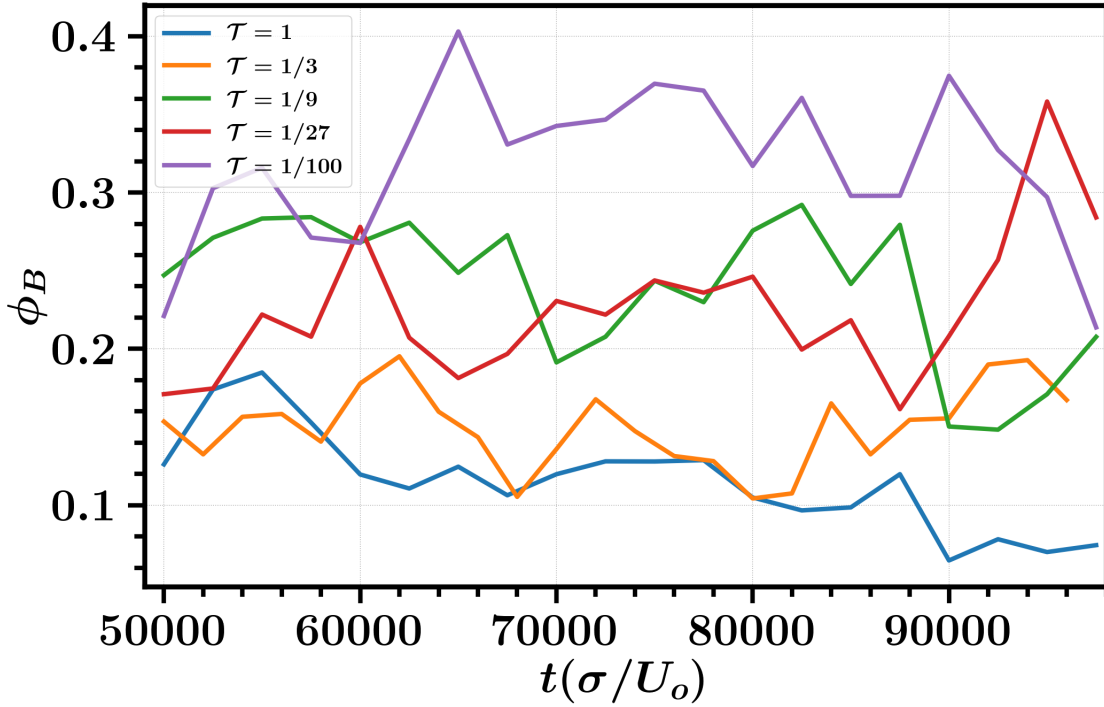


Figure D.5: Average bubble fraction ϕ_B over time for thermalized monodisperse systems.

main text) in Figs. D.8 and D.9. Additionally, we reproduce Fig. 5.2 of the main text up to higher values of A in order to display the cutoff size for the algebraic decay in Fig. D.10.

We also provide a table of the power law exponents fit to $n(A)$ as well as the area fraction of gas phase within bubbles in Table D.1

The hexatic order parameter was calculated for each particle, defined as:

$$\psi_{6j} = N_j^{-1} \sum_{k=1}^{N_j} \exp[i6\phi_{jk}], \quad (\text{D.6})$$

where N_j is the number of nearest neighbors of particle j and ϕ_{jk} is the angle between the vector connecting particles j and k with an arbitrary reference vector. The hexatic order can be easily calculated using the order module of the Freud software package [163].

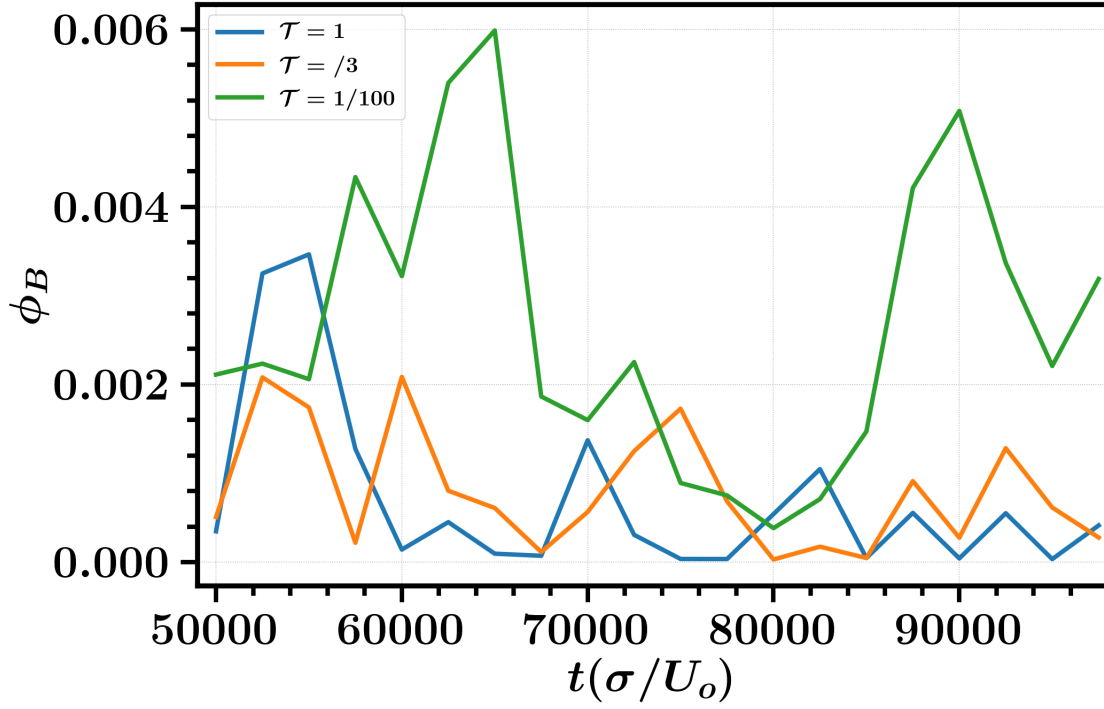


Figure D.6: Average bubble fraction ϕ_B over time for thermalized polydisperse systems.

Table D.1: Scaling exponents of $n(A)$ and area fraction of gas phase in bubbles, ϕ_B

Dispersity	$\mathcal{T}$	ω	ϕ_B
Monodisperse	1	1.44 ± 0.08	0.12 ± 0.03
Monodisperse	1/3	1.45 ± 0.07	0.15 ± 0.03
Monodisperse	1/9	1.47 ± 0.07	0.21 ± 0.05
Monodisperse	1/27	1.46 ± 0.05	0.21 ± 0.05
Monodisperse	1/100	1.44 ± 0.05	0.28 ± 0.07
Monodisperse	0	N/A	$(5.2 \pm 2.7) \times 10^{-4}$
Polydisperse	1	N/A	$(7.1 \pm 9.9) \times 10^{-4}$
Polydisperse	1/3	N/A	$(8.0 \pm 6.6) \times 10^{-4}$
Polydisperse	1/100	N/A	$(2.7 \pm 1.6) \times 10^{-3}$
Polydisperse	0	N/A	$(3.3 \pm 2.2) \times 10^{-4}$

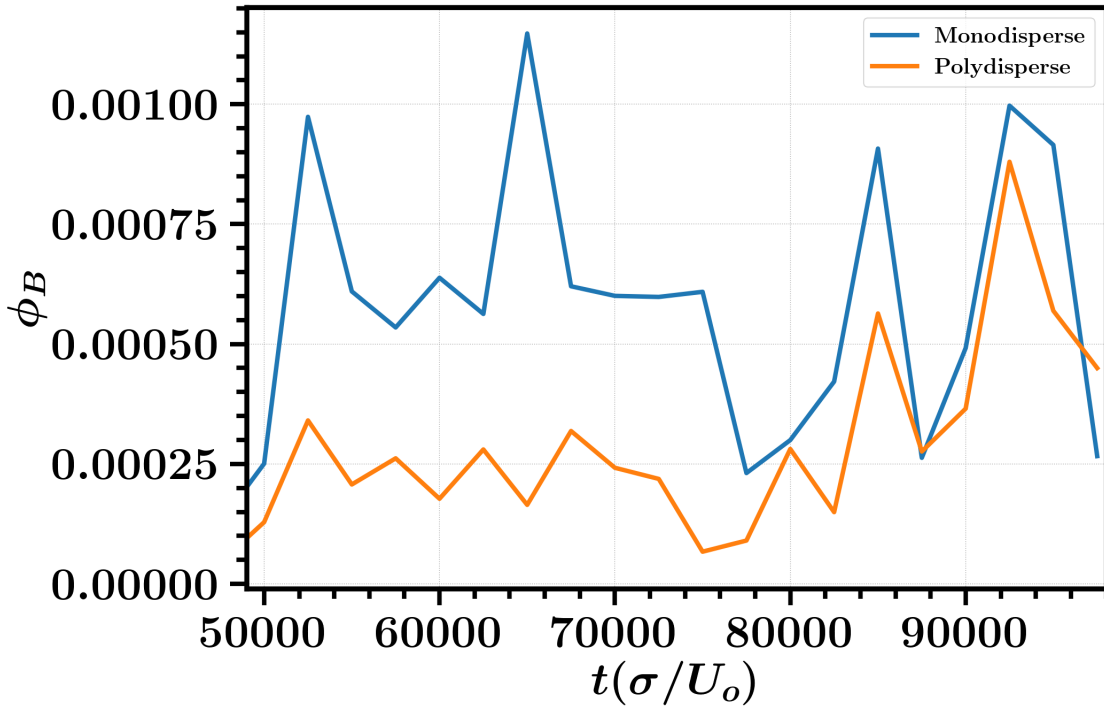


Figure D.7: Average bubble fraction ϕ_B over time for athermal systems.

Clearly the result of Eq. (D.6) is complex, which implies a phase angle. Following Ref. [35], hexatic domains were defined as clusters of particles with ψ_{6j} that had similar phase angle. Specifically, the interval $[0, 2\pi]$ was broken up into 15 evenly spaced bins and each particle was assigned a bin according to the phase angle of its ψ_{6j} . The DBSCAN algorithm outlined in the previous section was then applied while only considering particles of a particular bin with ϵ_{cut} set to 1.5σ and n_{min} set to 6. The result of this calculation is clusters of particles sharing the same bin of ψ_{6j} phase angle, which we identify as hexatic domains. The area of these domains are then calculated and added to a histogram, which is then normalized by the total number of domains identified and the bin spacing $10\sigma^2$.

The above calculation was only relevant for the monodisperse simulations which displayed a grain structure. Further, the athermal monodisperse simulation only contained a few large domains with sluggish kinetics, causing the measured distribution to be very noisy, with a few spikes corresponding to the sizes of the

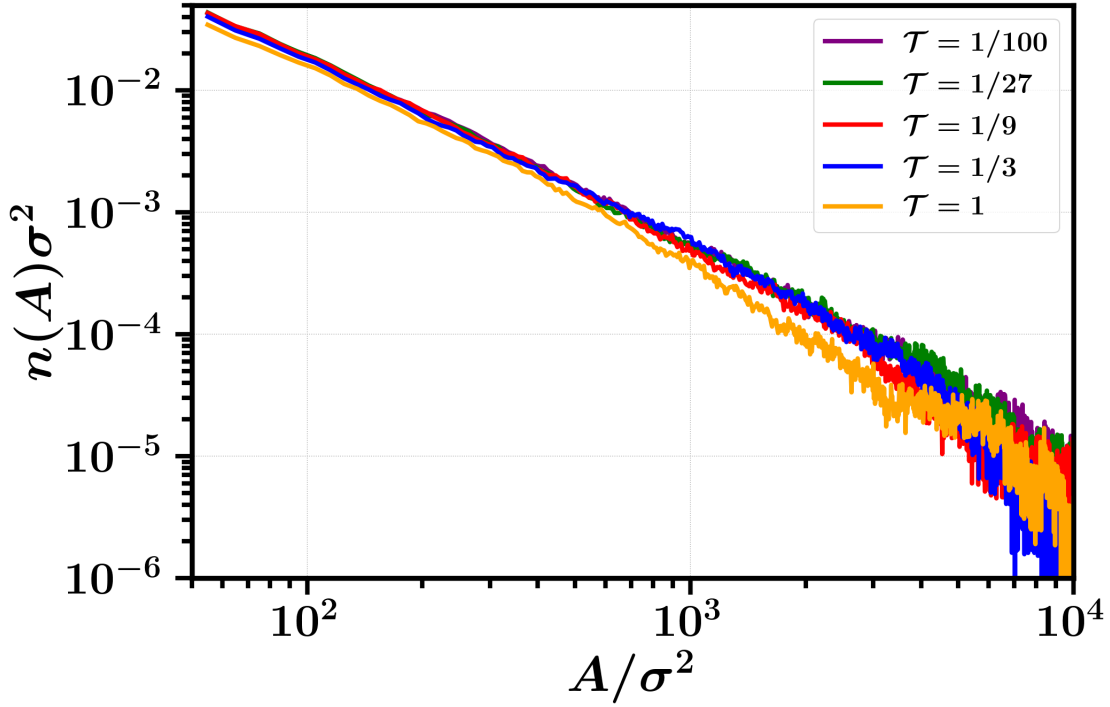
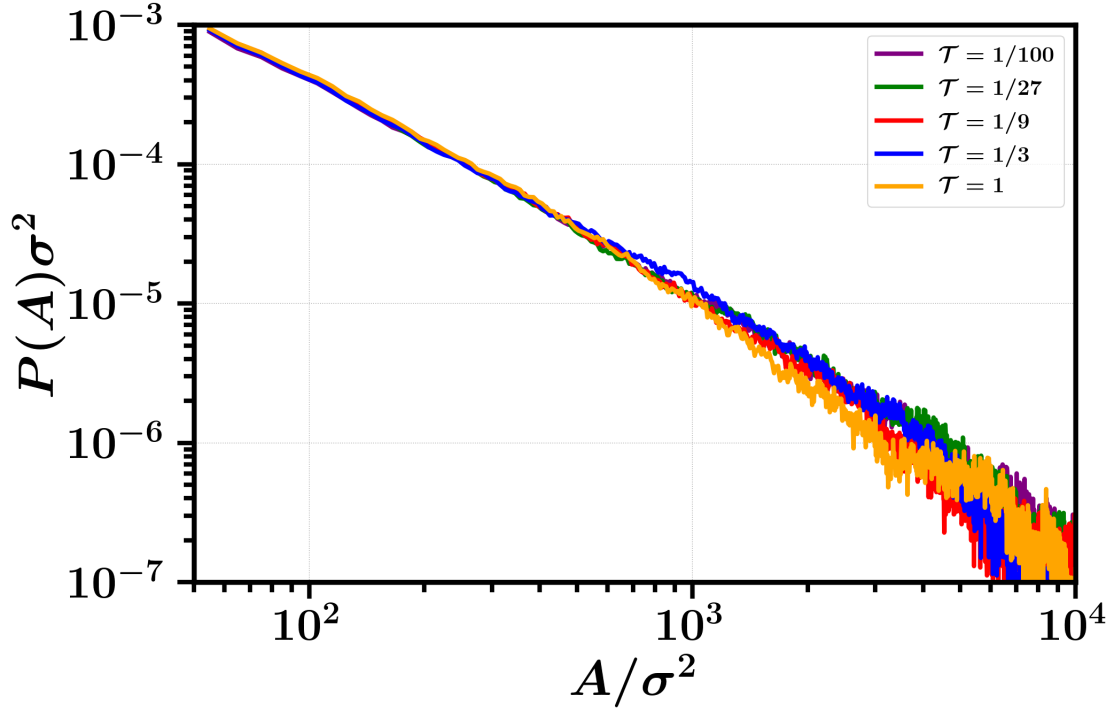


Figure D.8: $n(A)$ for all monodisperse simulated values of $\mathcal{T} > 0$.

large grains seen in Fig. 3(a) of the main text. We therefore only plot the domain size distribution for monodisperse thermal simulations in the main text. We include here the same figure but with the athermal case included.

In order to confirm the disappearance of medium-to-long ranged hexatic ordering in the polydisperse case, we also calculated the spatial correlations of the hexatic order for all simulations $\langle \psi_6(r) \psi_{6j}^*(0) \rangle$, where $\psi_6(r)$ is the average hexatic order of all particles located within some $r + \Delta r$ of particle j , and the angled brackets indicate an average over all particles in the dense phase. It should be noted that this quantity is not translationally invariant as the notation r might suggest: particles closer to the interface will clearly have different hexatic order than those deep in the bulk of the dense phase. However, in the polydisperse case where non-negligible correlations only persist for a few particle diameters, these differences minimally impact the calculation's results.

We also provide the hexatic correlation function for all thermalized systems

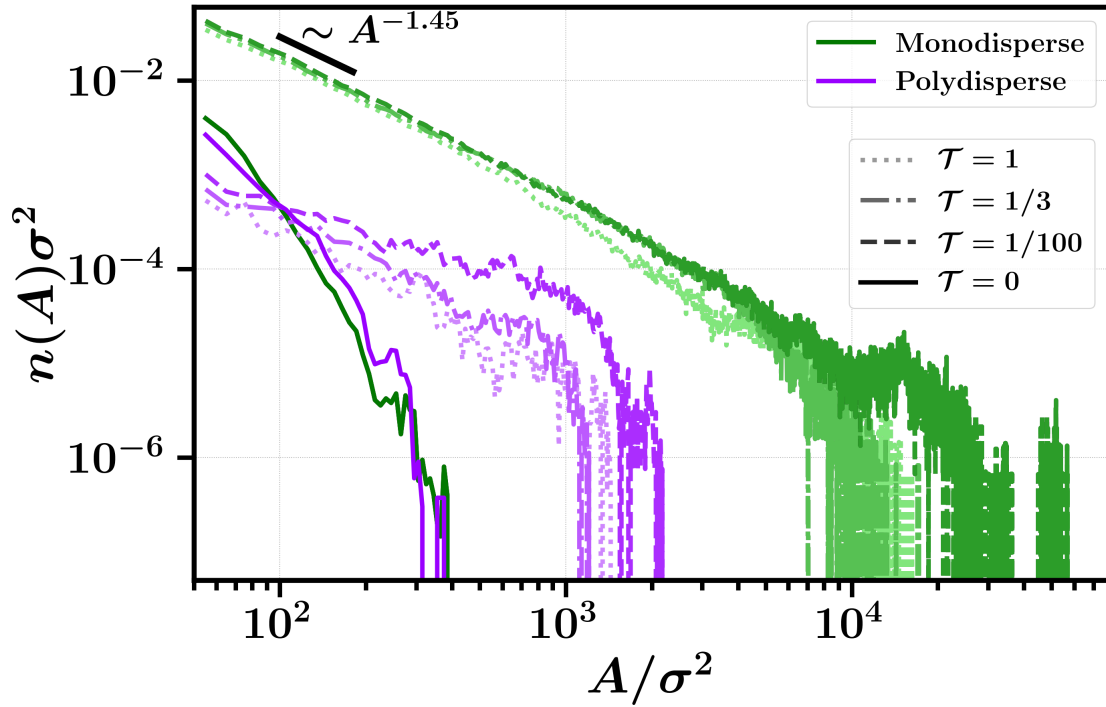

 Figure D.9: $P(A)$ for all monodisperse simulated values of $\mathcal{T} > 0$.

(not included in the main text) in Fig. D.12.

The videos shown in Section D.6 point towards collective motion being a dominant transport mechanism for thermalized simulations. In order to quantify this observation, we turn to the spatial correlation function of the velocity $Q(r)$ employed by Caprini *et al.* [151–153]:

$$Q_i(r) = 1 - 2 \sum_j \frac{\theta_{ij}}{\mathcal{N}_r \pi}, \quad (\text{D.7})$$

where $\mathcal{N}_r$ is the number of particles in a shell of some thickness Δr centered a distance r away from the i th particle and θ_{ij} is the angle between the velocities of particle i and particle j . Clearly, velocities are perfectly aligned θ_{ij} will be zero for all j , and $Q_i(r) = 1$. At an opposite extreme, if velocities are perfectly antialigned then $\theta_{ij} = \pi$ for all j , and $Q_i(r) = -1$. We use the DBSCAN algorithm from the


 Figure D.10: $n(A)$ plotted up to higher values of A .

previous Sections to find all particles within the dense phase, then calculate the average value of $Q(r) = (1/N_{\text{dense}}) \sum_{i=1}^{N_{\text{dense}}} Q_i(r)$ for each particle.

Note that we are analyzing Brownian data, and thus instantaneous velocities are poorly defined. We therefore must use *average* velocities $\mathbf{v}_i(t) \equiv \mathbf{r}_i(t + \Delta t) - \mathbf{r}_i(t)$ which is now a quantity dependent on the arbitrary choice of Δt . We therefore evaluate $Q(r)$ at the distance corresponding to the first nearest neighbor shell as a function of the choice of Δt across a wide range of time separations. The results of this calculation are plotted in Fig. D.13.

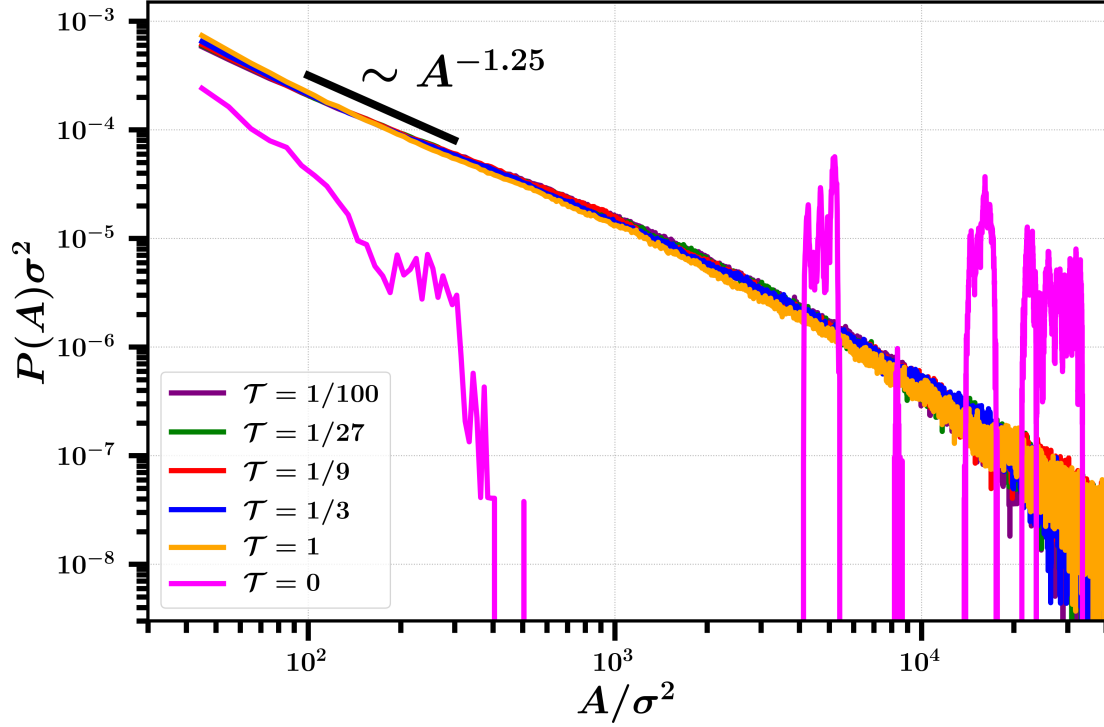


Figure D.11: Grain distribution for all monodisperse simulated values, including $\tau = 0$.

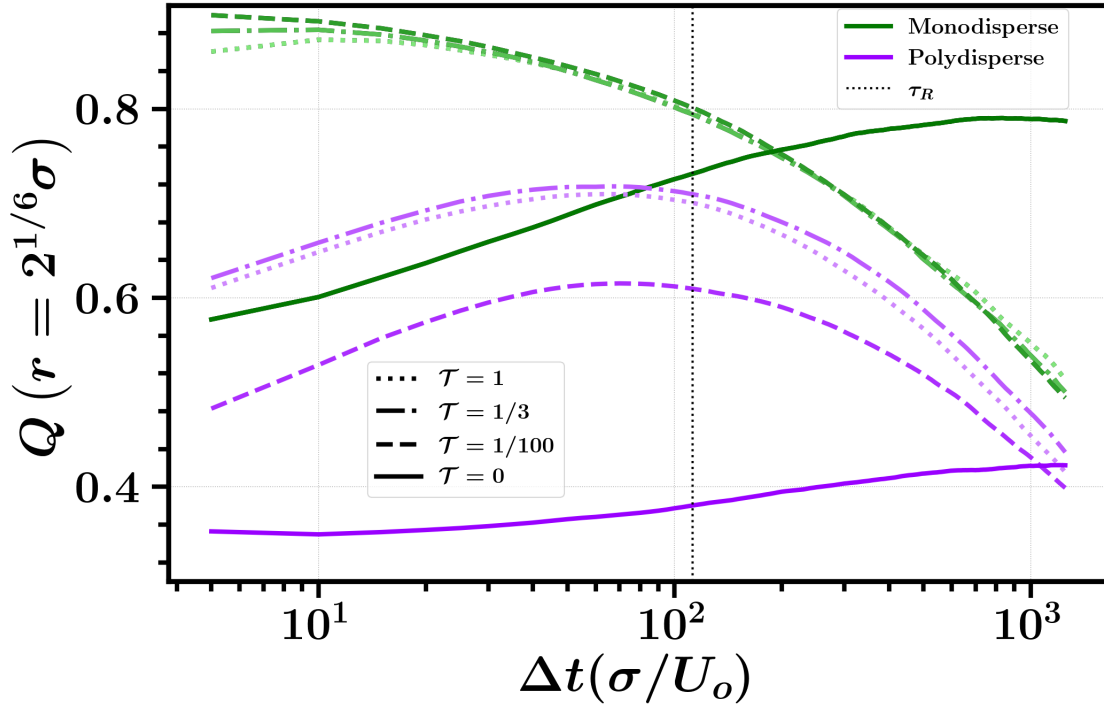


Figure D.13: $Q(r)$ evaluated at the nearest neighbor separation for different selections of Δt .

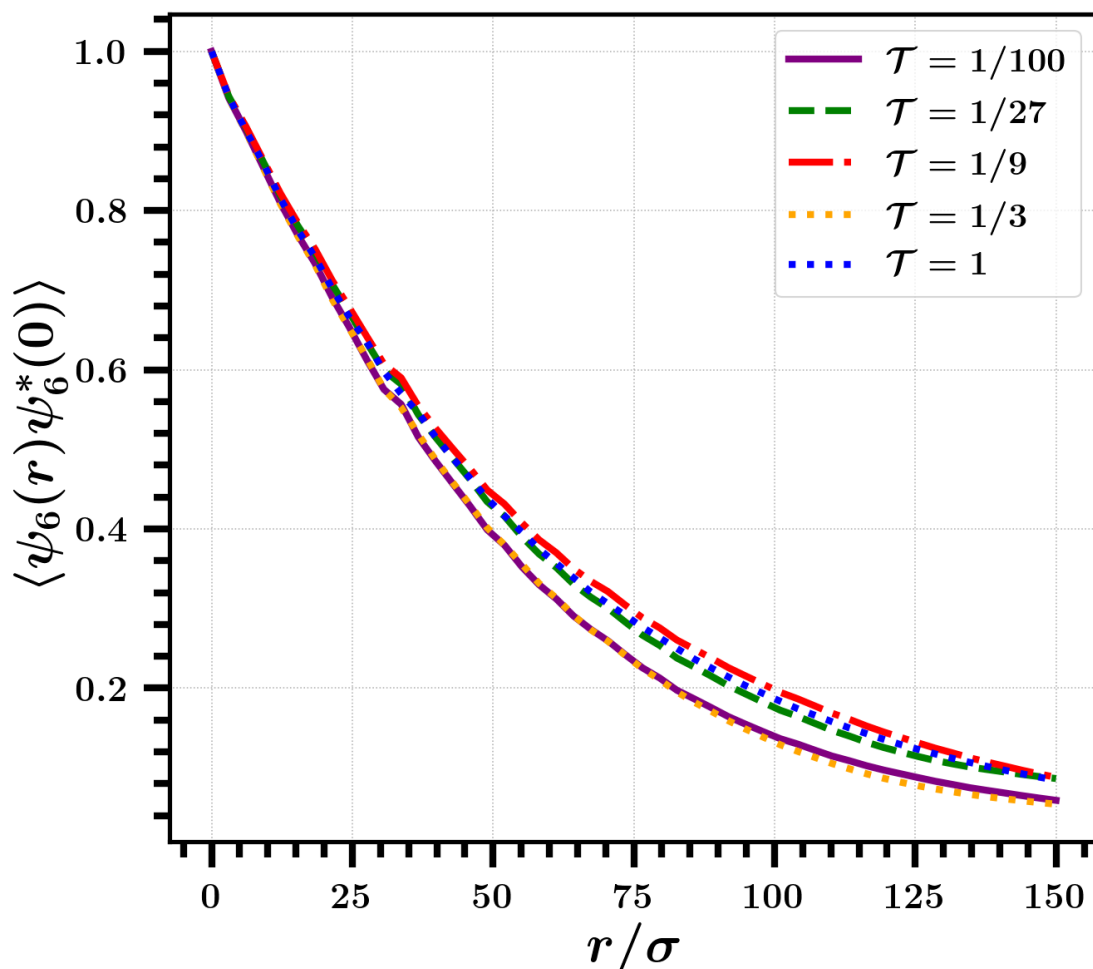


Figure D.12: Hexatic spatial correlations for all monodisperse simulated values of $\mathcal{T} > 0$.

From Fig. D.13 one can determine the value of Δt that maximizes spatial velocity correlations. All plots of $Q(r)$ in the main text uses the maximizing value of Δt at each particular simulation condition. It is interesting to note that correlations in thermal monodisperse simulations were maximized at relatively small time separations $\mathcal{O}(\sigma/U_o)$, thermal polydisperse simulations were maximized at time separations slightly less than $\mathcal{O}(\tau_R)$, and athermal simulations were maximized at time separations of multiples of τ_R . In general, these correlations were higher for thermalized monodisperse systems, indicating that collective motion is most

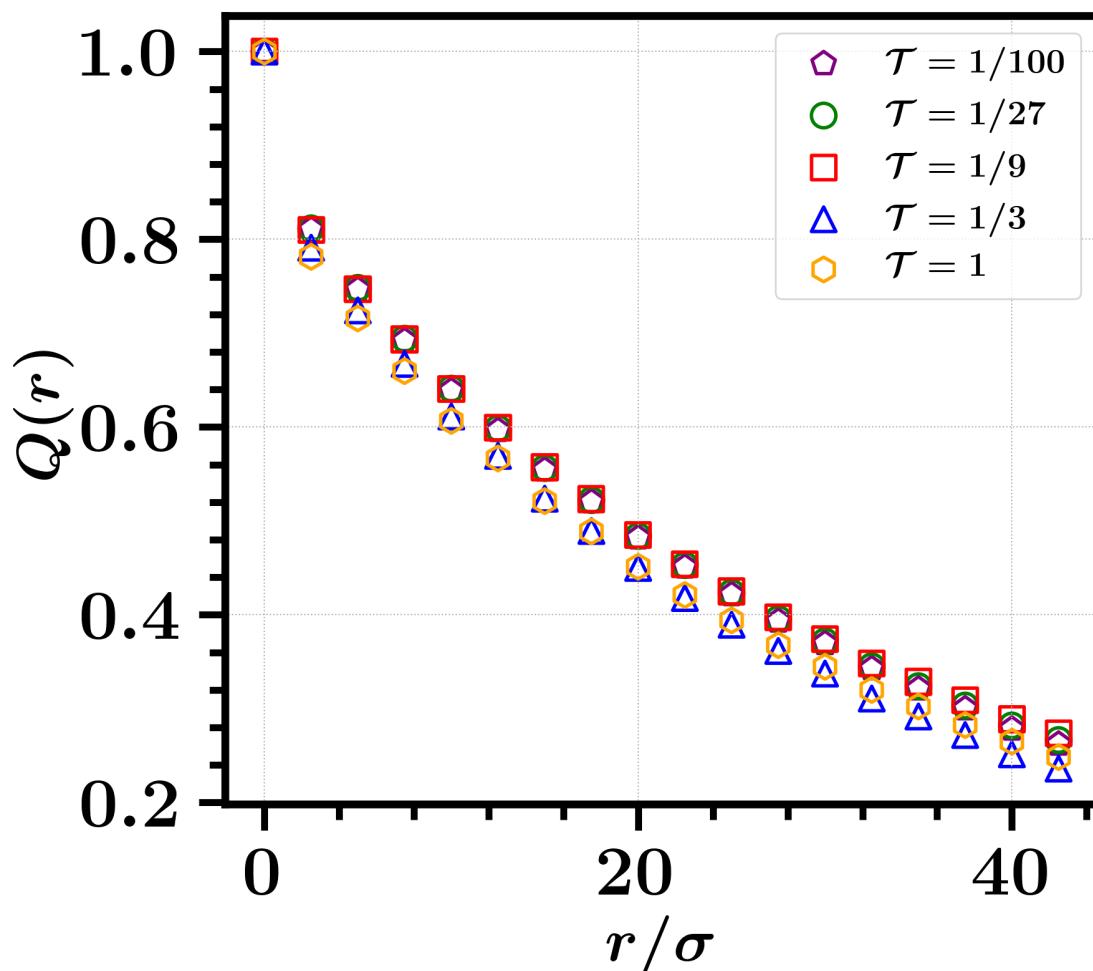


Figure D.14: $Q(r)$ for all monodisperse simulated values of $\tau > 0$.

significant at short time scales under these conditions. We also provide the plots of $Q(r)$ for all thermalized systems (not included in the main text) in Fig. D.14.

While the above provides insight on velocity correlations, they do not allow one to clearly understand the *speed* at which this correlated/uncorrelated transport occurs. We therefore calculate the mean squared displacement of particles in the dense phase. In order to ensure that the measurement was only of dense phase particles, we employed the DBSCAN algorithm to only consider the mean squared displacement of particles that remained in the dense phase. This systematically

excludes particles which escape to the gas phase, which have typically moved a greater distance than other particles, and therefore represents an underestimate of the true dense phase MSD. Therefore the plots in the main text which indicate subdiffusive transport in the athermal simulations may not necessarily be evidence of this.

D.5 Dense Polydisperse Simulation

All polydisperse simulations reported in the main text were done at an overall area fraction of $\phi_o = 0.49$ while all monodisperse simulations reported in the main text were done at an overall area fraction of $\phi_o = 0.64$. In order to confirm that this discrepancy is purely quantitative and did not result in any qualitative differences, we calculated the $\mathcal{T} = 1/3$ polydisperse simulation at $\phi_o = 0.64$. The statistics of the bubbles were similar, with the area fraction of gas trapped in the bubbles $\phi_B = (1.6 \pm 1.5) \times 10^{-3}$, which remains significantly smaller than that of the monodisperse thermal simulations done at similar overall densities.

$n(A)$ at this simulation is plotted along with the $n(A)$ from a simulation at the same conditions but $\phi_o = 0.49$ is shown below.

The hexatic order spatial correlations, $Q(r)$, and the mean squared displacement were also all calculated for both the polydisperse thermal $\phi_o = 0.49$ and $\phi_o = 0.64$ simulation and are plotted below.

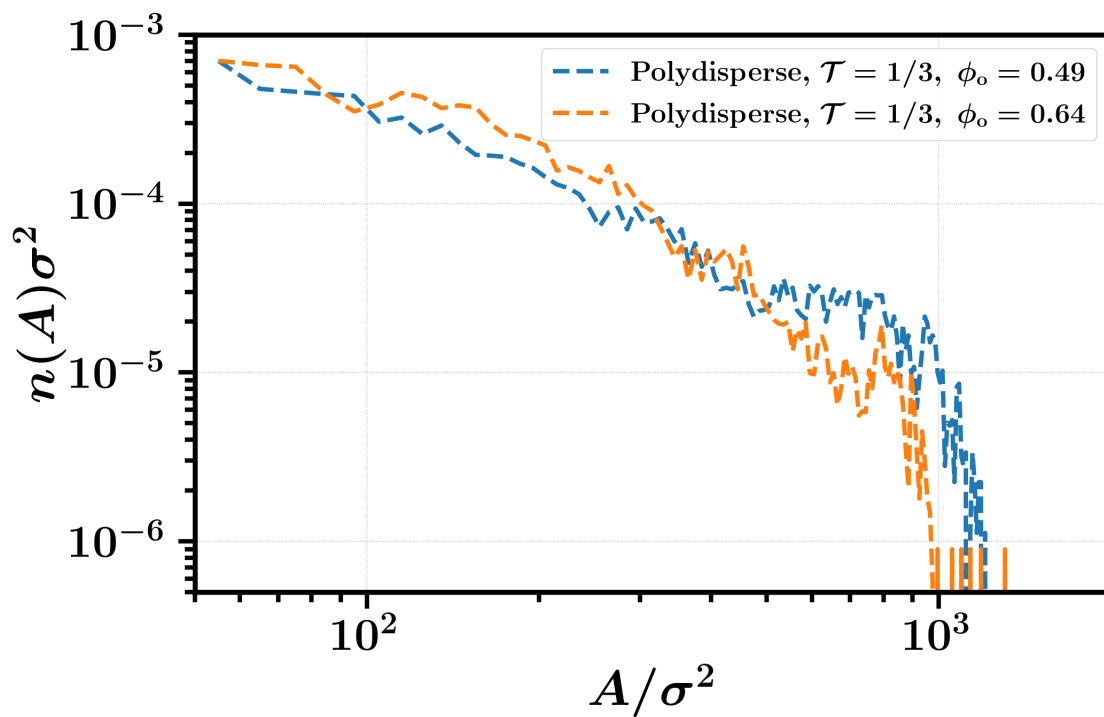


Figure D.15: $n(A)$ for polydisperse $\mathcal{T} = 1/3$ simulations at $\phi_o = 0.49$ and $\phi_o = 0.64$.

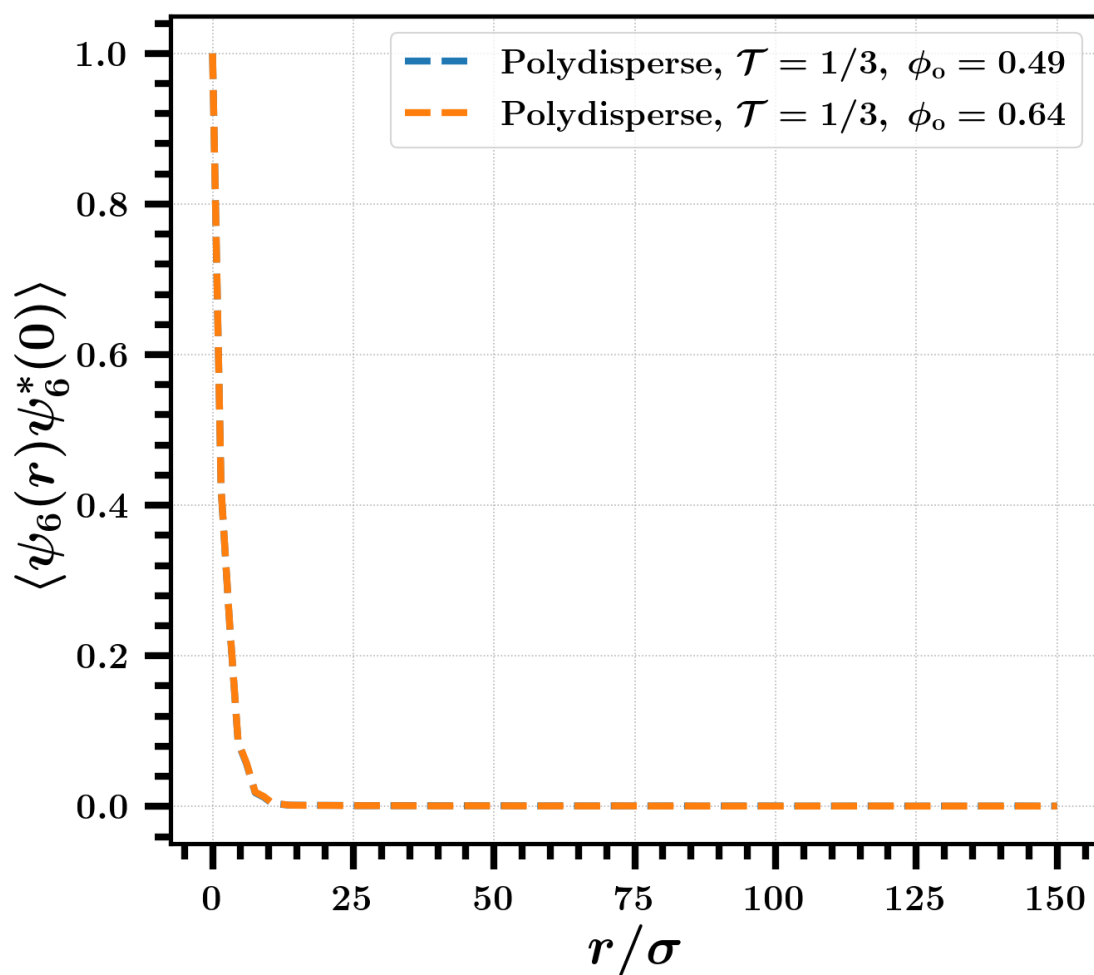


Figure D.16: Spatial correlations of hexatic order for polydisperse $\mathcal{T} = 1/3$ simulations at $\phi_o = 0.49$ and $\phi_o = 0.64$.

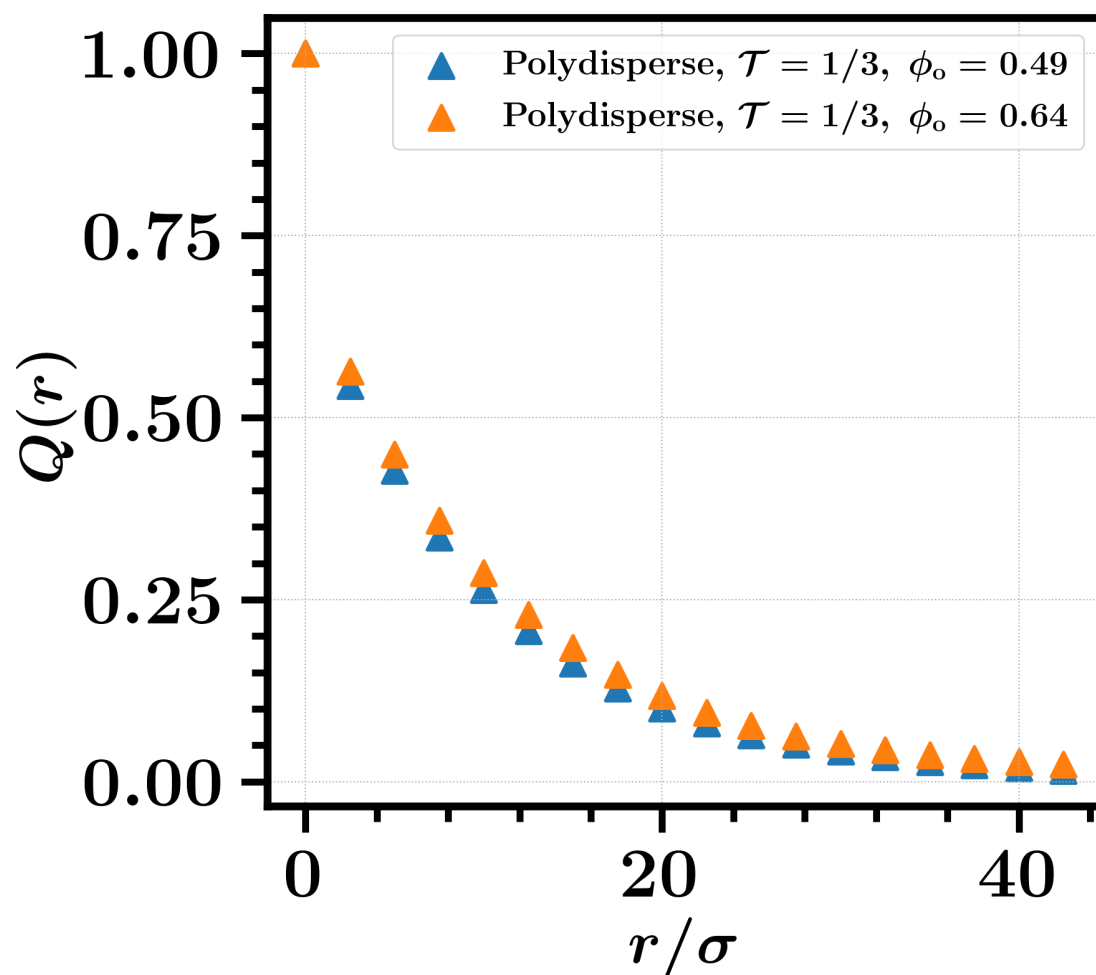


Figure D.17: Spatial correlations of velocity orientation for polydisperse $\mathcal{T} = 1/3$ simulations at $\phi_o = 0.49$ and $\phi_o = 0.64$.

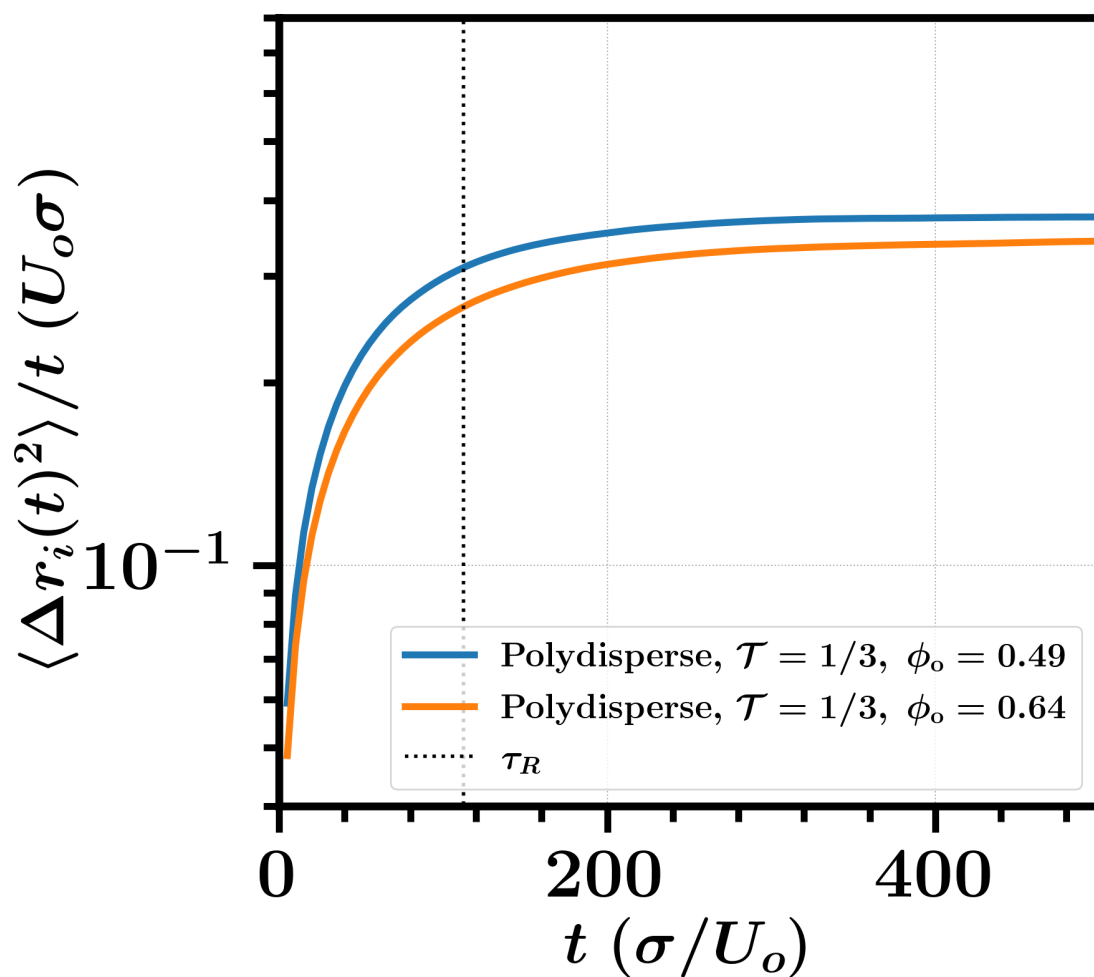


Figure D.18: Mean squared displacement for polydisperse $\mathcal{T} = 1/3$ simulations at $\phi_o = 0.49$ and $\phi_o = 0.64$.

D.6 Media

Each video listed below displays a window of selected simulations reported in the main text. All videos labeled “closeup” correspond to a total time of $2.5 \times 10^3 \sigma/U_o$ with frame spacing of $5 \sigma/U_o$ and all other videos correspond to a total time of $10^4 \sigma/U_o$ with frame spacing of $25 \sigma/U_o$. All videos were rendered using the OVITO software package [67] at a rate of 8 frames per second and are publicly available at the following URL:

<https://berkeley.box.com/s/rmjqhhvi368ae5j5fwo31b5aweeemjy9>

- [monodisperse_athermal_video.mp4]:
Dynamics of monodisperse system with $\mathcal{T} = 0$, zoomed out such that entire system is visible.
- [monodisperse_athermal_video_closeup.mp4]:
Dynamics of monodisperse system with $\mathcal{T} = 0$, zoomed in such that motion of individual particles is visible.
- [monodisperse_thermal_video.mp4]:
Dynamics of monodisperse system with $\mathcal{T} = 1/3$, zoomed out such that entire system is visible.
- [monodisperse_thermal_video_closeup.mp4]:
Dynamics of monodisperse system with $\mathcal{T} = 1/3$, zoomed in such that motion of individual particles is visible.
- [polydisperse_athermal_video.mp4]:
Dynamics of polydisperse system with $\mathcal{T} = 0$, zoomed out such that entire system is visible.
- [polydisperse_athermal_video_closeup.mp4]:
Dynamics of polydisperse system with $\mathcal{T} = 0$, zoomed in such that motion of individual particles is visible.

- [polydisperse_thermal_video.mp4]:
Dynamics of polydisperse system with $\mathcal{T} = 1/3$, zoomed out such that entire system is visible.
- [polydisperse_thermal_video_closeup.mp4]:
Dynamics of polydisperse system with $\mathcal{T} = 1/3$, zoomed in such that motion of individual particles is visible.

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