Supplementary Information – Theory of Nonequilibrium Crystallization and the Phase Diagram of Active Brownian Spheres

Daniel Evans and Ahmad K. Omar*

Department of Materials Science and Engineering,
University of California, Berkeley, California 94720, USA and
Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA

CONTENTS

Microscopic Derivation of the Evolution of the Density and Crystallinity Fields 1
Equations of Motion and Fokker-Planck Operator 2
Steady-State Mechanics of the Density Field 2
Steady-State Mechanics of the Crystallinity Field 4

Derivation of Coexistence Criteria 8

Equations of State of Active Brownian Spheres 12
Physical and Semi-Empirical Bulk Equations of State 12
Characterization of the "Pseudo"-spinodal 15

References 16

MICROSCOPIC DERIVATION OF THE EVOLUTION OF THE DENSITY AND CRYSTALLINITY FIELDS

In this Section, we derive the dynamics of the ensemble-averaged density and crystallinity fields to identify the steady-state force balance governing each field. We derive the density field dynamics in an identical manner as in Ref. [1] and use a similar approach to derive the crystallinity field dynamics.

Equations of Motion and Fokker-Planck Operator

We consider N interacting 3D active Brownian particles, where the position, $\mathbf{r}_i$, and orientation, $\mathbf{q}_i$, of the i^{th} particle undergo the following equations of motion:

$$\dot{\mathbf{r}}_i = U_0\mathbf{q}_i + \frac{1}{\zeta}\mathbf{F}_i^C, \tag{S1a}$$

$$\dot{\mathbf{q}}_i = \boldsymbol{\Omega}_i \times \mathbf{q}_i, \tag{S1b}$$

where $\dot{a} \equiv \partial a/\partial t$, U_0 is the active speed, ζ is the drag coefficient, $\mathbf{F}_i^C = \sum_{j\neq i}\mathbf{F}_{ij}$ is the sum of conservative interparticle forces on particle i, and $\boldsymbol{\Omega}_i$ is a random angular velocity with zero mean and variance $\langle\boldsymbol{\Omega}_i(t)\boldsymbol{\Omega}_j(t')\rangle = 2\tau_R^{-1}\delta_{ij}\delta(t-t')\mathbf{I}$ where τ_R is the characteristic reorientation time, δ_{ij} is the Kronecker delta, $\delta(t)$ is the Dirac delta function, and $\mathbf{I}$ is the identity tensor. Here, this mean and variance are taken over the noise statistics.

The noise-averaged N-body distribution function, P_N, evolves according to $\dot{P}_N = \mathcal{L}P_N$ where the Fokker-Planck operator, $\mathcal{L}$, is:

$$\mathcal{L} \equiv -\sum_i \left[\frac{\partial}{\partial\mathbf{r}_i}\cdot\left(U_0\mathbf{q}_i + \frac{1}{\zeta}\mathbf{F}_i^C\right) - \tau_R^{-1}\boldsymbol{\nabla}_i^R\cdot\boldsymbol{\nabla}_i^R\right], \tag{S2}$$

where $\boldsymbol{\nabla}_i^R \equiv \mathbf{q}_i \times \partial/\partial\mathbf{q}_i$ is the rotational gradient operator. The adjoint to this Fokker-Planck operator, $\mathcal{L}^\dagger$, is:

$$\mathcal{L}^\dagger \equiv \sum_i \left[\left(U_0\mathbf{q}_i + \frac{1}{\zeta}\mathbf{F}_i^C\right)\cdot\frac{\partial}{\partial\mathbf{r}_i} + \tau_R^{-1}\boldsymbol{\nabla}_i^R\cdot\boldsymbol{\nabla}_i^R\right]. \tag{S3}$$

This adjoint allows one to express the evolution of an observable $\mathcal{O} \equiv \left\langle\hat{\mathcal{O}}\right\rangle$ as $\dot{\mathcal{O}} = \left\langle\mathcal{L}^\dagger\hat{\mathcal{O}}\right\rangle$, where $\hat{\mathcal{O}}$ is the microscopic definition of the observable. Here, expectations are now taken over the N-body distribution, $\left\langle\hat{\mathcal{O}}\right\rangle = \int_\gamma d\boldsymbol{\Gamma} P_N\hat{\mathcal{O}}$ where $\boldsymbol{\Gamma} = \{\mathbf{r}^N, \mathbf{q}^N\}$ contains all N positions and N orientations and γ is the phase space volume.

Steady-State Mechanics of the Density Field

We now consider the evolution of the density field, $\rho(\mathbf{x},t) \equiv \langle\sum_i\delta(\mathbf{x}-\mathbf{r}_i)\rangle$. This derivation of the density field dynamics was previously performed in Ref. [1], using the same closures, approximations, and constitutive relations that we employ here. While a detailed discussion of this derivation and the nature of the approximations and closures can be found in Ref. [1], we briefly recapitulate the essence of the derivation here for convenience.

Using $\mathcal{L}^\dagger$, we find the dynamics of ρ to be:

$$\dot{\rho} = \left\langle \mathcal{L}^\dagger \sum_i \delta(\mathbf{x} - \mathbf{r}_i) \right\rangle = -\boldsymbol{\nabla} \cdot \left(U_0 \mathbf{m} + \frac{1}{\zeta} \boldsymbol{\nabla} \cdot \boldsymbol{\sigma}_C \right), \tag{S4}$$

where we have defined the polar order field, $\mathbf{m}(\mathbf{x}, t) \equiv \langle \sum_i \mathbf{q}_i \delta(\mathbf{x} - \mathbf{r}_i) \rangle$, and the conservative stress, $\boldsymbol{\sigma}_C \equiv \frac{1}{2} \left\langle \sum_i \sum_{j \neq i} \mathbf{r}_{ij} \mathbf{F}_{ij} b_{ij} \right\rangle$ with the distance vector $\mathbf{r}_{ij} \equiv \mathbf{r}_j - \mathbf{r}_i$ and bond function $b_{ij} \equiv \int_0^1 d\lambda \delta(\mathbf{x} - \mathbf{r}_j + \lambda \mathbf{r}_{ij})$.

The polar order field obeys its own evolution equation. Before examining the resulting equation, we introduce an approximation in the dynamics of *all* fields whose microscopic definition includes at least one orientation: the conservative interactions act to reduce the effective active speed from its ideal value U_0 to a field-dependent value. This dependence will ultimately require additional constitutive equations (see Ref. [1]). Using this approximation, the evolution of the polar order field is:

$$\dot{\mathbf{m}} = \left\langle \mathcal{L}^\dagger \sum_i \mathbf{q}_i \delta(\mathbf{x} - \mathbf{r}_i) \right\rangle = -\boldsymbol{\nabla} \cdot \left(U_0 \overline{U}^{\mathbf{m}} \tilde{\mathbf{Q}} \right) - \frac{2}{\tau_R} \mathbf{m}, \tag{S5}$$

where we have introduced the nematic order field $\tilde{\mathbf{Q}} \equiv \langle \sum_i \mathbf{q}_i \mathbf{q}_i \delta(\mathbf{x} - \mathbf{r}_i) \rangle$ and the renormalized active speed of the polar order field $U_0 \overline{U}^{\mathbf{m}}$. The nematic order field undergoes its own evolution equation:

$$\dot{\tilde{\mathbf{Q}}} = \left\langle \mathcal{L}^\dagger \sum_i \mathbf{q}_i \mathbf{q}_i \delta(\mathbf{x} - \mathbf{r}_i) \right\rangle = -\boldsymbol{\nabla} \cdot \left(U_0 \overline{U}^{\tilde{\mathbf{Q}}} \tilde{\mathbf{B}} \right) - \frac{6}{\tau_R} \left(\tilde{\mathbf{Q}} - \frac{\rho}{3} \mathbf{I} \right), \tag{S6}$$

where we have introduced the third orientational moment field, $\tilde{\mathbf{B}} \equiv \langle \sum_i \mathbf{q}_i \mathbf{q}_i \mathbf{q}_i \delta(\mathbf{x} - \mathbf{r}_i) \rangle$, and the renormalized active speed of the nematic field $U_0 \overline{U}^{\mathbf{Q}}$. Notably, the traceless nematic order parameter, defined as $\mathbf{Q} \equiv \tilde{\mathbf{Q}} - \rho \mathbf{I}/3$, has the following dynamics:

$$\dot{\mathbf{Q}} = -\boldsymbol{\nabla} \cdot \left(U_0 \overline{U}^{\tilde{\mathbf{Q}}} \tilde{\mathbf{B}} \right) - \frac{1}{3} \mathbf{I} \dot{\rho} - \frac{6}{\tau_R} \mathbf{Q} = -\boldsymbol{\nabla} \cdot \left(U_0 \overline{U}^{\tilde{\mathbf{Q}}} \tilde{\mathbf{B}} - \frac{1}{3} \mathbf{I} U_0 \mathbf{m} - \frac{1}{3\zeta} \boldsymbol{\nabla} \cdot \boldsymbol{\sigma}_C \right) - \frac{6}{\tau_R} \mathbf{Q}. \tag{S7}$$

While $\tilde{\mathbf{B}}$ undergoes its own equation of motion, we define $\tilde{\mathbf{B}} = \mathbf{B} + \boldsymbol{\alpha} \cdot \mathbf{m}/5$ (where $\boldsymbol{\alpha}$ is an isotropic fourth rank tensor with $\alpha_{ijkl} = \delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}$ in Einstein notation) and use the closure $\mathbf{B} = \mathbf{0}$. This closure allows us to simply express the third orientational moment with the polar order field. The evolution of the nematic order field is then:

$$\dot{\mathbf{Q}} = -\boldsymbol{\nabla} \cdot \left(U_0 \mathbf{I} \mathbf{m} \left(\frac{3}{5} \overline{U}^{\tilde{\mathbf{Q}}} - \frac{1}{3} \right) - \frac{1}{3\zeta} \boldsymbol{\nabla} \cdot \boldsymbol{\sigma}_C \right) - \frac{6}{\tau_R} \mathbf{Q}. \tag{S8}$$

Inserting the steady-state solution to Eq. (S5) into Eq. (S4) we have:

$$\dot{\rho} = -\boldsymbol{\nabla} \cdot \left(\frac{1}{\zeta} \boldsymbol{\nabla} \cdot \boldsymbol{\sigma}_{\text{act}} + \frac{1}{\zeta} \boldsymbol{\nabla} \cdot \boldsymbol{\sigma}_C \right), \tag{S9}$$

where we have defined the active stress $\boldsymbol{\sigma}_{\rm act} \equiv -\zeta U_0 \ell_0 \overline{U}^{\mathbf{m}} \tilde{\mathbf{Q}}/2$ with the run length $\ell_0 \equiv U_0 \tau_R$.

We now explicitly look for the quasi-1D dynamics in the z-direction. Defining $\mathcal{P} \equiv -\sigma^{zz}_{\rm act} + p_C$ where $p_C \equiv -\sigma^{zz}_C$ and assuming the polar and nematic order fields relax faster than the density field (quasi-static density field dynamics), we find:

$$\dot{\rho} = \frac{\partial}{\partial z}\left(\frac{1}{\zeta}\frac{\partial}{\partial z}\mathcal{P}\right) = -\frac{\partial}{\partial z}\left(L f_\rho\right), \tag{S10a}$$

and identify $J_\rho = L f_\rho = -\zeta \partial \mathcal{P}/\partial z$, mapping these dynamics to Ref. [2]. We see that $L = \zeta^{-1}$ and therefore find the flux-driving force to be:

$$f_\rho = -\frac{\partial}{\partial z}\mathcal{P} = -\frac{\partial}{\partial z}\left[p_C + p^{\rm bulk}_{\rm act} - \frac{\ell_0^2 \overline{U}}{20}\frac{\partial}{\partial z}\left(\overline{U}\frac{\partial p^{\rm bulk}_C}{\partial z}\right)\right], \tag{S10b}$$

where we have defined the active pressure $p^{\rm bulk}_{\rm act} \equiv \zeta U_0 \ell_0 \overline{U} \rho/6$ and, in line with our quasi-static approximation, have substituted the steady-state solution to Eq. (S8), the steady-state solution $\zeta U_0 m_z = -d\sigma^{zz}/dz$, and the approximation $\overline{U}^{\mathbf{m}} = \overline{U}^{\tilde{\mathbf{Q}}} = \overline{U}$ into $\mathcal{P}$ and truncated at second order in spatial gradients. Equation (S10b) immediately yields the "chemical pseudopotential" $u_\rho = \mathcal{P}$ with $\mathcal{T}_{\rho\rho} = -1$ and $\mathcal{T}_{\rho\psi} = 0$, noting that $\partial/\partial z \to d/dz$ in a steady-state.

Steady-State Mechanics of the Crystallinity Field

We now consider the evolution of a crystallinity field, $\psi_V(\mathbf{x}, t) \equiv \langle \sum_i \psi_i \delta(\mathbf{x} - \mathbf{r}_i)\rangle$, where ψ_i provides a measure of the local crystalline order around particle i that in principle can depend on the distance vectors $\mathbf{r}_{ij}$ between i and all other particles j. This derivation has not been performed previously to our knowledge, however we aim to parallel the derivation of the density field dynamics when possible. Using $\mathcal{L}^\dagger$ we find:

$$\dot{\psi}_V = \left\langle \mathcal{L}^\dagger \sum_i \psi_i \delta(\mathbf{x} - \mathbf{r}_i)\right\rangle = -\boldsymbol{\nabla}\cdot\mathbf{J}_\psi + U_0 m^\psi + s_C, \tag{S11}$$

where we have defined $m^\psi \equiv \left\langle \sum_i \sum_j \mathbf{q}_j \cdot \frac{\partial \psi_i}{\partial \mathbf{r_j}} \delta(\mathbf{x} - \mathbf{r}_i)\right\rangle$, $s_C \equiv \frac{1}{\zeta}\left\langle \sum_i \sum_j \mathbf{F}^C_j \cdot \frac{\partial \psi_i}{\partial \mathbf{r_j}} \delta(\mathbf{x} - \mathbf{r}_i)\right\rangle$, and the crystallinity flux $\mathbf{J}_\psi$:

$$\mathbf{J}_\psi \equiv \left\langle \sum_i \left(U_0 \mathbf{q}_i + \frac{1}{\zeta}\mathbf{F}^C_i\right)\delta\left(\mathbf{x} - \mathbf{r}_i\right)\right\rangle. \tag{S12}$$

While the flux is clearly not identically zero, we nevertheless approximate it as negligible in comparison to the generation terms, $|\boldsymbol{\nabla}\cdot\mathbf{J}_\psi| << |s_\psi|$. This results in model A dynamics for the crystallinity field and allows us to apply our recently proposed coexistence framework [2].

As was the case in the previous section, m^ψ undergoes its own evolution equation:

$$\dot{m}^\psi = \left\langle \mathcal{L}^\dagger \sum_i \sum_j \mathbf{q}_j \cdot \frac{\partial \psi_i}{\partial \mathbf{r_j}} \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle = -\boldsymbol{\nabla} \cdot \left(U_0 \overline{U}_{\mathbf{J}}^{m^\psi} \tilde{\mathbf{Q}}_1^\psi \right) + U_0 \overline{U}_s^{m^\psi} \tilde{Q}_2^\psi - \frac{2}{\tau_R} m^\psi, \tag{S13}$$

where we have defined:

$$\tilde{\mathbf{Q}}_1^\psi = \left\langle \sum_i \sum_j \mathbf{q}_i \mathbf{q}_j \cdot \frac{\partial \psi_i}{\partial \mathbf{r}_j} \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle, \tag{S14a}$$

$$\tilde{Q}_2^\psi = \left\langle \sum_i \sum_j \sum_k \mathbf{q}_j \cdot \frac{\partial^2 \psi_i}{\partial \mathbf{r}_j \partial \mathbf{r}_k} \cdot \mathbf{q}_k \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle, \tag{S14b}$$

and the renormalized active speeds $U_0 \overline{U}_{\mathbf{J}}^{m^\psi}$ and $U_0 \overline{U}_s^{m^\psi}$.

We now seek expressions for the evolution of $\tilde{\mathbf{Q}}_1^\psi$ and $\tilde{Q}_2^\psi$. Beginning with $\tilde{Q}_2^\psi$:

$$\begin{aligned}\dot{\tilde{Q}}_2^\psi &= \left\langle \mathcal{L}^\dagger \sum_i \sum_j \sum_k \mathbf{q}_j \cdot \frac{\partial^2 \psi_i}{\partial \mathbf{r}_j \partial \mathbf{r}_k} \cdot \mathbf{q}_k \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle \\ &= -\boldsymbol{\nabla} \cdot \left(U_0 \overline{U}_{\mathbf{J}}^{\tilde{Q}_2^\psi} \tilde{\mathbf{B}}_2 \right) + U_0 \overline{U}_s^{\tilde{Q}_2^\psi} \tilde{B}_3 - \frac{4}{\tau_R} \tilde{Q}_2^\psi + \frac{2}{\tau_R} \left\langle \sum_i \sum_j \sum_k \frac{\partial}{\partial \mathbf{r}_j} \cdot \frac{\partial \psi_i}{\partial \mathbf{r}_k} \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle \\ &\quad - \frac{1}{\tau_R} \left\langle \sum_i \sum_j \sum_k (\mathbf{q}_j \mathbf{q}_j + \mathbf{q}_k \mathbf{q}_k) : \frac{\partial^2 \psi_i}{\partial \mathbf{r}_j \partial \mathbf{r}_k} \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle, \end{aligned} \tag{S15}$$

where we have defined:

$$\tilde{\mathbf{B}}_2^\psi = \left\langle \sum_i \sum_j \sum_k \mathbf{q}_i \mathbf{q}_j \cdot \frac{\partial^2 \psi_i}{\partial \mathbf{r}_j \partial \mathbf{r}_k} \cdot \mathbf{q}_k \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle, \tag{S16a}$$

$$\tilde{B}_3^\psi = \left\langle \sum_i \sum_j \sum_k \sum_l \mathbf{q}_j \mathbf{q}_k \mathbf{q}_l \vdots \frac{\partial^3 \psi_i}{\partial \mathbf{r}_j \partial \mathbf{r}_k \partial \mathbf{r}_l} \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle, \tag{S16b}$$

along with the renormalized active speeds $U_0 \overline{U}_{\mathbf{J}}^{\tilde{Q}_2^\psi}$ and $U_0 \overline{U}_s^{\tilde{Q}_2^\psi}$. We now recognize that as each ψ_i is a function of the bond vectors $\mathbf{r}_{ij}$, $\partial \psi_i / \partial \mathbf{r}_i = -\sum_{j \neq i} \partial \psi_i / \partial \mathbf{r}_{ij}$ and $\partial \psi_i / \partial \mathbf{r}_j = \partial \psi_i / \partial \mathbf{r}_{ij}$. This implies:

$$\left\langle \sum_i \sum_j \sum_k \frac{\partial}{\partial \mathbf{r}_j} \cdot \frac{\partial \psi_i}{\partial \mathbf{r}_k} \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle = \left\langle \sum_i \sum_j \frac{\partial}{\partial \mathbf{r}_j} \cdot \left(\frac{\partial \psi_i}{\partial \mathbf{r}_i} + \sum_{k \neq i} \frac{\partial \psi_i}{\partial \mathbf{r}_k} \right) \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle = 0, \tag{S17a}$$

and:

$$\begin{aligned} &\left\langle \sum_i \sum_j \sum_k (\mathbf{q}_j \mathbf{q}_j + \mathbf{q}_k \mathbf{q}_k) : \frac{\partial^2 \psi_i}{\partial \mathbf{r}_j \partial \mathbf{r}_k} \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle = \\ &\qquad \left\langle \sum_i \left(\sum_j \mathbf{q}_j \mathbf{q}_j : \frac{\partial}{\partial \mathbf{r}_j} \left[\frac{\partial \psi_i}{\partial \mathbf{r}_i} + \sum_{k \neq i} \frac{\partial \psi_i}{\partial \mathbf{r}_k} \right] \right.\right. \\ &\qquad\qquad \left.\left. + \sum_k \mathbf{q}_k \mathbf{q}_k : \frac{\partial}{\partial \mathbf{r}_k} \left[\frac{\partial \psi_i}{\partial \mathbf{r}_i} + \sum_{j \neq i} \frac{\partial \psi_j}{\partial \mathbf{r}_k} \right] \right) \delta(\mathbf{x}-\mathbf{r}_i) \right\rangle = 0, \end{aligned} \tag{S17b}$$

and hence:

$$\dot{\tilde{Q}}_2^{\psi} = -\boldsymbol{\nabla}\cdot\left(U_0\overline{U}_{\mathbf{J}}^{\tilde{Q}_2^{\psi}}\tilde{\mathbf{B}}_2\right) + U_0\overline{U}_s^{\tilde{Q}_2^{\psi}}\tilde{B}_3 - \frac{4}{\tau_R}\tilde{Q}_2^{\psi}. \tag{S18}$$

We examine the evolution of $\tilde{\mathbf{Q}}_1^{\psi}$:

$$\begin{aligned}
\dot{\tilde{\mathbf{Q}}}_1^{\psi} &= \left\langle \mathcal{L}^{\dagger}\sum_i\sum_j \mathbf{q}_i\mathbf{q}_j\cdot\frac{\partial\psi_i}{\partial\mathbf{r}_j}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle \\
&= -\boldsymbol{\nabla}\cdot\left(U_0\overline{U}_{\mathbf{J}}^{\tilde{\mathbf{Q}}_1^{\psi}}\tilde{\mathbf{B}}_1\right) + U_0\overline{U}_s^{\tilde{\mathbf{Q}}_1^{\psi}}\tilde{\mathbf{B}}_2 - \frac{4}{\tau_R}\tilde{\mathbf{Q}}_1^{\psi} + \frac{2}{\tau_R}\left\langle\sum_i\sum_j\frac{\partial\psi_i}{\partial\mathbf{r}_j}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle \\
&\qquad - \frac{1}{\tau_R}\left\langle\sum_i\sum_j(\mathbf{q}_j\mathbf{q}_j+\mathbf{q}_i\mathbf{q}_i)\cdot\frac{\partial\psi_i}{\partial\mathbf{r}_j}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle,
\end{aligned} \tag{S19}$$

where we have defined:

$$\tilde{\mathbf{B}}_1^{\psi} = \left\langle\sum_i\sum_j\mathbf{q}_i\mathbf{q}_i\mathbf{q}_j\cdot\frac{\partial\psi_i}{\partial\mathbf{r}_j}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle, \tag{S20}$$

and the renormalized active speeds $U_0\overline{U}_{\mathbf{J}}^{\tilde{Q}_1^{\psi}}$ and $U_0\overline{U}_s^{\tilde{Q}_1^{\psi}}$. We again find:

$$\left\langle\sum_i\sum_j\frac{\partial\psi_i}{\partial\mathbf{r}_j}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle = \left\langle\sum_i\left(\frac{\partial\psi_i}{\partial\mathbf{r}_i}+\sum_{j\neq i}\frac{\partial\psi_i}{\partial\mathbf{r}_j}\right)\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle = 0. \tag{S21}$$

Contrasting this, we find:

$$\begin{aligned}
&\left\langle\sum_i\sum_j(\mathbf{q}_j\mathbf{q}_j+\mathbf{q}_i\mathbf{q}_i)\cdot\frac{\partial\psi_i}{\partial\mathbf{r}_j}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle \\
&\qquad = \left\langle\sum_i\left[2\mathbf{q}_i\mathbf{q}_i\cdot\frac{\partial\psi_i}{\partial\mathbf{r}_i}+\sum_{j\neq i}(\mathbf{q}_j\mathbf{q}_j+\mathbf{q}_i\mathbf{q}_i)\cdot\frac{\partial\psi_i}{\partial\mathbf{r}_j}\right]\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle \\
&\qquad = \left\langle\sum_i\sum_{j\neq i}(\mathbf{q}_j\mathbf{q}_j-\mathbf{q}_i\mathbf{q}_i)\cdot\frac{\partial\psi_i}{\partial\mathbf{r}_{ij}}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle \neq 0,
\end{aligned} \tag{S22}$$

which makes one unable to obtain closed expressions for the source of ψ_V at this order. To circumvent this, we assume that this vector can be expressed as the product of an isotropic second order tensor and $\left\langle\sum_i\sum_j\frac{\partial\psi_i}{\partial\mathbf{r}_j}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle$ which vanishes. We then have:

$$\dot{\tilde{\mathbf{Q}}}_1^{\psi} = -\boldsymbol{\nabla}\cdot\left(U_0\overline{U}_{\mathbf{J}}^{\tilde{\mathbf{Q}}_1^{\psi}}\tilde{\mathbf{B}}_1\right) + U_0\overline{U}_s^{\tilde{\mathbf{Q}}_1^{\psi}}\tilde{\mathbf{B}}_2 - \frac{4}{\tau_R}\tilde{\mathbf{Q}}_1^{\psi}. \tag{S23}$$

To close these equations, we introduce the following forms for $\tilde{\mathbf{B}}_1^{\psi}$:

$$\tilde{\mathbf{B}}_1^{\psi} = \mathbf{B}_i^{\psi} + \frac{1}{3}\mathbf{I}\left\langle\sum_i\sum_j\mathbf{q}_j\cdot\frac{\partial\psi_i}{\partial\mathbf{r}_j}\delta(\mathbf{x}-\mathbf{r}_i)\right\rangle = \mathbf{B}_1^{\psi} + \frac{1}{3}\mathbf{I}m^{\psi}, \tag{S24a}$$

for $\tilde{\mathbf{B}}_2^\psi$:

$$\tilde{\mathbf{B}}_2^\psi = \mathbf{B}_2^\psi + \left\langle \sum_i \sum_j \sum_k \mathbf{q}_j \cdot \frac{\partial^2 \psi_i}{\partial \mathbf{r}_j \partial \mathbf{r}_k} \delta(\mathbf{x} - \mathbf{r}_i) \right\rangle$$
$$= \mathbf{B}_2^\psi + \left\langle \sum_i \sum_j \mathbf{q}_j \cdot \frac{\partial}{\partial \mathbf{r}_j} \left(\frac{\partial \psi_i}{\partial \mathbf{r}_i} + \sum_{k \neq i} \frac{\partial \psi_i}{\partial \mathbf{r}_k} \right) \delta(\mathbf{x} - \mathbf{r}_i) \right\rangle = \mathbf{B}_2^\psi, \quad \text{(S24b)}$$

and for $\tilde{B}_3^\psi$:

$$\tilde{B}_3^\psi = B_3^\psi + \left\langle \sum_i \sum_j \sum_k \sum_l \mathbf{q}_j \cdot \frac{\partial}{\partial \mathbf{r}_j} \frac{\partial}{\partial \mathbf{r}_k} \cdot \frac{\partial \psi_i}{\partial \mathbf{r}_l} \delta(\mathbf{x} - \mathbf{r}_i) \right\rangle$$
$$= B_3^\psi + \left\langle \sum_i \sum_j \sum_k \mathbf{q}_j \cdot \frac{\partial}{\partial \mathbf{r}_j} \frac{\partial}{\partial \mathbf{r}_k} \cdot \left(\frac{\partial \psi_i}{\partial \mathbf{r}_i} + \sum_{l \neq i} \frac{\partial \psi_i}{\partial \mathbf{r}_l} \right) \delta(\mathbf{x} - \mathbf{r}_i) \right\rangle = B_3^\psi. \quad \text{(S24c)}$$

We then introduce the closures $\mathbf{B}_1^\psi = \mathbf{0}$, $\mathbf{B}_2^\psi = \mathbf{0}$, and $B_3^\psi = 0$ and substitute the result into Eqs. (S18) and (S23), finding:

$$\dot{\tilde{Q}}_2^\psi = -\frac{4}{\tau_R} \tilde{Q}_2^\psi, \quad \text{(S25a)}$$
$$\dot{\tilde{\mathbf{Q}}}_1^\psi = -\boldsymbol{\nabla} \left(U_0 \overline{U}_{\mathbf{J}}^{\tilde{\mathbf{Q}}_1^\psi} \frac{1}{3} m^\psi \right) - \frac{4}{\tau_R} \tilde{\mathbf{Q}}_1^\psi. \quad \text{(S25b)}$$

We now return to Eq. (S11). Solving Eq. (S13) and substituting the result into Eq. (S11) we have:

$$\dot{\psi}_V = -\boldsymbol{\nabla} \cdot \left(\frac{U_0 \ell_0 \overline{U}_{\mathbf{J}}^{m^\psi}}{2} \tilde{\mathbf{Q}}_1^\psi \right) + \frac{U_0 \ell_0 \overline{U}_s^{m^\psi}}{2} \tilde{Q}_2^\psi + s_C \quad \text{(S26)}$$

Substituting the solutions to Eq. (S25) we find:

$$\dot{\psi}_V = \boldsymbol{\nabla} \cdot \left(\frac{\ell_0^2 \overline{U}_{\mathbf{J}}^{m^\psi}}{24} \boldsymbol{\nabla} \left(\overline{U}_{\mathbf{J}}^{\tilde{\mathbf{Q}}_1^\psi} U_0 m^\psi \right) \right) + s_C. \quad \text{(S27)}$$

We now use the steady-state solution $U_0 m^\psi = -s_C$ to find:

$$\dot{\psi}_V = s_C - \boldsymbol{\nabla} \cdot \left(\frac{\ell_0^2 \overline{U}_{\mathbf{J}}^{m^\psi}}{24} \boldsymbol{\nabla} \left(\overline{U}_{\mathbf{J}}^{\tilde{\mathbf{Q}}_1^\psi} s_C \right) \right). \quad \text{(S28)}$$

Moving to a quasi-1D profile oriented along the z-direction, the dynamics of the crystallinity field are:

$$\dot{\psi}_V = M f_\psi = s_C - \frac{\ell_0^2}{24} \frac{\partial}{\partial z} \left(\overline{U} \frac{\partial}{\partial z} \left(\overline{U} s_C^{\text{bulk}} \right) \right), \quad \text{(S29)}$$

where we have truncated at second order in spatial gradients by retaining only the bulk part of the conservative generation ($s_C \equiv s_C^{\text{bulk}} + s_C^{(2)}$) and assumed $\overline{U}_{\mathbf{J}}^{m^\psi} = \overline{U}_{\mathbf{J}}^{\tilde{\mathbf{Q}}_1^\psi} = \overline{U}$. Mapping these dynamics to Ref. [2], we may set $M = 1$ and identify the generation-driving force as:

$$f_\psi = s_C - \frac{\ell_0^2}{24}\frac{\partial}{\partial z}\left(\overline{U}\frac{\partial}{\partial z}\left(\overline{U}s_C^{\text{bulk}}\right)\right), \tag{S30}$$

where $\partial/\partial z \to d/dz$ in a steady-state.

DERIVATION OF COEXISTENCE CRITERIA

With the expressions for f_ρ and f_ψ determined in the previous section, we now derive the nonequilibrium coexistence criteria used in the main text following Ref. [2]. We note that as f_ρ is the spatial derivative of $-\mathcal{P}$, we can immediately identify that the steady-state condition of the conserved field (in flux-free boundary conditions), $f_\rho = 0$, can be expressed as $\mathcal{P} = \text{const}$, i.e., the dynamic pressure must be spatially uniform. In the language of Ref. [2], $\mathcal{P}$ is the pseudopotential of the conserved field, $u_\rho = \mathcal{P}$. As spatial gradients vanish in the bulk coexisting phases, the steady-state conditions $\mathcal{P} = \text{const}$ and $f_\psi = 0$ can be immediately converted to three coexistence criteria:

$$\mathcal{P}^{\text{bulk}}\left(\rho^f, \psi_V^f\right) = \mathcal{P}^{\text{bulk}}\left(\rho^s, \psi_V^s\right) = \mathcal{P}^{\text{coexist}}, \tag{S31a}$$

$$s_C^{\text{bulk}}\left(\rho^f, \psi_V^f\right) = 0, \tag{S31b}$$

$$s_C^{\text{bulk}}\left(\rho^s, \psi_V^s\right) = 0, \tag{S31c}$$

where $\mathcal{P}^{\text{bulk}} \equiv p_C^{\text{bulk}} + p_{\text{act}}^{\text{bulk}}$ and $\mathcal{P}^{\text{coexist}}$ is a constant that must be determined. We therefore seek one additional equation to determine the four variables of interest: ρ^f, ρ^s, ψ_V^f, and ψ_V^s.

In equilibrium, the final criterion is the Maxwell construction. Noting that it is the dynamic pressure that must be spatially uniform (and not just the conservative contribution), the equilibrium Maxwell construction for active solid-fluid coexistence is:

$$\int_{\rho^f}^{\rho^s}\left(\mathcal{P}^{\text{bulk}}(\rho, \psi_V^*) - \mathcal{P}^{\text{coexist}}\right)d\rho^{-1} = 0, \tag{S32}$$

where $\psi_V^*(\rho)$ is defined to satisfy $s_C^{\text{bulk}}(\rho, \psi_V^*) = 0$. Combining this Maxwell construction with Eq. (S31) is what we refer to as the "passive" criteria in the main text – this is what was used in a previous attempt to quantitatively predict active solid-fluid binodals (Ref. [3]). However, as we will show, this integral is generally nonzero and path-dependent as it requires the Gibbs-Duhem relation, $d\mathcal{G} = \rho^{-1}d\mathcal{P} + \psi_N df_\psi$ ($\mathcal{G}$ is the chemical potential in equilibrium), to hold.

Following Ref. [2], we introduce an ansatz of a generalized Gibbs-Duhem relation:

$$\frac{d\mathcal{G}}{dz} = \mathcal{E}_\rho \frac{d\mathcal{P}}{dz} - \mathcal{E}_\psi \frac{df_\psi}{dz}, \tag{S33}$$

where $\mathcal{E}_\rho$ and $\mathcal{E}_\psi$ are "Maxwell construction variables" which depend on ρ and ψ_V but not their spatial gradients while $\mathcal{G}$ is a "global quantity" that depends on both ρ and ψ_V and their spatial gradients. In particular, we expand $\mathcal{G}$ as:

$$\mathcal{G} = \mathcal{G}^{\text{bulk}} - \mathcal{G}_\rho^{(2,2)} \frac{d^2\rho}{dz^2} - \mathcal{G}_\psi^{(2,2)} \frac{d^2\psi_V}{dz^2} - \mathcal{G}_{\rho\rho}^{(2,1)} \left(\frac{d\rho}{dz}\right)^2 - 2\mathcal{G}_{\rho\psi}^{(2,1)} \frac{d\rho}{dz}\frac{d\psi_V}{dz} - \mathcal{G}_{\psi\psi}^{(2,1)} \left(\frac{d\psi_V}{dz}\right)^2, \tag{S34}$$

where $\mathcal{G}^{\text{bulk}}$ and each $\mathcal{G}_i^{(2,2)}$ and $\mathcal{G}_{ij}^{(2,1)}$ for $i,j \in \{\rho,\psi\}$ are equations of state that depend on ρ and ψ_V but not their spatial gradients. When $\mathcal{E}_\rho$, $\mathcal{E}_\psi$, and the coefficients of $\mathcal{G}$ can be determined, $\mathcal{G}$ is spatially uniform in steady-states and $\mathcal{G}^{\text{bulk}}$ is equal in coexisting phases. This final criterion can be expressed as a generalized Maxwell construction:

$$\int_{\mathcal{E}_\rho^f}^{\mathcal{E}_\rho^s} \left(\mathcal{P}^{\text{bulk}}(\rho,\psi_V^*) - \mathcal{P}^{\text{coexist}}\right) d\mathcal{E}_\rho = 0. \tag{S35}$$

We now expand the conservative pressure:

$$p_C = p_C^{\text{bulk}} - p_{C\rho}^{(2,2)} \frac{d^2\rho}{dz^2} - p_{C\psi}^{(2,2)} \frac{d^2\psi_V}{dz^2} - p_{C\rho\rho}^{(2,1)} \left(\frac{d\rho}{dz}\right)^2 - 2p_{C\rho\psi}^{(2,1)} \frac{d\rho}{dz}\frac{d\psi_V}{dz} - p_{C\psi\psi}^{(2,1)} \left(\frac{d\psi_V}{dz}\right)^2, \tag{S36a}$$

and generation:

$$s_C = s_C^{\text{bulk}} - s_{C\rho}^{(2,2)} \frac{d^2\rho}{dz^2} - s_{C\psi}^{(2,2)} \frac{d^2\psi_V}{dz^2} - s_{C\rho\rho}^{(2,1)} \left(\frac{d\rho}{dz}\right)^2 - 2s_{C\rho\psi}^{(2,1)} \frac{d\rho}{dz}\frac{d\psi_V}{dz} - s_{C\psi\psi}^{(2,1)} \left(\frac{d\psi_V}{dz}\right)^2, \tag{S36b}$$

where p_C^{bulk}, s_C^{bulk}, and each $p_{Ci}^{(2,2)}$, $s_{Ci}^{(2,2)}$, $p_{Cij}^{(2,1)}$, and $s_{Cij}^{(2,1)}$ for $i,j \in \{\rho,\psi\}$ are equations of state that depend on ρ and ψ_V but not their spatial gradients. We now define:

$$\mathcal{P}_\rho^{(2,2)} = p_{C\rho}^{(2,2)} + \frac{\ell_0^2\overline{U}^2}{20}\frac{\partial p_C^{\text{bulk}}}{\partial\rho}, \tag{S37a}$$

$$\mathcal{P}_\psi^{(2,2)} = p_{C\psi}^{(2,2)} + \frac{\ell_0^2\overline{U}^2}{20}\frac{\partial p_C^{\text{bulk}}}{\partial\psi_V}, \tag{S37b}$$

$$\mathcal{P}_{\rho\rho}^{(2,1)} = p_{C\rho\rho}^{(2,1)} + \frac{\ell_0^2\overline{U}}{20}\left(\frac{\partial p_C^{\text{bulk}}}{\partial\rho}\frac{\partial\overline{U}}{\partial\rho} + \overline{U}\frac{\partial^2 p_C^{\text{bulk}}}{\partial\rho^2}\right), \tag{S37c}$$

$$\mathcal{P}_{\rho\psi}^{(2,1)} = p_{C\rho\psi}^{(2,1)} + \frac{\ell_0^2\overline{U}}{20}\left(\frac{\partial p_C^{\text{bulk}}}{\partial\psi_V}\frac{\partial\overline{U}}{\partial\rho} + \frac{\partial p_C^{\text{bulk}}}{\partial\rho}\frac{\partial\overline{U}}{\partial\psi_V} + 2\overline{U}\frac{\partial^2 p_C^{\text{bulk}}}{\partial\rho\partial\psi_V}\right), \tag{S37d}$$

$$\mathcal{P}_{\psi\psi}^{(2,1)} = p_{C\psi\psi}^{(2,1)} + \frac{\ell_0^2\overline{U}}{20}\left(\frac{\partial p_C^{\text{bulk}}}{\partial\psi_V}\frac{\partial\overline{U}}{\partial\psi_V} + \overline{U}\frac{\partial^2 p_C^{\text{bulk}}}{\partial\psi_V^2}\right), \tag{S37e}$$

and:

$$s_\rho^{(2,2)} = s_{C\rho}^{(2,2)} + \frac{\ell_0^2\overline{U}}{24}\left(\overline{U}\frac{\partial s_C^{\text{bulk}}}{\partial\rho} + s_C^{\text{bulk}}\frac{\partial\overline{U}}{\partial\rho}\right), \tag{S37f}$$

$$s_{\psi}^{(2,2)} = s_{C\psi}^{(2,2)} + \frac{\ell_0^2 \overline{U}}{24}\left(\overline{U}\frac{\partial s_C^{\rm bulk}}{\partial \psi_V} + s_C^{\rm bulk}\frac{\partial \overline{U}}{\partial \psi_V}\right), \tag{S37g}$$

$$s_{\rho\rho}^{(2,1)} = s_{C\rho\rho}^{(2,1)} + \frac{\ell_0^2}{24}\left[\overline{U}\left(\overline{U}\frac{\partial^2 s_C^{\rm bulk}}{\partial \rho^2} + 2\frac{\partial \overline{U}}{\partial \rho}\frac{\partial s_C^{\rm bulk}}{\partial \rho} + s_C^{\rm bulk}\frac{\partial^2 \overline{U}}{\partial \rho^2}\right) + \frac{\partial \overline{U}}{\partial \rho}\left(\overline{U}\frac{\partial s_C^{\rm bulk}}{\partial \rho} + s_C^{\rm bulk}\frac{\partial \overline{U}}{\partial \rho}\right)\right], \tag{S37h}$$

$$s_{\rho\psi}^{(2,1)} = s_{C\rho\psi}^{(2,1)} + \frac{\ell_0^2}{24}\left[2\overline{U}\left(\overline{U}\frac{\partial^2 s_C^{\rm bulk}}{\partial \rho \partial \psi_V} + \frac{\partial \overline{U}}{\partial \rho}\frac{\partial s_C^{\rm bulk}}{\partial \psi_V} + \frac{\partial \overline{U}}{\partial \psi_V}\frac{\partial s_C^{\rm bulk}}{\partial \rho} + s_C^{\rm bulk}\frac{\partial^2 \overline{U}}{\partial \rho \psi_V}\right)\right. \tag{S37i}$$

$$\left. + \frac{\partial \overline{U}}{\partial \psi_V}\left(\overline{U}\frac{\partial s_C^{\rm bulk}}{\partial \rho} + s_C^{\rm bulk}\frac{\partial \overline{U}}{\partial \rho}\right) + \frac{\partial \overline{U}}{\partial \rho}\left(\overline{U}\frac{\partial s_C^{\rm bulk}}{\partial \psi_V} + s_C^{\rm bulk}\frac{\partial \overline{U}}{\partial \psi_V}\right)\right], \tag{S37j}$$

$$s_{\psi\psi}^{(2,1)} = s_{C\psi\psi}^{(2,1)} + \frac{\ell_0^2}{24}\left[\overline{U}\left(\overline{U}\frac{\partial^2 s_C^{\rm bulk}}{\partial \psi_V^2} + 2\frac{\partial \overline{U}}{\partial \psi_V}\frac{\partial s_C^{\rm bulk}}{\partial \psi_V} + s_C^{\rm bulk}\frac{\partial^2 \overline{U}}{\partial \psi_V^2}\right) + \frac{\partial \overline{U}}{\partial \psi_V}\left(\overline{U}\frac{\partial s_C^{\rm bulk}}{\partial \psi_V} + s_C^{\rm bulk}\frac{\partial \overline{U}}{\partial \psi_V}\right)\right], \tag{S37k}$$

such that:

$$\mathcal{P} = \mathcal{P}^{\rm bulk} - \mathcal{P}_{\rho}^{(2,2)}\frac{d^2\rho}{dz^2} - \mathcal{P}_{\psi}^{(2,2)}\frac{d^2\psi_V}{dz^2} - \mathcal{P}_{\rho\rho}^{(2,1)}\left(\frac{d\rho}{dz}\right)^2 - 2\mathcal{P}_{\rho\psi}^{(2,1)}\frac{d\rho}{dz}\frac{d\psi_V}{dz} - \mathcal{P}_{\psi\psi}^{(2,1)}\left(\frac{d\psi_V}{dz}\right)^2, \tag{S38a}$$

$$f_{\psi} = s_C^{\rm bulk} - s_{\rho}^{(2,2)}\frac{d^2\rho}{dz^2} - s_{\psi}^{(2,2)}\frac{d^2\psi_V}{dz^2} - s_{\rho\rho}^{(2,1)}\left(\frac{d\rho}{dz}\right)^2 - 2s_{\rho\psi}^{(2,1)}\frac{d\rho}{dz}\frac{d\psi_V}{dz} - s_{\psi\psi}^{(2,1)}\left(\frac{d\psi_V}{dz}\right)^2. \tag{S38b}$$

Substituting the expressions for $\mathcal{P}$ and f_{ψ} into the generalized Gibbs-Duhem relation [Eq. (S33)] and evaluating derivatives, we find:

$$\begin{aligned}
&\frac{\partial \mathcal{G}^{\rm bulk}}{\partial \rho}\frac{d\rho}{dz} + \frac{\partial \mathcal{G}^{\rm bulk}}{\partial \psi_V}\frac{d\psi_V}{dz} - \mathcal{G}_{\rho}^{(2,2)}\frac{d^3\rho}{dz^3} - \mathcal{G}_{\psi}^{(2,2)}\frac{d^3\psi_V}{dz^3} - \left(\frac{\partial \mathcal{G}_{\rho}^{(2,2)}}{\partial \rho} + 2\mathcal{G}_{\rho\rho}^{(2,1)}\right)\frac{d^2\rho}{dz^2}\frac{d\rho}{dz} \\
&- \left(\frac{\partial \mathcal{G}_{\rho}^{(2,2)}}{\partial \psi_V} + \mathcal{G}_{\rho\psi}^{(2,1)}\right)\frac{d^2\rho}{dz^2}\frac{d\psi}{dz} - \left(\frac{\partial \mathcal{G}_{\psi}^{(2,2)}}{\partial \rho} + \mathcal{G}_{\rho\psi}^{(2,1)}\right)\frac{d^2\psi_V}{dz^2}\frac{d\rho}{dz} - \left(\frac{\partial \mathcal{G}_{\psi}^{(2,2)}}{\partial \psi_V} + 2\mathcal{G}_{\psi\psi}^{(2,1)}\right)\frac{d^2\psi_V}{dz^2}\frac{d\psi_V}{dz} \\
&- \frac{\partial \mathcal{G}_{\rho\rho}^{(2,1)}}{\partial \rho}\left(\frac{d\rho}{dz}\right)^3 - \frac{\partial \mathcal{G}_{\psi\psi}^{(2,1)}}{\partial \psi_V}\left(\frac{d\psi_V}{dz}\right)^3 - \left(\frac{\partial \mathcal{G}_{\rho\rho}^{(2,1)}}{\partial \psi_V} + \frac{\partial \mathcal{G}_{\rho\psi}^{(2,1)}}{\partial \rho}\right)\left(\frac{d\rho}{dz}\right)^2\frac{d\psi_V}{dz} - \\
&\left(\frac{\partial \mathcal{G}_{\psi\psi}^{(2,1)}}{\partial \rho} + \frac{\partial \mathcal{G}_{\rho\psi}^{(2,1)}}{\partial \psi_V}\right)\left(\frac{d\psi_V}{dz}\right)^2\frac{d\rho}{dz} = \mathcal{E}_{\rho}\left[\frac{\partial \mathcal{P}^{\rm bulk}}{\partial \rho}\frac{d\rho}{dz} + \frac{\partial \mathcal{P}^{\rm bulk}}{\partial \psi_V}\frac{d\psi_V}{dz} - \mathcal{P}_{\rho}^{(2,2)}\frac{d^3\rho}{dz^3} - \mathcal{P}_{\psi}^{(2,2)}\frac{d^3\psi_V}{dz^3}\right. \\
&- \left(\frac{\partial \mathcal{P}_{\rho}^{(2,2)}}{\partial \rho} + 2\mathcal{P}_{\rho\rho}^{(2,1)}\right)\frac{d^2\rho}{dz^2}\frac{d\rho}{dz} - \left(\frac{\partial \mathcal{P}_{\rho}^{(2,2)}}{\partial \psi_V} + \mathcal{P}_{\rho\psi}^{(2,1)}\right)\frac{d^2\rho}{dz^2}\frac{d\psi_V}{dz} - \left(\frac{\partial \mathcal{P}_{\psi}^{(2,2)}}{\partial \rho} + \mathcal{P}_{\rho\psi}^{(2,1)}\right)\frac{d^2\psi_V}{dz^2}\frac{d\rho}{dz} \\
&- \left(\frac{\partial \mathcal{P}_{\psi}^{(2,2)}}{\partial \psi_V} + 2\mathcal{P}_{\psi\psi}^{(2,1)}\right)\frac{d^2\psi_V}{dz^2}\frac{d\psi_V}{dz} - \frac{\partial \mathcal{P}_{\rho\rho}^{(2,1)}}{\partial \rho}\left(\frac{d\rho}{dz}\right)^3 - \frac{\partial \mathcal{P}_{\psi\psi}^{(2,1)}}{\partial \psi_V}\left(\frac{d\psi_V}{dz}\right)^3 \\
&\left. - \left(\frac{\partial \mathcal{P}_{\rho\rho}^{(2,1)}}{\partial \psi_V} + \frac{\partial \mathcal{P}_{\rho\psi}^{(2,1)}}{\partial \rho}\right)\left(\frac{d\rho}{dz}\right)^2\frac{d\psi_V}{dz} - \left(\frac{\partial \mathcal{P}_{\psi\psi}^{(2,1)}}{\partial \rho} + \frac{\partial \mathcal{P}_{\rho\psi}^{(2,1)}}{\partial \psi_V}\right)\left(\frac{d\psi_V}{dz}\right)^2\frac{d\rho}{dz}\right] - \mathcal{E}_{\psi}\left[\frac{\partial s_C^{\rm bulk}}{\partial \rho}\frac{d\rho}{dz} + \frac{\partial s_C^{\rm bulk}}{\partial \psi_V}\frac{d\psi_V}{dz}\right. \\
&- s_{\rho}^{(2,2)}\frac{d^3\rho}{dz^3} - s_{\psi}^{(2,2)}\frac{d^3\psi_V}{dz^3} - \left(\frac{\partial s_{\rho}^{(2,2)}}{\partial \rho} + 2s_{\rho\rho}^{(2,1)}\right)\frac{d^2\rho}{dz^2}\frac{d\rho}{dz} - \left(\frac{\partial s_{\rho}^{(2,2)}}{\partial \psi_V} + s_{\rho\psi}^{(2,1)}\right)\frac{d^2\rho}{dz^2}\frac{d\psi_V}{dz}
\end{aligned}$$

$$- \left(\frac{\partial s_{\psi}^{(2,2)}}{\partial \rho} + s_{\rho\psi}^{(2,1)} \right) \frac{d^2\psi_V}{dz^2} \frac{d\rho}{dz} - \left(\frac{\partial s_{\psi}^{(2,2)}}{\partial \psi_V} + 2 s_{\psi\psi}^{(2,1)} \right) \frac{d^2\psi_V}{dz^2} \frac{d\psi_V}{dz} - \frac{\partial s_{\rho\rho}^{(2,1)}}{\partial \rho} \left(\frac{d\rho}{dz} \right)^3 - \frac{\partial s_{\psi\psi}^{(2,1)}}{\partial \psi_V} \left(\frac{d\psi_V}{dz} \right)^3$$

$$- \left(\frac{\partial s_{\rho\rho}^{(2,1)}}{\partial \psi_V} + \frac{\partial s_{\rho\psi}^{(2,1)}}{\partial \rho} \right) \left(\frac{d\rho}{dz} \right)^2 \frac{d\psi_V}{dz} - \left(\frac{\partial s_{\psi\psi}^{(2,1)}}{\partial \rho} + \frac{\partial s_{\rho\psi}^{(2,1)}}{\partial \psi_V} \right) \left(\frac{d\psi_V}{dz} \right)^2 \frac{d\rho}{dz} \Bigg]. \quad \text{(S39)}$$

Matching the coefficients of each term of order $(d^n\rho/dz^n)^k(d^m\psi_V/dz^m)^l$ (where $n^k + m^l = 1$ or 3), we find a system of partial differential-algebraic equations for $\mathcal{E}_\rho$, $\mathcal{E}_\psi$, and each of the coefficients in $\mathcal{G}$:

$$\frac{\partial \mathcal{G}^{\text{bulk}}}{\partial \rho} = \mathcal{E}_\rho \frac{\partial \mathcal{P}^{\text{bulk}}}{\partial \rho} - \mathcal{E}_\psi \frac{\partial s_C^{\text{bulk}}}{\partial \rho}, \quad \text{(S40a)}$$

$$\frac{\partial \mathcal{G}^{\text{bulk}}}{\partial \psi_V} = \mathcal{E}_\rho \frac{\partial \mathcal{P}^{\text{bulk}}}{\partial \psi_V} - \mathcal{E}_\psi \frac{\partial s_C^{\text{bulk}}}{\partial \psi_V}, \quad \text{(S40b)}$$

$$\mathcal{G}_\rho^{(2,2)} = \mathcal{E}_\rho \mathcal{P}_\rho^{(2,2)} - \mathcal{E}_\psi s_\rho^{(2,2)}, \quad \text{(S40c)}$$

$$\mathcal{G}_\psi^{(2,2)} = \mathcal{E}_\rho \mathcal{P}_\psi^{(2,2)} - \mathcal{E}_\psi s_\psi^{(2,2)}, \quad \text{(S40d)}$$

$$\frac{\partial \mathcal{G}_\rho^{(2,2)}}{\partial \rho} + 2\mathcal{G}_{\rho\rho}^{(2,1)} = \mathcal{E}_\rho \left(\frac{\partial \mathcal{P}_\rho^{(2,2)}}{\partial \rho} + 2\mathcal{P}_{\rho\rho}^{(2,1)} \right) - \mathcal{E}_\psi \left(\frac{\partial s_\rho^{(2,2)}}{\partial \rho} + 2 s_{\rho\rho}^{(2,1)} \right), \quad \text{(S40e)}$$

$$\frac{\partial \mathcal{G}_\rho^{(2,2)}}{\partial \psi_V} + \mathcal{G}_{\rho\psi}^{(2,1)} = \mathcal{E}_\rho \left(\frac{\partial \mathcal{P}_\rho^{(2,2)}}{\partial \psi_V} + \mathcal{P}_{\rho\psi}^{(2,1)} \right) - \mathcal{E}_\psi \left(\frac{\partial s_\rho^{(2,2)}}{\partial \psi_V} + s_{\rho\psi}^{(2,1)} \right), \quad \text{(S40f)}$$

$$\frac{\partial \mathcal{G}_\psi^{(2,2)}}{\partial \rho} + \mathcal{G}_{\rho\psi}^{(2,1)} = \mathcal{E}_\rho \left(\frac{\partial \mathcal{P}_\psi^{(2,2)}}{\partial \rho} + \mathcal{P}_{\rho\psi}^{(2,1)} \right) - \mathcal{E}_\psi \left(\frac{\partial s_\psi^{(2,2)}}{\partial \rho} + s_{\rho\psi}^{(2,1)} \right), \quad \text{(S40g)}$$

$$\frac{\partial \mathcal{G}_\psi^{(2,2)}}{\partial \psi_V} + 2\mathcal{G}_{\psi\psi}^{(2,1)} = \mathcal{E}_\rho \left(\frac{\partial \mathcal{P}_\psi^{(2,2)}}{\partial \psi_V} + 2\mathcal{P}_{\psi\psi}^{(2,1)} \right) - \mathcal{E}_\psi \left(\frac{\partial s_\psi^{(2,2)}}{\partial \psi_V} + 2 s_{\psi\psi}^{(2,1)} \right), \quad \text{(S40h)}$$

$$\frac{\partial \mathcal{G}_{\rho\rho}^{(2,1)}}{\partial \rho} = \mathcal{E}_\rho \frac{\partial \mathcal{P}_{\rho\rho}^{(2,1)}}{\partial \rho} - \mathcal{E}_\psi \frac{\partial s_{\rho\rho}^{(2,1)}}{\partial \rho}, \quad \text{(S40i)}$$

$$\frac{\partial \mathcal{G}_{\psi\psi}^{(2,1)}}{\partial \psi_V} = \mathcal{E}_\rho \frac{\partial \mathcal{P}_{\psi\psi}^{(2,1)}}{\partial \rho} - \mathcal{E}_\psi \frac{\partial s_{\psi\psi}^{(2,1)}}{\partial \psi_V}, \quad \text{(S40j)}$$

$$\frac{\partial \mathcal{G}_{\rho\rho}^{(2,1)}}{\partial \psi_V} + \frac{\partial \mathcal{G}_{\rho\psi}^{(2,1)}}{\partial \rho} = \mathcal{E}_\rho \left(\frac{\partial \mathcal{P}_{\rho\rho}^{(2,1)}}{\partial \psi_V} + \frac{\partial \mathcal{P}_{\rho\psi}^{(2,1)}}{\partial \rho} \right) - \mathcal{E}_\psi \left(\frac{\partial s_{\rho\rho}^{(2,1)}}{\partial \psi_V} + \frac{\partial s_{\rho\psi}^{(2,1)}}{\partial \rho} \right), \quad \text{(S40k)}$$

$$\frac{\partial \mathcal{G}_{\psi\psi}^{(2,1)}}{\partial \rho} + \frac{\partial \mathcal{G}_{\rho\psi}^{(2,1)}}{\partial \psi_V} = \mathcal{E}_\rho \left(\frac{\partial \mathcal{P}_{\psi\psi}^{(2,1)}}{\partial \rho} + \frac{\partial \mathcal{P}_{\rho\psi}^{(2,1)}}{\partial \psi_V} \right) - \mathcal{E}_\psi \left(\frac{\partial s_{\psi\psi}^{(2,1)}}{\partial \rho} + \frac{\partial s_{\rho\psi}^{(2,1)}}{\partial \psi_V} \right). \quad \text{(S40l)}$$

At finite activity, these equations do not have a solution. In the limit $\ell_0 \to 0$, the system is effectively passive and we find $\mathcal{E}_\rho = 1/\rho$ and $\mathcal{E}_\psi = -\psi_N$, recovering the equilibrium Gibbs-Duhem relation and Maxwell construction. In the high-activity limit, $\ell_0 \to \infty$, all $p_{Ci}^{(2,2)}$, $s_{Ci}^{(2,2)}$, $p_{Cij}^{(2,1)}$, and $s_{Cij}^{(2,1)}$ for $\{\rho, \psi\}$ can be ignored as the active contributions at this order scale as ℓ_0^2. In this limit, Eq. (S40) cannot be solved, however Eq. (S40c)-(S40l) can be, where one finds $\mathcal{E}_\rho = p_C^{\text{bulk}}$ and $\mathcal{E}_\psi = 0$. As shown in Ref. [2], when this is the case, the final coexistence criterion is an *approximate* generalized Maxwell construction:

$$\int_{p_C^{\text{bulk}}(\rho^f,\psi_V^f)}^{p_C^{\text{bulk}}(\rho^s,\psi_V^s)} \left(\mathcal{P}^{\text{bulk}}(\rho,\psi_V) - \mathcal{P}^{\text{coexist}}\right) dp_C^{\text{bulk}} \approx 0, \tag{S41}$$

which is path-dependent and generally only vanishes when evaluated along the integration path that corresponds to the spatial coexistence profiles (which is what we would like to circumvent). Evaluating Eq. (S41) along ψ_V^* thus represents and approximate form of the final criterion – combining this with Eq. (S31) is what we refer to as the "active" criteria in the main text, noting that it is only well-defined (albeit still approximate) in the infinite activity limit.

EQUATIONS OF STATE OF ACTIVE BROWNIAN SPHERES

Ultimately, the application of the coexistence criteria derived in the main text will require equations of state for p_C^{bulk}, $\overline{U}$, and ψ_V^*. Simulation data can *only* be obtained for systems in which a state of homogeneous ρ is at least locally stable. Consequently, it is not possible to obtain the complete relevant functional dependence of the required state functions directly from simulation. However, application of our coexistence criteria only requires knowledge of the equations of state at $\psi_V = \psi_V^*(\rho)$ for each density ρ. We therefore proceed by devising a simple simulation protocol to obtain as much of this limited data as possible. We then use this data, along with the known physical limits we require our equations of state to capture, in order to develop physical and semi-empirical bulk equations of state.

We now seek the minimal functional forms of these equations of state that capture the physical limits we impose (e.g., established equations of state for the fluid states and passive crystal, diverging active crystal pressure at close-packing, zero crystallinity at zero density and maximal crystallinity at close-packing) while capturing trends in our data. The chosen forms will necessarily not be unique, however we opt for simple forms that minimize the number of tuning parameters. We then determine the values of the parameters in the equations through straightforward regressions in `Python`.

Physical and Semi-Empirical Bulk Equations of State

To construct the ABP solid-fluid phase diagram by applying our derived coexistence criteria, we need equations of state for the preferred crystallinity, $\psi_N^*\left(\phi;\ell_0/D\right)$, and pressures,

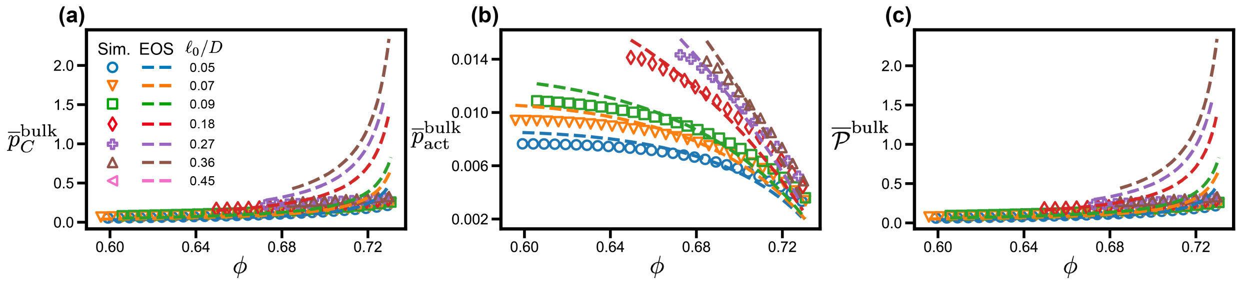

FIG. S1. Conservative interaction (p_C^{bulk}), active ($p_{\text{act}}^{\text{bulk}}$), and total ($\mathcal{P}^{\text{bulk}}$) pressure equations of state at low activity. Pressures are nondimensionalized by the scale $6\zeta U_0/\pi D^2$.

$p_C^{\text{bulk}}(\phi, \psi_N; \ell_0/D)$ and $p_{\text{act}}^{\text{bulk}}(\phi, \psi_N; \ell_0/D)$, that accurately describe both fluid ($\psi_N \approx 0$) and solid ($\psi_N > 0$) phases at all activities. We combine existing equations of state for an ABP fluid [1] (developed for moderate activities $\ell_0/D > 1$) and an equilibrium hard sphere fluid [4] to develop accurate equations of state for ABP fluids at all activities. To extend these equations of state to describe crystalline systems, we develop auxiliary equations of state [e.g., an equation of state for the maximum possible packing fraction, $\phi^{\max}(\psi_N; \ell_0/D)$] to capture the effects of nonzero ψ_N.

The active pressure of ABP fluids developed in Ref. [1] ($p_{\text{act}}^{\text{bulk}}$) correctly recovers the ideal gas pressure in the reversible limit ($\ell_0/D \to 0$), i.e., $p_{\text{act}}^{\text{bulk}} = \rho k_B T_{\text{act}}$ where the active energy scale is $k_B T_{\text{act}} \equiv \zeta U_0 \ell_0/6$. We extend $p_{\text{act}}^{\text{bulk}}$ to nonzero ψ_N by introducing an equation of state $\phi^{\max}(\psi_N; \ell_0/D)$ capturing the crystallinity-dependent maximum volume fraction:

$$p_{\text{act}}^{\text{bulk}}(\phi, \psi_N; \ell_0/D) = \frac{\zeta U_0}{\pi D^2}\left(\frac{\ell_0}{D}\right)\phi \frac{1 + \tanh(A_{\text{act}}\psi_N)\left[1 - c_{\text{act}}^{(1)} t_{100}\left(\frac{\ell_0}{D}, \frac{\ell_0^c}{D}\right) + c_{\text{act}}^{(2)} t_1\left(\frac{\ell_0}{D}, \frac{\ell_0^{\text{act}}}{D}\right)\right]}{1 + \left(1 - \exp\left[-2^{7/6}\left(\frac{\ell_0}{D}\right)\right]\right)\frac{\phi}{1 - \phi/\phi^{\max}(\psi_N; \ell_0/D)}}, \tag{S42a}$$

$$t_B(\ell_0/D, \ell_0^*/D) = \frac{1 + \tanh\left(B\frac{\ell_0 - \ell_0^*}{D}\right)}{2} \tag{S42b}$$

where $\ell_0^c = 18.8 \times 2^{-1/6} D$ is the MIPS critical point [5], $\phi^{\max}(\psi_N = 0; \ell_0/D) = \phi^{\text{RCP}} = 0.645$ to recover the fluid pressure in Ref. [1], and $\phi^{\max}(\psi_N = 1; \ell_0/D) = \phi^{\text{CP}} = 0.74$ when the system has perfect crystalline order. We emphasize that Eq. (S42a) recovers the active pressure in Ref. [1] when $\psi_N = 0$. We fit the coefficients $A_{\text{act}} = 10$, $c_{\text{act}}^{(1)} = 29$, $c_{\text{act}}^{(2)} = 30$, and $\ell_0^{\text{act}} = 3 \times 2^{-1/6} D$. The conservative interaction pressure in Ref. [1] ($p_C^{\text{bulk,ABP}}$) *does not* recover the equilibrium hard sphere pressure ($p_C^{\text{bulk,HS}}$) [4] in the low activity limit. We remedy this by including an interpolation [through an equation of state $x(\ell_0/D)$] between the conservative interaction pressures of an ABP fluid and an equilibrium hard sphere fluid (for which we use a simplified version of that in Ref. [4] that we verify on Brownian dynamics simulations of passive hard spheres). Extending $p_C^{\text{bulk,ABP}}$ to nonzero ψ_N requires an equation of state capturing an empirical crystallinity-induced slowing of its divergence $[\beta(\psi;\ell_0/D)]$ in addition to using $\phi^{\max}(\psi_N;\ell_0/D)$ as the maximum volume fraction:

$$p_C^{\text{bulk}} = x\big(\ell_0/D\big)p_C^{\text{bulk,ABP}} + [1 - x(\ell_0/D)]\, p_C^{\text{bulk,HS}}, \tag{S43a}$$

$$p_C^{\text{bulk,ABP}}(\phi,\psi_N;\ell_0/D) = 2^{-7/6}\frac{\phi^2}{(1-\phi/\phi^{\max})^{\beta}}\left(1 - c_C^{(1)}\tanh(A_C\psi_N)\left[1 - c_C^{(2)} t_{100}\left(\frac{\ell_0}{D},\frac{\ell_0^{\mathrm{c}}}{D}\right)\right]\right) \tag{S43b}$$

$$p_C^{\text{bulk,HS}}(\phi,\psi_N;k_BT) = \frac{6k_BT}{\pi D^3}\frac{\phi}{1-\phi/\phi^{\max}}, \tag{S43c}$$

where $\beta(\psi_N = 0;\ell_0/D) = 1/2$ to recover the pressure in Ref. [1] and $A_C = 10$, $c_C^{(1)} = 5/6$, and $c_C^{(2)} = 0.9$ are coefficients we have fit. We again emphasize that Eq. (S43a) recovers the conservative pressure in Ref. [1] when $\psi_N = 0$ and $\ell_0/D \gg 1$ and recovers the pressure of a hard sphere fluid (see Ref. [4]) when $\psi_N = 0$ and $\ell_0/D = 0$. We have introduced the thermal energy k_BT, which, in systems of active hard spheres, is generally density (and crystallinity) dependent and can be defined as $k_BT \equiv p_{\text{act}}^{\text{bulk}}/\rho$. We find no appreciable differences in the resulting phase diagram when approximating this active temperature with that of ideal ABPs in 3D, $k_BT = k_BT_{\text{act}} = \zeta U_0\ell_0/6$ [6], however. We then use the simpler density-independent effective temperature, k_BT_{act}, when constructing phase diagrams but note that the density dependence of the effective temperature may be more important for other systems.

The equations of state $x(\ell_0/D)$, $\phi^{\max}(\psi_N;\ell_0/D)$, and $\beta(\psi_N;\ell_0/D)$ were empirically fit:

$$\begin{aligned} x\big(\ell_0/D\big) = {} & (1-\tanh(A_x\psi_N))\left[\tanh\left(\log\left(\frac{\ell_0}{D}+1\right) + t_{10}\left(\frac{\ell_0}{D},\frac{\ell_0^{\mathrm{c}}}{D}\right)\left[e^{\ell_0/D}-1\right]\right)\right] \\ & + \tanh(10\psi_N)\left[\tanh\left(\log\left(\frac{\ell_0}{D}+1\right) + t_{100}\left(\frac{\ell_0}{D},\frac{\ell_0^{\mathrm{c}}}{D}\right)^{r_x^{(1)}}\left[e^{(\ell_0/\ell_0^{\mathrm{c}})^{r_x^{(2)}}}-1\right]\right)\right], \end{aligned} \tag{S44a}$$

$$\phi^{\max}(\psi_N;\ell_0/D) = \phi^{\text{RCP}} + \big(\phi^{\text{CP}} - \phi^{\text{RCP}}\big)\tanh(A_{\max}\psi_N), \tag{S44b}$$

$$\beta(\psi_N;\ell_0/D) = \frac{1}{2} - \tanh(A_\beta\psi_N)\left[c_\beta^{(1)} t_5(\ell_0/D,\ell_0^{\mathrm{c}}/D) + c_\beta^{(2)} t_{0.01}\left(\ell_0/D,\ell_0^{\beta}/D\right)\right], \tag{S44c}$$

where $A_x = 10$, $r_x^{(1)} = r_x^{(2)} = 10$, $A_{\max} = 15.848$, $A_\beta = 10$, $c_\beta^{(1)} = 0.4$, $c_\beta^{(2)} = 0.175$, and $\ell_0^{\beta} = 50\times 2^{-1/6}D$ are coefficients we have fit. The forms of these fits were motivated by the previously discussed physical limits that we require to be met (e.g., $\phi^{\max}$ must be 0.74 when $\psi_N = 1$ and ≈ 0.645 when $\psi_N = 0$).

In order to use the equations of state in Eqs. (S42a) and (S43) we require an equation of state for the preferred per-particle crystallinity, ψ_N^*, which we fit an expression for $\psi_N^*(\phi;\ell_0/D)$ (see

Fig. 1 in the main text):

$$\psi_N^*\left(\phi;\ell_0/D\right) = \Theta\left(\phi-\phi^{\mathrm{ODT}}\right)\tanh\left[\exp\left(m_\psi\phi + c_\psi + A_\psi\frac{\phi}{\sqrt{1-\phi/\phi^{\mathrm{CP}}}}\right)\right.$$
$$\left.\times\left(\Delta_\psi^{(1)} + \frac{\left(\ell_0/D\right)^{r_\psi^{(1)}}}{\Delta_\psi^{(2)} + \ln\left[\Delta_\psi^{(3)} + \left(\ell_0/D\right)^{r_\psi^{(2)}}\right]}\right)^{-r_\psi^{(3)}\left(1-\phi/\phi^{\mathrm{CP}}\right)}\right], \quad \text{(S45)}$$

where $m_\psi = 18.8$, $c_\psi = -13.1$, $A_\psi = 0.05$, $\Delta_\psi^{(1)} = 0.01$, $\Delta_\psi^{(2)} = \Delta_\psi^{(3)} = 1$, $r_\psi^{(1)} = 1.16$ and $r_\psi^{(2)} = r_\psi^{(3)} = 2$, are again constants that have been fit. The equation of state for the order-disorder volume fraction, $\phi^{\mathrm{ODT}}\left(\ell_0/D\right)$, [see the inset of Fig. 1 in the main text] was determined to be:

$$\phi^{\mathrm{ODT}}\left(\ell_0/D\right) = \phi_{\mathrm{eqm}}^{\mathrm{ODT}} + \left(\phi^{\mathrm{RCP}} - \phi_{\mathrm{eqm}}^{\mathrm{ODT}}\right)\tanh\left(A_{\mathrm{ODT}}\log\left[c_{\mathrm{ODT}}\frac{\ell_0}{D} + 1\right]\right), \quad \text{(S46)}$$

where $\phi_{\mathrm{eqm}}^{\mathrm{ODT}} = 0.515$ is the equilibrium hard sphere ϕ^{ODT} (determined by imposing that the equilibrium Maxwell construction on $p_C^{\mathrm{bulk,HS}}$ returns the hard sphere crystal-fluid binodals measured in simulations [7]) and $A_{\mathrm{ODT}} = 1.381$ and $c_{\mathrm{ODT}} = 0.909$ are fitted constants.

We see that since our equation for ψ_N^* in Eq. (S45) experiences a discontinuity at ϕ^{ODT}, our equation for p_C^{bulk} in Eq. (S43) does as well. This discontinuity is necessary for passive solid-fluid coexistence, as the pressure (evaluated at ψ_N^*) must be non-monotonic with increasing ρ in order to find binodals.

Figure S1 shows the fits for p_C^{bulk} and $p_{\mathrm{act}}^{\mathrm{bulk}}$ at low activities after inserting the expressions for x, ϕ^{max}, β, ϕ^{ODT}, and ψ^* into Eqs. (S42a) and (S43). While the fit for p_C^{bulk} is an overestimate, the qualitative ℓ_0/D and ϕ dependent trends are captured, whereas the fit for $p_{\mathrm{act}}^{\mathrm{bulk}}$ is more quantitatively accurate.

Characterization of the "Pseudo"-spinodal

There are two spinodals, or regions of instability, in our dynamic pressure $\left(\mathcal{P}^{\mathrm{bulk}} = p_C^{\mathrm{bulk}} + p_{\mathrm{act}}^{\mathrm{bulk}}\right)$ of active hard spheres in Eqs. (S43) and (S42a). The first is a true spinodal indicating that the fluid phase ($\psi_N \approx 0$) is unstable at certain densities. The fluid spinodal, which occurs above the critical activity, arises from a non-monotonic active pressure and results in MIPS. The second is a "pseudo"-spinodal which drives crystallization, even in the reversible limit. We distinguish this spinodal as it indicates that states of intermediate density and finite ψ_N (which cannot generally be prepared) are unstable.

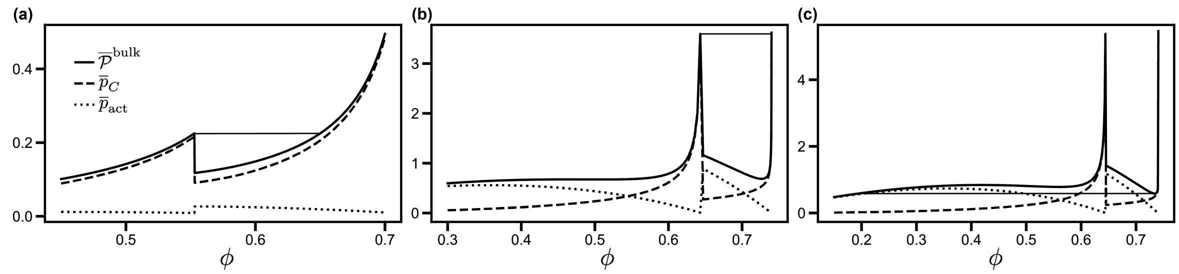

FIG. S2. ABP pressure (nondimensionalized by the scale $6\zeta U_0/\pi D^2$) at (a) low activity $\ell_0/D = 0.9$, (b) intermediate activity $\ell_0/D = 17.4$, and (c) high activity $\ell_0/D = 22.3$. p_C^{bulk} is shown in dashed lines while $p_{\mathrm{act}}^{\mathrm{bulk}}$ is shown in dotted lines.

For a solid-fluid transition to occur for passive hard spheres, p_C^{bulk} must contain a discontinuity at the order-disorder volume fraction, ϕ^{ODT}. This discontinuity represents a region of instability that occurs over an infinitely narrow range of ϕ where ψ_N^* adopts a nonzero value, representing a pseudo-spinodal. The pseudo-spinodal widens at finite activity due to the non-monotonicity of $p_{\mathrm{act}}^{\mathrm{bulk}}$, encompassing a finite range of volume fractions above ϕ^{ODT}. Figure S2 shows the widening of this pseudo-spinodal, showing the active and conservative interaction contributions to $\mathcal{P}^{\mathrm{bulk}}$ at low, intermediate, and high activity (the same activities as Fig. 2 in the main text).

* aomar@berkeley.edu

[S1] A. K. Omar, H. Row, S. A. Mallory, and J. F. Brady, Proc. Natl. Acad. Sci. U.S.A. **120** (2023).
[S2] D. Evans and A. K. Omar, Phys. Rev. Res. **7**, 043229 (2025).
[S3] S. Hermann and M. Schmidt, Phys. Rev. E **109**, L022601 (2024).
[S4] Y. Song, R. M. Stratt, and E. A. Mason, J. Chem. Phys. **88**, 1126 (1988).
[S5] A. K. Omar, K. Klymko, T. GrandPre, and P. L. Geissler, Phys. Rev. Lett. **126**, 188002 (2021).
[S6] S. C. Takatori, W. Yan, and J. F. Brady, Phys. Rev. Lett. **113**, 028103 (2014).
[S7] B. J. Alder and T. E. Wainwright, J. Chem. Phys. **27**, 1208 (1957).