

Supporting Information: Mechanical Theory of Nonequilibrium Coexistence and Motility-Induced Phase Separation

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ACTIVE BROWNIAN PARTICLE CONSERVATION EQUATIONS AND CLOSURES

We provide a derivation of the equations presented in the main text: the conservation equations for density, linear momentum, and the other orientational moment fields for a collection of N interacting active Brownian particles (ABPs). We emphasize that these equations for interacting ABPs have appeared in various forms throughout the literature [1–3]. However, in addition to deriving the equations, we will introduce the closures and assumptions that allow us to determine the stationary states of coexistence.

The Fokker-Planck (or Smoluchowski) equation describing the N -body distribution of particle positions and orientations follows from the particle equations-of-motion and is given by:

$$\frac{\partial f_N(\mathbf{\Gamma}, t)}{\partial t} + \sum_{\alpha} \nabla_{\alpha} \cdot \mathbf{j}_{\alpha}^T + \sum_{\alpha} \nabla_{\alpha}^R \cdot \mathbf{j}_{\alpha}^R = 0. \quad (\text{S1a})$$

Here, $f_N(\mathbf{\Gamma}, t)$ is the probability density of observing a configuration $\mathbf{\Gamma} \equiv (\mathbf{r}_1, \dots, \mathbf{r}_N, \mathbf{q}_1, \dots, \mathbf{q}_N)$ at time t , \mathbf{r}_{α} and \mathbf{q}_{α} ($|\mathbf{q}_{\alpha}| = 1$) are the position and orientation vectors of particle α , \mathbf{j}_{α}^T and \mathbf{j}_{α}^R are the translational and rotational fluxes of particle α , and $\nabla_{\alpha} = \partial/\partial \mathbf{r}_{\alpha}$ and $\nabla_{\alpha}^R = \mathbf{q}_{\alpha} \times \partial/\partial \mathbf{q}_{\alpha}$ are translational and rotational gradient operators. The fluxes are given by:

$$\mathbf{j}_{\alpha}^T = U_0 \mathbf{q}_{\alpha} f_N + \frac{1}{\zeta} \mathbf{F}_{\alpha} f_N - D_T \nabla_{\alpha} f_N, \quad (\text{S1b})$$

$$\mathbf{j}_{\alpha}^R = -\frac{1}{\tau_R} \nabla_{\alpha}^R f_N, \quad (\text{S1c})$$

where U_0 is the intrinsic active speed, ζ is the translational drag coefficient, D_T is the translational Brownian diffusivity (neglected in the main text but provided here for completeness), τ_R is the reorientation time scale, and \mathbf{F}_{α} is the conservative force on particle α arising from a the potential energy $U(\mathbf{r}_1, \dots, \mathbf{r}_N)$. We consider pairwise additive isotropic interparticle interactions such that $U(\mathbf{r}_1, \dots, \mathbf{r}_N) = \sum_{\alpha} U^{\text{ext}}(\mathbf{r}_{\alpha}) + \sum_{\alpha} \sum_{\beta \neq \alpha} U_2(r_{\alpha\beta})/2$. Here, $U^{\text{ext}}(\mathbf{x})$ is the externally imposed potential at position \mathbf{x} , $U_2(r)$ is the pair interaction potential, and $\mathbf{r}_{\alpha\beta} = \mathbf{r}_{\alpha} - \mathbf{r}_{\beta}$ ($r_{\alpha\beta} = |\mathbf{r}_{\alpha\beta}|$). The force on particle α arising from the conservative potentials thus has two contributions $\mathbf{F}_{\alpha} = \mathbf{F}_{\alpha}^{\text{ext}} + \mathbf{F}_{\alpha}^C$, where $\mathbf{F}_{\alpha}^{\text{ext}} = -\nabla_{\alpha} U^{\text{ext}}(\mathbf{r}_{\alpha})$, $\mathbf{F}_{\alpha}^C = \sum_{\beta \neq \alpha} \mathbf{F}_{\alpha\beta}^C$, and $\mathbf{F}_{\alpha\beta}^C = -\nabla_{\alpha} U_2(r_{\alpha\beta})$. From Eq. (S1) the dynamical operator is defined as $\mathcal{L} \equiv \sum_{\alpha} [\nabla_{\alpha} \cdot (-U_0 \mathbf{q}_{\alpha} - \zeta^{-1} \mathbf{F}_{\alpha} + D_T \nabla_{\alpha}) + \nabla_{\alpha}^R \cdot (\tau_R^{-1} \nabla_{\alpha}^R)]$ such that $\partial f_N / \partial t = \mathcal{L} f_N$.

The full N -body distribution function f_N allows for the determination of the ensemble average of any observable \mathcal{O} : $\langle \mathcal{O} \rangle = \langle \hat{\mathcal{O}}(\mathbf{\Gamma}) \rangle$, where $\langle \cdot \rangle \equiv \int (\cdot) f_N(\mathbf{\Gamma}, t) d\mathbf{\Gamma}$ is the ensemble average and $\hat{\mathcal{O}}(\mathbf{\Gamma})$ is the microscopic definition of the observable \mathcal{O} . The time evolution of an observable is then:

$$\frac{\partial \mathcal{O}}{\partial t} = \int \hat{\mathcal{O}} \left(\frac{\partial f_N}{\partial t} \right) d\mathbf{\Gamma} = \int \hat{\mathcal{O}} (\mathcal{L} f_N) d\mathbf{\Gamma} = \int (\mathcal{L}^* \hat{\mathcal{O}}) f_N d\mathbf{\Gamma} = \langle \mathcal{L}^* \hat{\mathcal{O}} \rangle, \quad (\text{S2})$$

where, in the case of Eq. (S1), the adjoint of \mathcal{L} is $\mathcal{L}^* \equiv \sum_{\alpha} [(U_0 \mathbf{q}_{\alpha} + \zeta^{-1} \mathbf{F}_{\alpha} + D_T \nabla_{\alpha}) \cdot \nabla_{\alpha} + \tau_R^{-1} \nabla_{\alpha}^R \cdot \nabla_{\alpha}^R]$.

We first derive an evolution equation of the (number) density field $\rho(\mathbf{x}, t) = \langle \hat{\rho}(\mathbf{x}) \rangle$, where $\hat{\rho}(\mathbf{x}) = \sum_{\alpha} \delta(\mathbf{x} - \mathbf{x}_{\alpha})$ is the microscopic density of particles. The continuity equation directly follows from this procedure:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{j}^{\rho} = 0, \quad (\text{S3a})$$

where:

$$\mathbf{j}^{\rho} = U_0 \mathbf{m}(\mathbf{x}, t) + \frac{1}{\zeta} \mathbf{F}^{\text{ext}}(\mathbf{x}) \rho + \frac{1}{\zeta} \nabla \cdot \boldsymbol{\sigma}^C(\mathbf{x}, t) - D_T \nabla \rho. \quad (\text{S3b})$$

Here, $\nabla \equiv \partial/\partial \mathbf{x}$ is the spatial gradient operator, \mathbf{j}^ρ is the particle flux, $\mathbf{m}(\mathbf{x}, t) \equiv \langle \hat{\mathbf{m}}(\mathbf{x}) \rangle$ is the polarization density field, $\hat{\mathbf{m}}(\mathbf{x}) = \sum_\alpha \mathbf{q}_\alpha \delta(\mathbf{x} - \mathbf{r}_\alpha)$ is the microscopic density of polarization, and:

$$\boldsymbol{\sigma}^C(\mathbf{x}, t) = \left\langle -\frac{1}{2} \sum_\alpha \sum_{\alpha \neq \beta} \mathbf{r}_{\alpha\beta} \mathbf{F}_{\alpha\beta}^C b_{\alpha\beta} \right\rangle, \quad (\text{S4})$$

is the stress generated by the pairwise interparticle forces and $b_{\alpha\beta}(\mathbf{x}; \mathbf{r}_\alpha, \mathbf{r}_\beta) = \int_0^1 \delta(\mathbf{x} - \mathbf{r}_\beta - \lambda \mathbf{r}_{\alpha\beta}) d\lambda$ is the bond function [4, 5].

The four terms in the particle flux (S3b) correspond to the four modes of particle transport: transport driven by the active force, external forcing, interparticle forces, and Brownian motion. Equation (S3b) is also a statement of linear momentum conservation. To see this we rearrange and find:

$$\mathbf{0} = \nabla \cdot \boldsymbol{\sigma}(\mathbf{x}, t) + \mathbf{b}(\mathbf{x}, t), \quad (\text{S5a})$$

where the body forces are the terms in Eq. S3b that *generally* may not be expressed as divergences of a tensor:

$$\mathbf{b}(\mathbf{x}, t) = -\zeta \mathbf{j}^\rho(\mathbf{x}, t) + \zeta U_0 \mathbf{m}(\mathbf{x}, t) + \mathbf{F}^{\text{ext}}(\mathbf{x}) \rho(\mathbf{x}, t), \quad (\text{S5b})$$

and the stresses are:

$$\boldsymbol{\sigma}(\mathbf{x}, t) = \boldsymbol{\sigma}^C(\mathbf{x}, t) - \zeta D_T \rho(\mathbf{x}, t) \mathbf{I}, \quad (\text{S5c})$$

where $-\zeta D_T \rho \mathbf{I}$ represents the Brownian “ideal gas” stress and \mathbf{I} is the second-rank identity tensor.

In order to solve Eq. (S3), we require the polarization density \mathbf{m} and the stress $\boldsymbol{\sigma}^C$. The polarization density has its own evolution equation. By letting $\hat{\mathcal{O}} = \hat{\mathbf{m}}$ in Eq. (S2) and following procedures similar to what were applied in order to obtain Eq. (S3), we find:

$$\frac{\partial \mathbf{m}}{\partial t} + \nabla \cdot \mathbf{j}^{\mathbf{m}} + \frac{d-1}{\tau_R} \mathbf{m} = 0, \quad (\text{S6a})$$

$$\mathbf{j}^{\mathbf{m}} = U_0 \tilde{\mathbf{Q}}(\mathbf{x}, t) + \frac{1}{\zeta} \mathbf{F}^{\text{ext}} \mathbf{m} + \frac{1}{\zeta} \boldsymbol{\kappa}^{\mathbf{m}}(\mathbf{x}, t) + \frac{1}{\zeta} \nabla \cdot \boldsymbol{\Sigma}^{\mathbf{m}}(\mathbf{x}, t) - D_T \nabla \mathbf{m}, \quad (\text{S6b})$$

where $\mathbf{j}^{\mathbf{m}}$ is the polarization flux, d is the spatial dimension, and $\tilde{\mathbf{Q}}(\mathbf{x}, t) \equiv \langle \hat{\mathbf{Q}}(\mathbf{x}) \rangle$ is the nematic order density field (where $\hat{\mathbf{Q}}(\mathbf{x}) = \sum_\alpha \mathbf{q}_\alpha \mathbf{q}_\alpha \delta(\mathbf{x} - \mathbf{r}_\alpha)$ is the microscopic nematic density). The interparticle forces contribute to the transport of the polarization in body-force-like (i.e., no divergence) $\boldsymbol{\kappa}^{\mathbf{m}}$ and stress-like $\boldsymbol{\Sigma}^{\mathbf{m}}$ manners, which are defined as:

$$\boldsymbol{\kappa}^{\mathbf{m}}(\mathbf{x}, t) = \left\langle \frac{1}{2} \sum_\alpha \sum_{\alpha \neq \beta} \mathbf{F}_{\alpha\beta}^C (\mathbf{q}_\alpha - \mathbf{q}_\beta) \delta(\mathbf{x} - \mathbf{r}_\alpha) \right\rangle, \quad (\text{S7})$$

$$\boldsymbol{\Sigma}^{\mathbf{m}}(\mathbf{x}, t) = \left\langle -\frac{1}{2} \sum_\alpha \sum_{\alpha \neq \beta} \mathbf{r}_{\alpha\beta} \mathbf{F}_{\alpha\beta}^C \mathbf{q}_\alpha b_{\alpha\beta} \right\rangle. \quad (\text{S8})$$

Equation (S8) makes clear that $d\mathbf{S}(\mathbf{x}) \cdot \boldsymbol{\Sigma}^{\mathbf{m}}/\zeta$ is the average transport of polarization due to the interparticle forces acting *across* the infinitesimal area dS from the direction of $d\mathbf{S}$. From the definition [Eq. (S7)] of the body-force-like term $\boldsymbol{\kappa}^{\mathbf{m}}$, we observe that configurations with $\mathbf{q}_\alpha = \mathbf{q}_\beta$ do not contribute to $\boldsymbol{\kappa}^{\mathbf{m}}$ and configurations with $\mathbf{q}_\alpha = -\mathbf{q}_\beta$ contribute the most to $\boldsymbol{\kappa}^{\mathbf{m}}$ in magnitude. This observation is indicative that $\boldsymbol{\kappa}^{\mathbf{m}}$ is correlated with the reduction in the effective active speed U_{eff} due to interparticle interactions — a pair of particles slow down when they collide head to head but active motion is largely unaffected when interacting particles are oriented in the same direction. From the scaling analysis of the active pressure $p_{\text{act}} \sim \rho \zeta U_0 \tau_R U_{\text{eff}}$ [6] and recognizing that the active pressure is proportional to the trace of the polarization flux (with $\mathbf{j}^{\mathbf{m}} \sim \rho \langle \dot{\mathbf{x}} \mathbf{q} \rangle$) such that $p_{\text{act}} \sim \rho \zeta U_0 \tau_R \langle \dot{\mathbf{x}} \cdot \mathbf{q} \rangle$ [2, 7–10], we can indeed identify that $\boldsymbol{\kappa}^{\mathbf{m}}$ is directly related to the reduction in the effective speed of active transport of polarization. This motivates a constitutive equation

$$\boldsymbol{\kappa}^{\mathbf{m}} = -\zeta (U_0 - U_{\text{eff}}^{\mathbf{m}}) \tilde{\mathbf{Q}}, \quad (\text{S9})$$

which leads to

$$\mathbf{j}^{\mathbf{m}} = U_0 \bar{U}^{\mathbf{m}} \tilde{\mathbf{Q}} + \frac{1}{\zeta} \mathbf{F}^{\text{ext}} \mathbf{m} + \frac{1}{\zeta} \nabla \cdot \Sigma^{\mathbf{m}} - D_T \nabla \mathbf{m}, \quad (\text{S10})$$

where $U_{\text{eff}}^{\mathbf{m}} = U_0 \bar{U}^{\mathbf{m}}$ is the effective speed of active polarization transport. The dimensionless quantity $\bar{U}^{\mathbf{m}} (\in [0, 1])$ represents the effective speed relative to the intrinsic speed U_0 and is an equation-of-state depending on the system volume (area) fraction ϕ and activity ℓ_0/D . $\bar{U}^{\mathbf{m}} \approx 1$ when particles move nearly freely at low ($\phi \ll 1$) densities and $\bar{U}^{\mathbf{m}} \approx 0$ when particles mobility is limited due to interparticle interactions.

To close our equations, expressions for $\sigma^{\mathbf{C}}$, $\tilde{\mathbf{Q}}$, $\bar{U}^{\mathbf{m}}$, and $\Sigma^{\mathbf{m}}$ are required. The nematic order density $\tilde{\mathbf{Q}}$ follows its own evolution equation which can again be derived from Eq. (S2) with $\hat{\mathcal{O}} = \tilde{\mathbf{Q}}$:

$$\frac{\partial \tilde{\mathbf{Q}}}{\partial t} + \nabla \cdot \mathbf{j}^{\tilde{\mathbf{Q}}} + \frac{2d}{\tau_R} \left(\tilde{\mathbf{Q}} - \frac{1}{d} \rho \mathbf{I} \right) = \mathbf{0}, \quad (\text{S11a})$$

$$\mathbf{j}^{\tilde{\mathbf{Q}}} = U_0 \tilde{\mathbf{B}}(\mathbf{x}, t) + \frac{1}{\zeta} \mathbf{F}^{\text{ext}} \tilde{\mathbf{Q}} + \frac{1}{\zeta} \kappa^{\tilde{\mathbf{Q}}}(\mathbf{x}, t) + \frac{1}{\zeta} \nabla \cdot \Sigma^{\tilde{\mathbf{Q}}}(\mathbf{x}, t) - D_T \nabla \tilde{\mathbf{Q}}. \quad (\text{S11b})$$

Here, $\mathbf{j}^{\tilde{\mathbf{Q}}}$ is the nematic order flux and $\tilde{\mathbf{B}}(\mathbf{x}, t) \equiv \langle \sum_{\alpha} \mathbf{q}_{\alpha} \mathbf{q}_{\alpha} \mathbf{q}_{\alpha} \delta(\mathbf{x} - \mathbf{r}_{\alpha}) \rangle$. Interparticle interactions again result in body-force- and stress-like terms in the nematic order flux:

$$\kappa^{\tilde{\mathbf{Q}}}(\mathbf{x}, t) = \left\langle \frac{1}{2} \sum_{\alpha} \sum_{\alpha \neq \beta} \mathbf{F}_{\alpha\beta}^{\mathbf{C}} (\mathbf{q}_{\alpha} \mathbf{q}_{\alpha} - \mathbf{q}_{\beta} \mathbf{q}_{\beta}) \delta(\mathbf{x} - \mathbf{r}_{\alpha}) \right\rangle, \quad (\text{S12})$$

$$\Sigma^{\tilde{\mathbf{Q}}}(\mathbf{x}, t) = \left\langle -\frac{1}{2} \sum_{\alpha} \sum_{\alpha \neq \beta} \mathbf{r}_{\alpha\beta} \mathbf{F}_{\alpha\beta}^{\mathbf{C}} \mathbf{q}_{\alpha} \mathbf{q}_{\alpha} b_{\alpha\beta} \right\rangle. \quad (\text{S13})$$

Again, the stress-like term $\Sigma^{\tilde{\mathbf{Q}}}(\mathbf{x}, t)$ represents the average transport of nematic order due to interparticle forces acting across \mathbf{x} and $\kappa^{\tilde{\mathbf{Q}}}$ is related to the reduction in the effective speed of active transport of the nematic order. We again propose a constitutive relation:

$$\kappa^{\tilde{\mathbf{Q}}} = -\zeta (U_0 - U_{\text{eff}}^{\tilde{\mathbf{Q}}}) \tilde{\mathbf{B}}. \quad (\text{S14})$$

Consequently, the nematic order flux becomes:

$$\mathbf{j}^{\tilde{\mathbf{Q}}} = U_0 \bar{U}^{\tilde{\mathbf{Q}}} \tilde{\mathbf{B}}(\mathbf{x}, t) + \frac{1}{\zeta} \mathbf{F}^{\text{ext}} \tilde{\mathbf{Q}} + \frac{1}{\zeta} \nabla \cdot \Sigma^{\tilde{\mathbf{Q}}}(\mathbf{x}, t) - D_T \nabla \tilde{\mathbf{Q}}, \quad (\text{S15})$$

where $U_{\text{eff}}^{\tilde{\mathbf{Q}}} = U_0 \bar{U}^{\tilde{\mathbf{Q}}}$ is the effective speed of active nematic order transport. The dimensionless quantity $\bar{U}^{\tilde{\mathbf{Q}}}$ is again an equation-of-state depending on ϕ and ℓ_0/D .

It proves convenient to rewrite our equations with the traceless tensorial orientational moments to exclude the portions that are dependent on the lower order orientational moments (i.e., ρ and \mathbf{m}). The traceless nematic order is defined as $\mathbf{Q} = \tilde{\mathbf{Q}} - \rho \mathbf{I}/d$ while $\mathbf{B} = \tilde{\mathbf{B}} - \alpha \cdot \mathbf{m}/(d+2)$, where α is a fourth-rank isotropic tensor. (In indicial notation, $\alpha_{ijkl} = \delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}$ where δ_{ij} is the second-rank identity tensor.) The polarization flux [Eq. (S10)] and nematic order evolution equation [Eqs. (S11a) and (S15)] become:

$$\mathbf{j}^{\mathbf{m}} = U_0 \bar{U}^{\mathbf{m}} \left(\mathbf{Q} + \frac{1}{d} \rho \mathbf{I} \right) + \frac{1}{\zeta} \mathbf{F}^{\text{ext}} \mathbf{m} + \frac{1}{\zeta} \nabla \cdot \Sigma^{\mathbf{m}} - D_T \nabla \mathbf{m}, \quad (\text{S16})$$

$$\frac{\partial \mathbf{Q}}{\partial t} + \nabla \cdot \mathbf{j}^{\mathbf{Q}} + \frac{2d}{\tau_R} \mathbf{Q} = \mathbf{0}, \quad (\text{S17a})$$

where the traceless nematic flux $\mathbf{j}^{\mathbf{Q}} = \mathbf{j}^{\tilde{\mathbf{Q}}} - \frac{1}{d} \mathbf{j}^{\rho} \mathbf{I}$ is:

$$\mathbf{j}^{\mathbf{Q}} = U_0 \bar{U}^{\tilde{\mathbf{Q}}} \left(\mathbf{B} + \frac{1}{d+2} \alpha \cdot \mathbf{m} \right) - \frac{1}{d} U_0 \mathbf{m} \mathbf{I} + \frac{1}{\zeta} \mathbf{F}^{\text{ext}} \mathbf{Q} + \frac{1}{\zeta} \nabla \cdot \Sigma^{\tilde{\mathbf{Q}}} - \frac{1}{\zeta d} \nabla \cdot \sigma^{\mathbf{C}} \mathbf{I} - D_T \nabla \mathbf{Q}. \quad (\text{S17b})$$

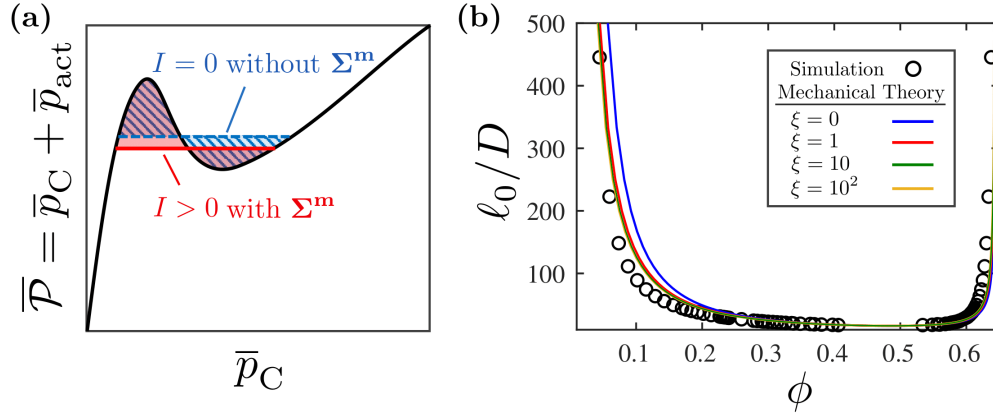


FIG. S1. (a) Schematic illustrating the effect of Σ^m to the coexistence effective pressure $\mathcal{P}^{\text{coexist}}$. On the $\mathcal{P} - p_C$ diagram, the integral I [Eq. (S18)] represents the area between the constant activity curve (black) and horizontal line $\mathcal{P} = \mathcal{P}^{\text{coexist}}$. When the stress-like contribution Σ^m is neglected, $I = 0$ since the equal-area construction variable is $\mathcal{E}(\rho) = p_C$. However, $I > 0$ when Σ^m is considered and the corresponding coexistence effective pressure (red) should be lower than the coexistence effective pressure predicted by $I = 0$ (blue). (b) Coexistence curves for athermal active Brownian spheres (3d) obtained by the mechanical theory with the stress-like contribution of the interparticle forces to the polar order Σ^m . The parameter ξ represents the magnitude of Σ^m . The reduced $\mathcal{P}^{\text{coexist}}$ resulting from Σ^m increases the difference between the coexisting densities. However, this difference, even for extreme values of ξ , is not significant.

Now, we have coupled evolution equations for the density [Eq. (S3)], polar order [Eqs. (S6a) and (S16)], and nematic order [Eq. (S17)]. Closing these equations will require expressions for the following unknowns: \mathbf{B} , $\Sigma^{\hat{Q}}$, Σ^m , σ^C , \bar{U}^m , and $\bar{U}^{\hat{Q}}$. Importantly, since our mechanical theory for phase coexistence requires density gradients only up to second order, \mathbf{B} and $\Sigma^{\hat{Q}}$ can be safely discarded as they will contribute third order ($\mathcal{O}(k^3)$ in the Fourier space) gradient terms. We postulate that the effective speeds of active transport for polar and nematic orders are identical, i.e. $\bar{U}(\ell_0/D, \phi) \equiv \bar{U}^m = \bar{U}^{\hat{Q}}$.

The conservative interaction stress σ^C is a familiar quantity. In the absence of spatial gradients, its sole contribution is the isotropic pressure arising from interparticle interactions $\sigma^C = -p_C(\ell_0/D, \phi)\mathbf{I}$, which again will simply depend on activity and the particle density. The stress will of course also have gradient terms (the Korteweg stresses). However, these Korteweg stresses, which arise due to the distortion of the pair-distribution function in the presence of density gradients, are significantly smaller than the gradient terms generated by the active stress. Indeed, the nonisotropic contribution to the Korteweg stress (which has the same scale as the isotropic contributions, see main text) was measured in Ref. [9] in order to compute the surface tension of phase-separated ABPs. These Korteweg stresses were found to be negligibly small, scaling as $\sim \zeta U_0 D$ while the active stress gradient terms scale as $\sim \zeta U_0 \ell_0$. As we are interested in scenarios where MIPS occurs (i.e., $\ell_0/D \gg 1$), we neglect the gradient terms arising from the conservative stress such that $\sigma^C = -p_C(\ell_0/D, \phi)\mathbf{I}$.

Finally, we show that including the stress-like contribution of the interparticle forces to the polar order (i.e. Σ^m in the polar order flux [Eq. (S16)]) broadens the binodals predicted by our mechanical theory, yet only inconsequentially. We therefore close our equations by neglecting the stress-like contribution. We first note that our first coexistence criterion for MIPS $\mathcal{P}(p_C^{\text{liq}}) = \mathcal{P}(p_C^{\text{gas}}) = \mathcal{P}^{\text{coexist}}$ is always true regardless of whether we include Σ^m or not, as Σ^m vanishes in regions of homogeneous density. However, since density gradients generate Σ^m , it can in principle alter the second coexistence criterion, the equal-area construction, by altering \mathcal{E} .

From the definition Eq. (S8), it is seen that Σ^m represents the correlation between the interaction stress σ^C and orientation \mathbf{m}/ρ . We examine the effect of the correlation by considering a constitutive relation $\Sigma^m = \xi \sigma^C \mathbf{m}/\rho$. Here, a parameter $\xi(> 0)$ is introduced in order to investigate effects of the magnitude of this term systematically. The effect of Σ^m on the binodal is most clearly seen by considering the following integral:

$$I \equiv \int_{p_C^{\text{gas}}}^{p_C^{\text{liq}}} [\mathcal{P}(p_C) - \mathcal{P}^{\text{coexist}}] dp_C = -\frac{\ell_0}{d-1} \int_{p_C^{\text{gas}}}^{p_C^{\text{liq}}} \frac{d\Sigma_{zzz}^m}{dz} dp_C. \quad (\text{S18})$$

When the correlation term is neglected, the integral I trivially vanishes and the corresponding equal-area construction variable is $\mathcal{E}(\rho) = p_C$ as discussed in the main text. With the simple model $\Sigma^m = \xi \sigma^C \mathbf{m}/\rho$, we find that:

$$I = \frac{\xi \tau_R}{2(d-1)\zeta} \int_{p_C^{\text{gas}}}^{p_C^{\text{liq}}} \left(\frac{dp_C}{dz} \right)^2 \frac{d}{dp_C} \left(\frac{p_C}{\rho} \right) dp_C. \quad (\text{S19})$$

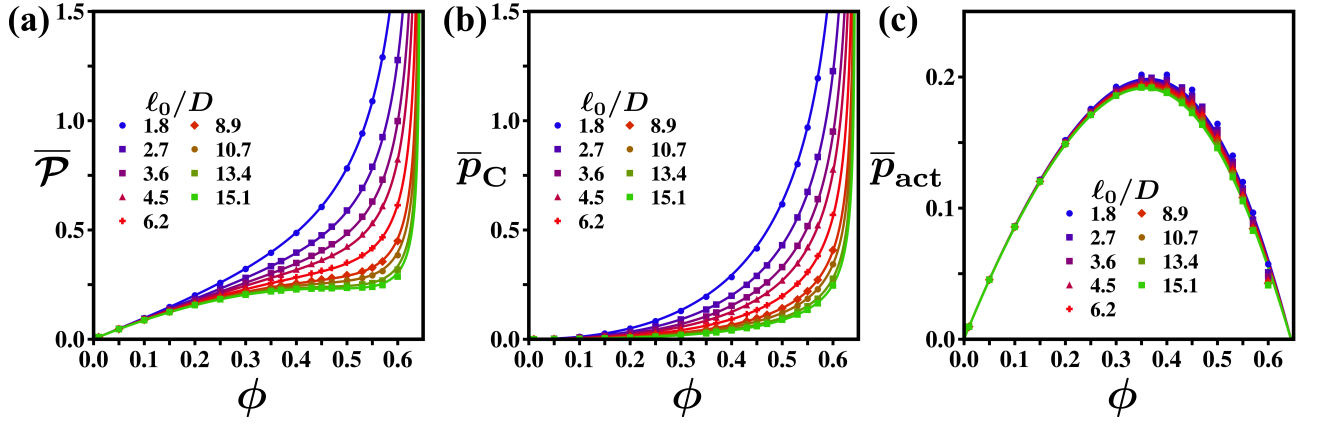


FIG. S2. Comparison between simulation data for active Brownian spheres and derived equations-of-state [Eq. (S20)] as a function of volume fraction ϕ . We observe excellent agreement with simulation data for the (a) dynamical pressure, (b) interaction pressure, and (c) active pressure for a range of run lengths ℓ_0/D approaching the critical activity ℓ_0^c . All pressures have been made dimensionless using the active energy scale $\zeta U_0 \ell_0 / D^3$ to highlight the collapse and linear behavior of \bar{p}_{act} and \bar{P} at low volume fractions.

It can be easily seen that the integrand in Eq. (S19) is always non-negative. Consequently, the integral $I > 0$ and this implies that the coexistence effective pressure $\mathcal{P}^{\text{coexist}}$ is reduced upon including Σ^m [see Fig. S1(a)]. Accordingly, the difference between the coexisting densities ρ^{liq} and ρ^{gas} increases as shown in Fig. S1(b). This is because ρ^{gas} decreases with $\mathcal{P}^{\text{coexist}}$ more rapidly than ρ^{liq} does due to the larger $dp_C/d\rho$ in the liquid phase. Figure S1(b) shows that the coexistence curve is not modified significantly by Σ^m even when its magnitude (ξ) is large, which allows us to close equations by discarding the term.

With our closures, only two quantities are required to describe athermal ABP phase coexistence: the effective active speed $U_0 \bar{U}(\ell_0/D, \phi)$ (or equivalently the active pressure) and the conservative interaction pressure $p_C(\ell_0/D, \phi)$. Accurate equations-of-state in 2d were developed in Ref. [10], and a detailed derivation of those expressions and a comparison to simulation data can be found in the main text and supplementary material of that work. A similar procedure to that utilized in Ref. [10] can be used to obtain equations-of-state for active Brownian spheres in three dimensions. The functional form of these expressions are:

$$\frac{p_{act}}{\zeta U_0 / (\pi D^2)} = \phi \left(\frac{\ell_0}{D} \right) \bar{U} = \phi \left(\frac{\ell_0}{D} \right) \left[1 + \left(1 - \exp \left[-2^{7/6} \left(\frac{\ell_0}{D} \right) \right] \right) \frac{\phi}{1 - \phi/\phi_{\max}} \right]^{-1}, \quad (\text{S20a})$$

$$\frac{p_C}{\zeta U_0 / (\pi D^2)} = 6 \times 2^{-7/6} \frac{\phi^2}{\sqrt{1 - \phi/\phi_{\max}}}, \quad (\text{S20b})$$

where $\phi_{\max} = 0.645$ is the maximum random packing fraction achieved in 3d from the simulations, $D = 2^{1/6} \sigma_{\text{LJ}}$ is the hard-sphere-like diameter, and σ_{LJ} is the Lennard-Jones diameter. Figure S2 provides a comparison between simulation data obtained using Brownian dynamics and the equations-of-state above.

MECHANICAL DERIVATION OF THE SPINODAL CONDITION

The primary focus of the main text is to obtain the coexistence criteria for mechanically determining the binodal without invoking thermodynamic arguments. Here, we show that the spinodal – the region of a phase diagram in which a homogeneous density profile is unstable – can also be determined mechanically without invoking thermodynamic stability arguments. The temporal description of a density profile is provided by the continuity equation:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \mathbf{j}^\rho = 0, \quad (\text{S21})$$

where we now require an expression for the number density flux $\mathbf{j}^\rho(\mathbf{x}; t) = \rho(\mathbf{x}; t) \mathbf{u}(\mathbf{x}, t)$ which follows from conservation of linear momentum:

$$\frac{\partial (m \mathbf{j}_\rho)}{\partial t} + \nabla \cdot (m \mathbf{j}^\rho \mathbf{j}^\rho / \rho) = \nabla \cdot \boldsymbol{\sigma} + \mathbf{b}, \quad (\text{S22})$$

where m is the particle mass. Let us consider a *passive* system initially at rest $\mathbf{u}(\mathbf{x}, t_0) = \mathbf{0}$ with an initially homogeneous density profile $\rho(\mathbf{x}, t_0) = \rho_0$. We now consider small perturbations to the density and velocity fields such that $\rho(\mathbf{x}) = \rho_0 + \delta\rho(\mathbf{x})$ and $\mathbf{u}(\mathbf{x}) = \delta\mathbf{u}(\mathbf{x})$. Substituting the perturbed density and velocity fields into Eq. (S21) and (S22) and retaining only terms linear in these perturbations results in:

$$\frac{\partial\delta\rho}{\partial t} + \rho_0 \nabla \cdot \delta\mathbf{u} = 0, \quad (\text{S23a})$$

$$m\rho_0 \frac{\partial\delta\mathbf{u}}{\partial t} = \nabla \cdot \boldsymbol{\sigma}, \quad (\text{S23b})$$

where we have, for now, neglected body forces. We require an expression for $\boldsymbol{\sigma}$ to describe the evolution of the density perturbations. As our focus will be on the behavior of long wavelength perturbations, we omit the spatial gradient terms (e.g., the Korteweg stress or viscous stresses), resulting in $\boldsymbol{\sigma} = -p(\rho)\mathbf{I}$. The divergence of the stress can now be expressed as $\nabla \cdot \boldsymbol{\sigma} = -(\partial p/\partial\rho)\nabla\delta\rho$ where the compressibility $(\partial p/\partial\rho)$ is to be evaluated at ρ_0 . Differentiating Eq. (S23a) with respect to time and substituting in Eq. (S23b) we arrive at:

$$\frac{\partial^2\delta\rho}{\partial t^2} = \frac{1}{m} \left(\frac{\partial p}{\partial\rho} \right)_{\rho=\rho_0} \nabla^2\delta\rho, \quad (\text{S24})$$

which we recognize as a wave equation for $\delta\rho$ with the wave speed c given by $c^2 = (\partial p/\partial\rho)/m$. Spatially Fourier transforming Eq. (S24) we arrive at:

$$\frac{\partial^2\delta\rho_k}{\partial t^2} = -(ck)^2\delta\rho_k, \quad (\text{S25})$$

where k is the magnitude of the wavevector \mathbf{k} and $\delta\rho_k(\mathbf{k}, t)$ is the Fourier-transformed density perturbation. Equation (S25) admits a plane-wave solution with:

$$\delta\rho_k = A_k \exp[-ickt] + B_k \exp[ickt], \quad (\text{S26})$$

where A_k and B_k are to-be-determined constants. Clearly, if c is imaginary, the Fourier modes of the density perturbations will grow in time and a homogeneous density is linearly unstable. This condition only occurs when $(\partial p/\partial\rho) < 0$, thus recovering the same spinodal condition as expected from thermodynamic stability while using only mechanical arguments.

In the case of driven systems, internal body forces $\mathbf{b}(\mathbf{x}, t)$ may be generated. These internal body forces often have their own evolution equations. However, there is often a separation of timescales between the relaxation dynamics of the density field and the dynamics of active body forces. Indeed, in the case of ABPs, the body force arises due to the polarization density of the active force which relaxes on a timescale proportional τ_R [see Eq. (S6)]. With a separation of timescales (e.g., for timescales large compared to τ_R in the case of ABPs), we can ignore the dynamics of the body force and, just as in the case of statics (see main text), define an effective stress $\boldsymbol{\Sigma}$ which incorporates the effects of the body force (for ABPs, $\boldsymbol{\Sigma} = \boldsymbol{\sigma}^C + \boldsymbol{\sigma}^{\text{act}}$). Provided that the dynamics of the body force permit the use of this dynamic stress in nonstationary conditions, the analysis applied above for passive systems can be repeated and will result in the spinodal condition of $(\partial\mathcal{P}/\partial\rho) < 0$ with the dynamic pressure ($\boldsymbol{\Sigma} = -\mathcal{P}\mathbf{I}$) now playing a determining role.

In unsteady conditions, the active force density is not the only body force in our model. The drag force density also acts as a body force, altering the equation-of-motion of the velocity field from Eq. (S23b) to:

$$m\rho_0 \frac{\partial\delta\mathbf{u}}{\partial t} = \nabla \cdot \boldsymbol{\Sigma} - \zeta\rho_0\delta\mathbf{u}, \quad (\text{S27})$$

where ζ is the translational drag coefficient and we now use the dynamic stress. We can again take a time derivative of Eq. (S23a) and now, using Eq. (S27), obtain:

$$\frac{\partial^2\delta\rho}{\partial t^2} - \frac{1}{\tau_p} \frac{\partial\delta\rho}{\partial t} = \frac{1}{m} \left(\frac{\partial\mathcal{P}}{\partial\rho} \right)_{\rho=\rho_0} \nabla^2\delta\rho, \quad (\text{S28})$$

where $\tau_p = m/\zeta$ is the momentum relaxation time. Equation (S28) is a telegraph equation. Considering timescales much larger than the momentum relaxation time (i.e., $\tau_p \rightarrow 0$) results in a diffusion equation:

$$\frac{\partial\delta\rho}{\partial t} = \frac{1}{\zeta} \left(\frac{\partial\mathcal{P}}{\partial\rho} \right)_{\rho=\rho_0} \nabla^2\delta\rho. \quad (\text{S29})$$

We identify the diffusion coefficient as $D = (\partial\mathcal{P}/\partial\rho)/\zeta$. The Fourier space diffusion equation follows as:

$$\frac{\partial\delta\rho_k}{\partial t} = -Dk^2\delta\rho, \quad (\text{S30})$$

with a solution of:

$$\delta\rho_k = A_k \exp[-Dk^2t]. \quad (\text{S31})$$

Thus, density perturbations will be linearly unstable only when $D < 0$, again resulting in the same spinodal condition of $(\partial\mathcal{P}/\partial\rho) < 0$. Finally, we note that it is also straightforward to show that the same spinodal condition is also recovered using Eq. (S28) without taking the overdamped limit.

DERIVATION OF MECHANICAL FORCE BALANCE BY USING THERMODYNAMIC VARIATIONAL PRINCIPLE

We present steps for deriving the mechanical force balance equation (Eq. (6) in the main text) by using the thermodynamic variational principle. Let us consider the free energy functional presented in Eq. (5) of the main text: $\mathcal{F}[\rho] = \int_V \left[f + \rho\mathcal{U}^{\text{ext}} + \kappa|\nabla\rho|^2/2 \right] d\mathbf{x}$, where $f(\rho)$ is the mean-field free energy density, $\kappa(\rho)$ is a (positive) coefficient such that the square-gradient term penalizes density gradients [11], and $\mathcal{U}^{\text{ext}}(\mathbf{x})$ represents all externally applied potential fields. At constant (N, V, T) , the free energy is minimized with respect to any unconstrained internal variables (in this case the density field) when:

$$\frac{\delta F}{\delta\rho} = \frac{\partial f}{\partial\rho} + \mathcal{U}^{\text{ext}} - \frac{1}{2} \frac{\partial\kappa}{\partial\rho} |\nabla\rho|^2 - \kappa\nabla^2\rho = \mu^{\text{coexist}} \text{ (constant)}, \quad (\text{S32})$$

where the constant (coexistence) chemical potential μ^{coexist} enters as a Lagrange multiplier enforcing the constraint $\int_V \rho d\mathbf{x} = N$. Equation (S32) simply states that chemical potential should be constant throughout the volume at equilibrium. We now cast this thermodynamic equilibrium condition in the form of a mechanical force balance. To do so, we recognize that the external force density per volume, i.e. $-\rho\nabla\mathcal{U}^{\text{ext}}$, should be present in the final mechanical force balance. This motivates us to multiply Eq. (S32) with $\nabla\rho$ and we obtain:

$$-\rho\nabla\mathcal{U}^{\text{ext}} + \nabla \cdot \left[\left(f + \rho\mathcal{U}^{\text{ext}} - \mu\rho + \frac{1}{2}\kappa|\nabla\rho|^2 \right) \mathbf{I} - \kappa\nabla\rho\nabla\rho \right] = \mathbf{0}. \quad (\text{S33})$$

Using Eq. (S32) and a thermodynamic relation $p(\rho) = -f(\rho) + \rho\partial f/\partial\rho$, we finally obtain:

$$-\rho\nabla\mathcal{U}^{\text{ext}} + \nabla \cdot \left[-p\mathbf{I} + \left(\frac{1}{2} \frac{\partial(\kappa\rho)}{\partial\rho} |\nabla\rho|^2 + \kappa\rho\nabla^2\rho \right) \mathbf{I} - \kappa\nabla\rho\nabla\rho \right] = \mathbf{0}. \quad (\text{S34})$$

Equation (S34) is the mechanical force balance in equilibrium – the externally imposed body force $\mathbf{b} = -\rho\nabla\mathcal{U}^{\text{ext}}$ is balanced by the divergence of stress $\boldsymbol{\sigma} = -p\mathbf{I} + \left(\partial(\kappa\rho)/\partial\rho |\nabla\rho|^2/2 + \kappa\rho\nabla^2\rho \right) \mathbf{I} - \kappa\nabla\rho\nabla\rho$.

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