

Capillary Interfacial Tension in Active Phase Separation

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In passive fluid-fluid phase separation, a single interfacial tension sets both the capillary fluctuations of the interface and the rate of Ostwald ripening. We show that these phenomena are governed by two different tensions in active systems, and compute the capillary tension σ_{cw} which sets the relaxation rate of interfacial fluctuations in accordance with capillary wave theory. We discover that strong enough activity can cause negative σ_{cw} . In this regime, depending on the global composition, the system self-organizes, either into a microphase-separated state in which coalescence is highly inhibited, or into an “active foam” state. Our results are obtained for Active Model B+, a minimal continuum model which, although generic, admits significant analytical progress.

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Active particles extract energy from the environment and dissipate it to self-propel [1,2]. Among their notable self-organizing features is phase separation into dense (liquid) and dilute (vapor) regions, even for purely repulsive particles [3–5]. Although generically a far-from-equilibrium effect, active phase separation was first described via an approximate mapping onto equilibrium liquid-vapor phase separation [3,4], leading to early speculation that time reversal symmetry might be restored macroscopically in steady state [3,6–11]. Indeed, activity is an irrelevant perturbation near the liquid-vapor critical point, albeit without causing emergent reversibility [12].

Recently it has become clear, however, that bulk phase separation in active systems displays strongly nonequilibrium features. Bubbly phase separation [13] was evidenced in simulations of repulsive self-propelled particles [14,15]: here large liquid droplets contain a population of mesoscopic vapor bubbles that are continuously created in the bulk, coarsen, and are ejected into the exterior vapor, creating a circulating phase-space current in the steady state. Microphase separation of vapor bubbles [15,16] has been further observed numerically, alongside a similar phase of finite dense clusters, often found in experiments with self-propelled colloids [17,18] and bacteria [19]. Recently, even more intriguing forms of phase separation have been reported in an active system of nematodes, comprising a phase where dense filaments continuously break up and reconnect [20].

Much understanding of active phase separation has been gained from continuum field theories. In the simplest setting [13,21,22], these only retain the evolution of the density field ϕ , while hydrodynamic [23,24] or polar [25]

fields can be added if the phenomenology requires. Their construction, via conservation laws and symmetry arguments, follows a path first introduced with Model B for passive phase separation [26–28]. Yet, these field theories differ from Model B because locally broken time-reversal symmetry implies that new nonlinear terms are allowed. The ensuing minimal theory, Active Model B+ (AMB+) [11,13], including all terms that break detailed balance up to order $\mathcal{O}(\nabla^4\phi^2)$ in a gradient expansion [11,13], is defined by

$$\partial_t\phi = -\nabla \cdot (\mathbf{J} + \sqrt{2DMA}), \quad (1)$$

$$\mathbf{J}/M = -\nabla\mu_\lambda + \zeta(\nabla^2\phi)\nabla\phi, \quad (2)$$

$$\mu_\lambda[\phi] = \frac{\delta\mathcal{F}}{\delta\phi} + \lambda|\nabla\phi|^2, \quad (3)$$

where $\mathcal{F} = \int d\mathbf{r}[f(\phi) + (K(\phi)/2)|\nabla\phi|^2]$, $f(\phi)$ is a double-well local free energy, and $\mathbf{\Lambda}$ is a vector of zero-mean, unit-variance, Gaussian white noises. Model B is recovered at vanishing activity ($\lambda = \zeta = 0$), unit mobility ($M = 1$) and constant noise level D [26].

It is known that at low activity (small λ , ζ), AMB+ undergoes conventional bulk phase separation. At higher activity, Ostwald ripening [29], the classical diffusive pathway to macroscopic phase separation, can go into reverse [13]. This explains the emergence of bubbly phase separation and microphase-separated vapor bubbles. (These phases arise when $\zeta, \lambda > 0$; for $\zeta, \lambda < 0$ the identities of liquid and vapor phases are interchanged.) More specific

mechanisms, due to hydrodynamics [24,30] or chemotaxis [31,32], can also piecewise explain some of these phases. AMB+ does not refute such specific explanations, but offers a minimal framework to address generic features of active phase equilibria. Its simplicity admits both significant analytical progress, and efficient numerics.

For active systems showing bulk liquid-vapor phase separation it has been debated, on the basis of numerical and analytical studies, how to define the liquid-vapor interfacial tension [33–39]. One key result of this Letter is to confirm that no unique definition is possible. Inspired by work on equilibrium interfaces [28], we derive an effective equation for the interface height, and calculate the capillary tension σ_{cw} which sets the spectrum of capillary waves and the relaxation times of height fluctuations. We find σ_{cw} differs from σ , the tension introduced in Ref. [13] to describe the Ostwald process. Whereas $\sigma < 0$ in the reverse Ostwald regime, this does not ensure capillary instability, which instead requires $\sigma_{\text{cw}} < 0$. When the latter holds, depending on the global density, we find two new types of active phase separation (Fig. 4), driven by an interfacial instability of Mullins-Sekerka type [40]: a microphase-separated droplet state, where coalescence among droplets is highly inhibited, and an “active foam” state.

As is standard [13,26], we now set $M = 1$, assume constant D , K , and select $f(\phi) = a(-\phi^2/2 + \phi^4/4)$ with $a > 0$. [Our results can be extended to any double-well f and any $K(\phi) > 0$.] We set $\zeta > 0$, meaning that reversed Ostwald ripening happens only for vapor bubbles. The corresponding results for $\zeta < 0$ follow from the invariance of our model under $(\phi, \lambda, \zeta) \rightarrow -(\phi, \lambda, \zeta)$. We denote by ϕ_1 and ϕ_2 the coexisting vapor and liquid densities in the mean-field limit, $D = 0$; note that $\phi_{1,2} = \pm 1$ in the passive case only. More generally they are found by changing variables from ϕ and f to ψ and g : these “pseudovariables,” introduced in Ref. [41] for $\zeta = 0$ and then generalized to AMB+ [13], solve $K\partial^2\psi/\partial\phi^2 = (\zeta - 2\lambda)\partial\psi/\partial\phi$ and $\partial g/\partial\psi = \partial f/\partial\phi \equiv \mu$, whence $\psi = K(\exp[(\zeta - 2\lambda)\phi/K] - 1)/(\zeta - 2\lambda)$. In terms of them, the equilibrium conditions $\mu_1 = \mu_2$ and $(\mu\psi - g)_1 = (\mu\psi - g)_2$ which select the binodals $\phi_{1,2}$ still hold [13,41]. (This change of variables is primarily a mathematical device for constructing the phase equilibria; ψ and g have no direct physical significance beyond this.) All our analytic results are valid in dimensions $d \geq 2$, while our numerics were done in $d = 2$ with periodic boundary conditions and system size $L_x \times L_y$, using a pseudospectral algorithm with Euler updating [42].

We start, following Ref. [29], by deriving the effective dynamics for small fluctuations of the interface height $\hat{h}(\mathbf{x}, t)$ above a $(d - 1)$ plane, with in-plane and vertical coordinates $(\mathbf{x}, y) = \mathbf{r}$. We assume the absence of overhangs. On a rapid timescale, we expect diffusion to quasistatically relax $\phi(\mathbf{r}, t)$ to a value that depends only

on the distance to the interface. For small amplitude, long-wavelength perturbations, the vertical direction and the one normal to the interface are equivalent and we thus can assume that

$$\phi(\mathbf{r}, t) = \varphi[y - \hat{h}(\mathbf{x}, t)], \quad (4)$$

where φ is the interfacial profile. By mass conservation, the spatial average of \hat{h} is constant; we set $\hat{h} = 0$. It will turn out that \hat{h} solves a nonlocal equation in space, so we work in terms of its Fourier transform $h(\mathbf{q}_x, t)$. We proceed by plugging Eq. (4) into Eq. (1) and inverting the Laplace operator. We multiply $\nabla^{-2}\partial_t\varphi$ by $\partial_y\psi$, integrate across the interface, Fourier transform along the \mathbf{x} direction, and expand in powers of h . Denoting $q = |\mathbf{q}_x|$, we obtain [42]

$$\partial_t h = -\frac{1}{\tau(q)}h + \chi + \mathcal{O}(q^2 h^2), \quad (5)$$

$$\frac{1}{\tau(q)} = \frac{2\sigma_{\text{cw}}(q)q^3}{A(q)}, \quad (6)$$

where

$$\sigma_{\text{cw}}(q) = \sigma_\lambda + \frac{3\zeta}{4} \int dy_1 dy_2 \frac{(y_1 - y_2) \psi'(y_1) \varphi'^2(y_2)}{|y_1 - y_2| e^{q|y_1 - y_2|}}, \quad (7)$$

$$\sigma_\lambda = K \int dy \varphi'(y) \psi'(y), \quad (8)$$

and χ is a zero-mean Gaussian noise with correlations $\langle \chi(\mathbf{q}_1, t_1) \chi(\mathbf{q}_2, t_2) \rangle = C_\chi(q_1) \delta(\mathbf{q}_1 + \mathbf{q}_2) \delta(t_1 - t_2)$, with

$$C_\chi(q) = 4(2\pi)^{d-1} \frac{DB(q)}{A^2(q)} q. \quad (9)$$

In Eqs. (6), (9), $A(q) \equiv \int dy_1 dy_2 \psi'(y_1) \varphi'(y_2) \exp(-q|y_1 - y_2|)$ and $B(q) \equiv \int dy_1 dy_2 \psi'(y_1) \psi'(y_2) \exp(-q|y_1 - y_2|)$. Note that (5) omits nonlinear terms, derived in the Supplemental Material [42], that previously arose in models of conserved surface roughening [43,44].

The effective height equations (5)–(9) are the fundamental analytic results of this Letter. For wavelengths much larger than an interfacial width $\xi \sim \xi_{eq} = (2K/a)^{1/2}$, we can replace $\sigma_{\text{cw}}(q)$, $A(q)$, and $B(q)$ with their limiting values as $q \rightarrow 0$. These, with a slight abuse of notation, are denoted as σ_{cw} , A , and B . Explicitly, the resulting capillary-wave tension σ_{cw} obeys

$$\sigma_{\text{cw}} = \sigma_\lambda - \frac{3\zeta}{2} \int dy \left[\psi(y) - \frac{\psi_1 + \psi_2}{2} \right] \varphi'^2(y), \quad (10)$$

where $\psi_{1,2} = \psi(\phi_{1,2})$ are the pseudodensities at the binodals. As expected, in the equilibrium limit $\lambda, \zeta \rightarrow 0$, σ_{cw}

reduces to the standard interfacial tension $\sigma_{\text{eq}} = K \int dy \varphi'^2(y)$. [45] which governs not only the capillary fluctuation spectrum, but the Laplace pressure and the rate of Ostwald ripening [28,29]. Switching on activity breaks this degeneracy. Indeed the tension determining the rate of Ostwald ripening of a bubble was given in Ref. [13] as $\sigma = \sigma_\lambda - \zeta \int dy [\psi - \psi(0)] \varphi'^2(y)$, where $\psi(0)$ is the value of the pseudodensity at the droplet center. Therefore σ is in general not equal to σ_{cw} .

To gain explicit predictions from Eqs. (5)–(9), we must evaluate σ_{cw} , A , and B . This requires knowledge of the interfacial shape $\varphi(y)$. At equilibrium, this is well known [45]: $\varphi_{\text{eq}}(y) = \pm \tanh(y/\xi_{\text{eq}})$, with $\xi_{\text{eq}} = \sqrt{2K/a}$ and $\sigma_{\text{eq}} = \sqrt{8Ka}/9$. (Note that $A = B = 4$ in this case.) Also, whenever $2\lambda = \zeta$ it is readily shown that $\varphi = \varphi_{\text{eq}}$ so that $\sigma_{\text{cw}} = \sigma_{\text{eq}}$, although the Ostwald tensions are $\sigma = \sigma_{\text{eq}}(1 \mp \zeta/K)$ for bubble growth (–) and liquid droplet growth (+), respectively [13]. We do not have closed-form results for σ_{cw} at general λ , ζ ; however, a change of variable to $w(\varphi) = \varphi'^2$ in the integrals defining σ_{cw} , A , B allows use of a simple numerical procedure introduced in Ref. [21] and detailed in Ref. [42] to find the low q behavior. To examine $q \neq 0$ below we instead extract the interface profile from simulations at $D = 0$.

Figure 1 shows a phase diagram of AMB+ for $\zeta > 0$ at mean-field level, delineating zones of negative σ and σ_{cw} . (There are none at $\zeta > 0$ and $\lambda < 0$). This provides the full phase diagram of AMB+: the case of $\zeta < 0$ follows from Fig. 1 using the symmetry $(\lambda, \zeta, \phi) \rightarrow -(\lambda, \zeta, \phi)$ of AMB+, which interchanges the liquid and vapor identities. For small activity, or for $\lambda\zeta < 0$, $\sigma_{\text{cw}} > 0$, even where $\sigma < 0$; here vapor bubbles undergoing reversed Ostwald ripening have stable interfaces and, depending on the global density, the system is either microphase separated or in bubbly phase separation [13]. At high activity a new

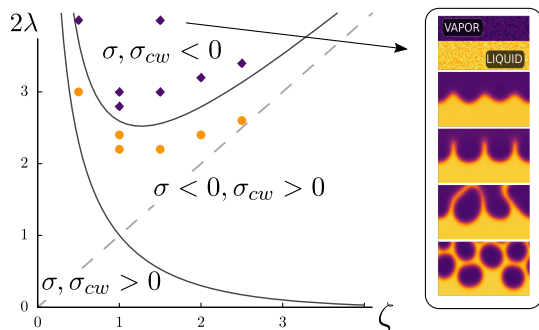


FIG. 1. Mean-field phase diagram for $\zeta > 0$, showing sign regimes of interfacial tensions σ and σ_{cw} . When $\sigma_{\text{cw}} > 0$, the interface is stable and unstable otherwise. Orange circles and blue squares respectively denote the results of direct simulations of AMB+ where the instability of the interface is or is not observed. Right: interfacial instability ($\zeta = 1.5$, $\lambda = 2$).

regime emerges where $\sigma_{\text{cw}} < 0$ implying that such interfaces (and also flat ones) become locally unstable.

We first consider the regime $\sigma_{\text{cw}} > 0$, where our theory predicts this capillary tension to govern, via Eq. (6), the relaxation times of the interface $\tau(q)$. To check this, we performed simulations of AMB+ for $D = 0$ starting from a phase separated state with the interface perturbed via a single mode; Fig. 2 confirms that $h(\mathbf{q}, t) = h(\mathbf{q}, 0) \exp[-t/\tau(q)]$ as predicted by (5)–(9), for either sign of the Ostwald tension σ . Our theory also predicts the stationary structure factor of the interface $S(q) = \lim_{t \rightarrow \infty} \langle |h(\mathbf{q}, t)|^2 \rangle$:

$$S(q) = \frac{(2\pi)^{d-1} D B(q)}{\sigma_{\text{cw}}(q) q^2 A(q)} \xrightarrow{q\xi^{-1} \ll 1} \frac{(2\pi)^{d-1} D_{\text{eff}}}{\sigma_{\text{cw}} q^2}, \quad (11)$$

where $D_{\text{eff}} = D(\psi_2 - \psi_1)/(\phi_2 - \phi_1)$ is an effective capillary temperature. Equation (11) generalizes capillary wave theory. Its equilibrium analog, $S(q) \propto D/\sigma_{\text{eq}} q^2$ [46], is often justified using equipartition arguments but, even in equilibrium, higher-order gradient terms give subleading corrections at finite q [47,48]. Activity impacts the interfacial fluctuations by renormalizing the temperature $D \rightarrow D_{\text{eff}}$ and, separately, replacing σ_{eq} with σ_{cw} . Even though Eq. (11) also neglects the additional nonlinearities omitted from Eq. (5), it is quite accurate at small D (Fig. 2). The use of capillary wave theory in phase-separated active systems was previously advocated heuristically [34,36,37], but until now, only qualitative estimates were provided for the coefficient $D_{\text{eff}}/\sigma_{\text{cw}}$ in Eq. (11).

When $\sigma_{\text{cw}} < 0$, a drastically new nonequilibrium phenomenology arises. Although the vapor-liquid interface is unstable to height fluctuations, the system remains phase separated. For, unlike in equilibrium where demixing itself cannot be sustained at negative tension, the active

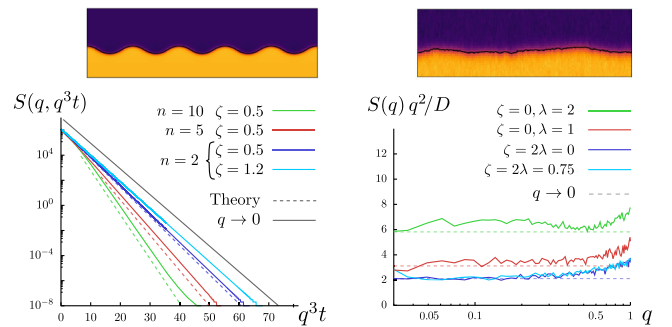


FIG. 2. (Left) liquid-vapor interface for $\sigma_{\text{cw}} > 0$ ($L_x = L_y = 256$), $D = 0$, $\zeta = 2\lambda$ and its relaxation compared to the theoretical predictions for initial perturbations at wave number $2\pi n/L_x$. Dashed lines are predictions obtained using $\tau(q)$, converging to the $q \rightarrow 0$ prediction (continuous line). (Right) snapshot in steady state for $D = 5 \times 10^{-3}$ and scaled structure factor $q^2 S(q)/D$ vs q compared to the $q \rightarrow 0$ analytical prediction; results are averaged over 30 realisations of duration 10^6 after equilibration.

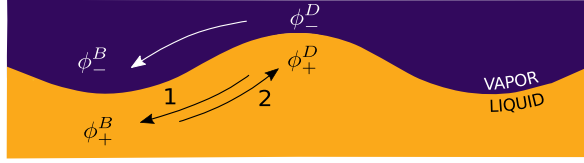


FIG. 3. Instability at $\sigma_{cw} < 0$. The densities on the two sides of the interface adjust quasistatically at values that depend on its local curvature. The ensuing diffusive density fluxes on the vapor side is always stabilising (white arrow); that in the liquid is stabilising when $\sigma > 0$ (arrow 1) and becomes destabilizing when $\sigma < 0$ (arrow 2). This (one-sided) reverse-Ostwald current does not trigger an instability unless the current in the liquid outweighs that in the vapor, which requires $\sigma_{cw} < 0$.

interface does not undergo diffusive collapse but remains linearly stable against normal perturbations $\phi(\mathbf{x}, y) = \phi(y) + \partial_y \epsilon(y)$ [42,49,50].

Next, we numerically simulated AMB+ at $D = 0$, with a noisy initial condition. Orange and blue dots in Fig. 1, respectively, represent cases where the interfacial fluctuation is damped or amplified (movie 1 in the Supplemental Material [42]), showing the accuracy of our analytical predictions. Computing $\tau(q)$ shows that the first unstable mode is at the lowest q available; thus the transition line $\sigma_{cw} = 0$ is critical.

The interfacial instability mechanism (Fig. 3) is reminiscent of the Mullins-Sekerka instability in solidification [40]. In both cases the instability is driven by a single diffusing field: latent heat in crystal growth, and density here. Such a diffusing field settles to quasistationary values $\phi_{\pm}^{B,D}$ on the two sides of the interface which depend on the local curvature. By approximating $\phi_{\pm}^{B,D}$ as the densities near the interface of a vapor bubble (B) or liquid droplet (D), we find that the diffusive current on the vapor side is always stabilizing. In contrast, depending on whether Ostwald ripening is normal or reversed, the current on the liquid side is stabilizing or destabilizing. Reversed Ostwald ripening is, however, not sufficient to drive overall

instability of the interface; this arises only if the current on the liquid side is stronger than the one on the vapor. This condition sets the threshold beyond which $\sigma_{cw} < 0$. Measuring the steady state currents confirms this mechanistic picture [42].

We now report simulations with a small but finite noise level to ensure reproducible steady states. Starting from a near-uniform initial state, we find that the final phase separation is strongly affected by interfacial instability. The stable case, $\sigma_{cw} > 0$, was explored in Ref. [13]. For the unstable case, $\sigma_{cw} < 0$, the stationary states seen by varying the global density $\phi_0 = \int \phi d\mathbf{r}/V$ are reported in Fig. 4 and movie 2 in the Supplemental Material [42]. When ϕ_0 lies outside the mean-field binodals $\phi_{1,2}$, the system remains homogeneous. Within them, at large ϕ_0 where the liquid is the majority phase, we find a microphase-separated state where coalescence of crowded bubbles is highly inhibited. The bubble size distribution $P(A)$ is strongly peaked, increasingly so as noise decreases, suggesting that the average bubble size $\langle A \rangle$ is finite when $D \rightarrow 0$. Our results are converged in time for $D > 0.1$; at lower noise the system gets trapped into metastable states, evolving only because of rare fluctuations of the bubbles interface [42]. Clearly, the average size is not set by the most unstable mode of the flat interface, as the steady state is attained through secondary instabilities (movies 1 and 3 in the Supplemental Material [42]). This phenomenology is at odds with the bubble phase at $\sigma_{cw} > 0$ [13], where a dynamical balance between nucleation, coalescence, and reversed Ostwald causes $\langle A \rangle \rightarrow \infty$ when $D \rightarrow 0$. The difference between these two microphase separated states is also apparent dynamically when starting from bulk phase separation (movie 3 in the Supplemental Material [42]).

When the liquid is the minority phase, bubbles cannot avoid touching and coalescing. One might expect that the system attains a microphase separated state of liquid droplets (for $\zeta > 0$); this is not the case because, as is clear from our mechanistic argument above, the interfaces bends toward the vapor side. Instead, we find a distinctive

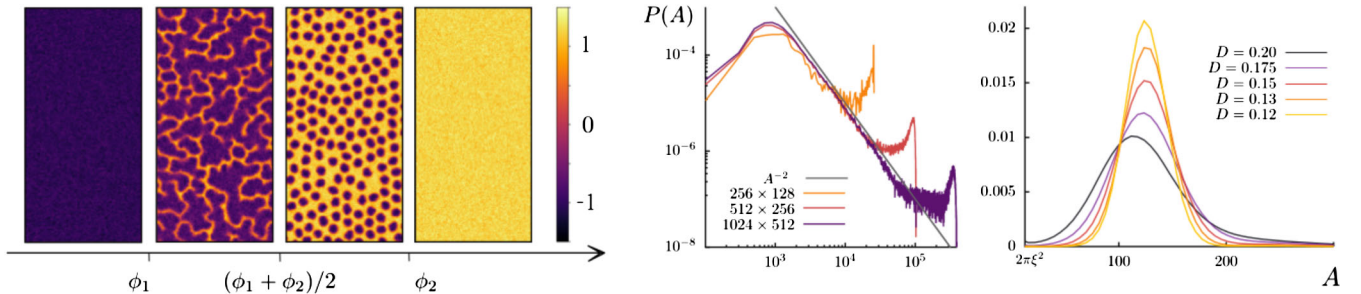


FIG. 4. (Left) phase diagram when $\sigma_{cw} < 0$ as a function of the global density $\phi_0 = -1, -0.4, 0.4, 1.2$ at $D = 0.05$, $L_x = 256$, $L_y = 512$, and $\lambda = 1.75$, $\zeta = 2$, for which $\phi_1 = -0.9$, $\phi_2 = 1.08$. At high and low ϕ_0 , the system is homogeneous (liquid or vapor states). Within the binodals, when $\phi_0 > (\phi_1 + \phi_2)/2$, the system shows microphase-separated vapor bubbles whose coalescence is highly inhibited. At lower ϕ_0 , the system forms a continuously evolving active foam state. (Middle and right): area distribution of vapor regions for the active foam state ($\phi_0 = -0.4$) and in the microphase-separated state ($\phi_0 = 0.2$, noise values in the legend).

form of phase separation, which we call the “active foam” state. Thin filaments of liquid are dispersed in the vapor phase, which continuously break up and reconnect. This state is previously unknown in active scalar models but resembles patterns that can arise, by a different mechanism, in active liquid crystals [51]. The filaments are bent on the most unstable lengthscale of the flat interface. The area distribution of vapor regions [Fig. 4(b)] is now peaked at size that corresponds to the merging of two bubbles, but a power-law tail A^{-2} emerges, only cutoff by the system size. The boundaries in ϕ_0 between the different phases of Fig. 4 are qualitative: while the vapor density is almost independent of ϕ_0 , the liquid density varies [42].

The techniques introduced here could help elucidate σ_{cw} in particle-based active models, by applying them to various field-theoretical descriptions obtained by explicit coarse-graining [13,41,52], or to describe confluent biological tissues, where the measured interfacial tension was shown to be dependent on the protocol [53]. The roughening properties of the interface also merit further study: the anomalous scaling found in particle-based simulations was interpreted to be in the Edwards-Wilkinson universality class [36,37]. Dimensional analysis [54] of our linear theory instead gives the critical exponents $z = 3$ and $\chi = (z - d)/2$, where $\langle \hat{h}(\mathbf{x}, t) \hat{h}(\mathbf{x}', t) \rangle \sim |\mathbf{x} - \mathbf{x}'|^{2\chi}$ and $\langle \hat{h}(\mathbf{x}, t) \hat{h}(\mathbf{x}, t') \rangle \sim |t - t'|^{2\chi/z}$. The impact of nonlinearities should be studied by renormalization methods.

Finally, it is remarkable that (a) the capillary tension can likewise become negative, and that (b) this leads to new types of phase separation including active foam states. Our generic field-theoretical approach is agnostic as to the microscopic mechanisms underlying activity (and even phase separation). Therefore the microscopic ingredients needed for our new phases remain to be identified. For the same reason, we expect them to be widely present in phase-separating systems with locally broken detailed balance: besides motility-induced phase separation [4], applications might encompass cell sorting in biological tissues [53], tumor invasion [55], and sociophysics [56].

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