

Phase separation with nonlocal interactions

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Phase separation in complex systems is a ubiquitous phenomenon. While simple theories predict coarsening until only macroscopically large phases remain, concrete models often exhibit patterns with finite length scales. To unify such models, we here propose a general field-theoretic model that combines phase separation with nonlocal interactions. Our analysis reveals that long-range interactions generally suppress coarsening, whereas systems with nonlocal short-range interactions additionally exhibit a continuous phase transition to patterned phases. Only the latter system allows for the coexistence of homogeneous and patterned phases, which we explain by mapping to the conserved Swift-Hohenberg model. Taken together, our generic model reveals an underlying framework that describes similar phenomena observed in many complex phase-separating systems.

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Phase separation, describing the demixing of a homogeneous mixture into dense and dilute regions, is ubiquitous in nature and materials science [1–9]. Standard demixing is qualitatively well described by classical theories, in particular Flory’s regular solution theory [10,11]. One key property of such theories is that they penalize interfaces, thus favoring one large droplet over multiple smaller ones. This leads to a coarsening process known as Ostwald ripening [12], which drives systems toward macroscopically large homogeneous phases. Yet suppressed coarsening, featuring patterns with finite length scales, has been observed for many phase-separating systems with complex interactions, including block copolymers [13–15], nonlocal elasticity [16–21], chemical reactions [22–27], material exchange between compartments [28], electrostatics [29–32], and membrane interactions [33,34]. Numerical analysis of these dynamics are time-consuming due to the complexity of the mechanisms leading to suppression of coarsening, thus limiting insights into the full nonlinear effects of such mechanisms and their interplay with phase separation, which ultimately dominates at long times. While theoretical analyses have yielded valuable insights into the mechanisms underlying suppressed droplet coarsening in these examples, such findings remain largely system dependent.

Suppressed coarsening can be attributed to effective nonlocal interactions in many phase-separating systems. This feature is immediately clear in the case of electrostatic interactions and nonlocal elasticity, but has also been demonstrated for block copolymers [13–15] and membrane interactions [33,34]. Perhaps surprisingly, even some active chemical re-

actions can be mapped onto effective Coulomb interactions that capture the different interactions mediated by reacting and diffusing species [9,22–25,35]. In these examples, the effective nonlocal interactions capture the dominant effects after microscopic degrees of freedom have been coarse grained, similar to a Ginzburg-Landau approach to phase transitions. However, while the suppression of coarsening can be qualitatively explained by such effective nonlocal interactions, the microscopic details differ fundamentally between the examples, and it remains unclear which observations depend on system-specific details and which result from the generic interplay between phase separation and nonlocal interactions.

In this Letter, we propose a generic model of phase separation with nonlocal interactions and show that coarsening is generally suppressed once nonlocal interactions are present. In addition to this common feature, we also reveal fundamental differences between long-range and short-range nonlocal interactions, which allow us to categorize the various complex interactions mentioned above. Finally, mapping complex mechanisms into effective nonlocal interactions greatly reduces the computational burden of numerical simulations, allowing us to explore the full nonlinear behavior of pattern formation and to uncover the most relevant effects of such mechanisms to droplet formation and growth.

To develop our theory, we consider an isothermal system of volume V described by a coarse-grained density field $\phi(\mathbf{x})$ of a single component. To analyze the effects of nonlocal interactions, we study the free energy

$$F[\phi] = \int d\mathbf{x} \left\{ f[\phi(\mathbf{x})] + \frac{\kappa h^2}{2} |\nabla \phi(\mathbf{x})|^2 \right\} + \frac{1}{2} \iint d\mathbf{x} d\mathbf{x}' g[\phi(\mathbf{x})] K(|\mathbf{x} - \mathbf{x}'|) g[\phi(\mathbf{x}')], \quad (1)$$

where f accounts for purely local interactions driving phase separation. The second term on the first line associates an energy penalty κ with compositional variations, resulting in interfaces of typical width h between regions of different density. The second line represents nonlocal interactions arising

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from coarse graining microscopic degrees of freedom not resolved at our field-theoretic level. We characterize these interactions by an isotropic kernel $K(|\mathbf{x} - \mathbf{x}'|)$ that sets the interaction strength between the density field at positions $\mathbf{x}$ and $\mathbf{x}'$ after a general weight function $g[\phi]$ has been applied. As discussed in more detail later, such a weight function is necessary to ensure the model is thermodynamically well defined and the free energy density remains intensive in the case of long-range interactions. For simplicity, we here focus on linear weight functions $g[\phi] = \phi - \bar{\phi}$ with mean density $\bar{\phi} = V^{-1} \int \phi dV$, which represent effective pairwise nonlocal interactions. Since we consider mass-conserving systems, we employ Model B dynamics [36],

$$\partial_t \phi(\mathbf{x}) = \nabla \cdot \left[\Lambda(\phi) \nabla \frac{\delta F}{\delta \phi(\mathbf{x})} \right], \quad (2)$$

where spatial fluxes are driven by gradients in chemical potential $\delta F / \delta \phi(\mathbf{x})$ and are proportional to the density-dependent mobility $\Lambda(\phi)$.

We start by analyzing the stability of homogeneous states for general nonlocal interaction kernels K and mobilities $\Lambda(\phi)$. Inserting Eq. (1) into Eq. (2) and using the perturbative expansion $\phi(\mathbf{x}, t) = \bar{\phi} + \int d\mathbf{q} \hat{\phi}(\mathbf{q}, t) e^{i\mathbf{q}\mathbf{x}}$, we find the dispersion relation

$$\omega(q) = -q^2 \Lambda(\bar{\phi}) \{ f''(\bar{\phi}) + h^2 \kappa q^2 + \hat{K}(q) \}, \quad (3)$$

with wave number $q = |\mathbf{q}|$ (due to isotropy) and Fourier-transformed interaction kernel $\hat{K} = \int e^{-i\mathbf{q}\mathbf{x}} K(\mathbf{x}) d\mathbf{x}$. Note that here, and below, primes denote derivatives with respect to the argument of the function.

To understand the influence of nonlocal interactions, we first ask whether the homogeneous state is stable. This can be determined from the fastest-growing Fourier mode q_* , for which $\omega'(q_*) = 0$. If $\omega(q_*) > 0$, the system is unstable, and thus in the *spinodal region*. Analogous to systems without nonlocal interactions [37], increasing f'' (e.g., by increasing the temperature) weakens the instability until the stability boundary is reached at the critical value f_s'' , i.e., when $\omega(q_*) = 0$. This boundary is thus characterized by the value of f_s'' and the corresponding most unstable mode q_s , which together satisfy $\omega(q_s) = \omega'(q_s) = 0$, such that

$$f_s''(\bar{\phi}) + h^2 \kappa q_s^2 + \hat{K}(q_s) = 0, \quad (4a)$$

$$2h^2 \kappa q_s + \hat{K}'(q_s) = 0 \quad (4b)$$

define the spinodal boundary. Consequently, phase separation will occur via spinodal decomposition whenever $f'' < f_s''$, e.g., for low enough temperatures. The associated spinodal mode q_s is equivalent to the marginal mode of the free energy F (obeying $\partial_q (\delta^2 F / \delta \phi^2)|_{q=q_s} = 0$). Since the dynamics given by Eq. (2) minimize the free energy, this marginal mode is independent of the dynamics and thus reveals fundamental long-term behavior of the system. In contrast, the fastest-growing mode q_* , which we derive in Appendix A, does depend on dynamics and is only relevant for the initial behavior.

Without nonlocal interactions ($\hat{K}(q) = 0$), Eq. (4b) implies a vanishing marginal mode ($q_s = 0$), so modes with arbitrarily large length scales will minimize the free energy. Additionally, Eq. (4a) implies that marginal stability is achieved when

$f''(\bar{\phi}) = 0$, consistent with the classical spinodal analysis of systems without nonlocal interactions [37]. Within the spinodal region ($f''(\bar{\phi}) < 0$), we have $\omega'(q) > 0$, so the system can always be made more stable by increasing length scales (decreasing q), leading to coarsening. Since we focus on long length scales, where only properties of the bulk interactions matter, we expect this conclusion to hold beyond the linear regime.

Nonlocal interactions affect the spinodal conditions fundamentally. First, nonlocal interactions modify the stability condition (4b), so the marginally stable mode q_s is typically nonzero, suggesting modes with finite length scale will prevail. Second, Eq. (4a) shows that nonlocal interactions shift f_s'' with respect to the case without nonlocal interactions. In particular, nonlocal interactions destabilize the system and enlarge the spinodal region for $\hat{K}(q_s) < 0$, so that weaker attractive forces are now sufficient to drive phase separation. In contrast, $\hat{K}(q_s) > 0$ stabilizes the system, so stronger attractive interactions are needed for phase separation.

To investigate the effect of nonlocal interactions further, we note that two general classes of kernels can be identified based on whether the spinodal exhibits macroscopic modes or not. Macroscopic modes require that $q_s = 0$ is a solution to Eq. (4), implying $\hat{K}'(q_s) = 0$. Consequently, the Fourier-transformed kernel must be differentiable at $q = 0$. This requirement immediately excludes kernels that decay algebraically for large distance in real space since the Paley-Wiener theorem implies that the Fourier transform of such functions is not analytic in the vicinity of $q = 0$ [38]. In contrast, analytic kernels that decay exponentially in real space exhibit an analytic Fourier transform, which implies $\hat{K}'(0) = 0$ since $\hat{K}(q) = \hat{K}(-q)$. In this case, $q = 0$ is always a solution to Eq. (4) for a particular f_s'' , although $\omega(q_s)$ is not necessarily a maximum and thus the most unstable mode. In summary, kernels that decay algebraically (long-range interactions) cannot exhibit macroscopically large modes at the spinodal boundary, suggesting that indefinite coarsening is impossible. In contrast, exponentially decaying kernels (short-range interactions) can in principle exhibit long-term coarsening. To understand the influence of nonlocal interactions in more detail, we next discuss particular examples of these two classes.

We first study long-range interactions, whose interaction kernels fall off algebraically in space. Examples of such interactions include electrostatics [39], the Ohta-Kawasaki free energy of block copolymers [13,14], systems with short-range attraction and long-range repulsion [40–43], and single molecule chemical reactions with linear kinetics [22–27]. All of these interactions can be described by the generic form $\hat{K}_r(q) = \epsilon / q^n$, where $\epsilon > 0$ sets the energy scale and $n \leq d$ in d spatial dimensions. Such long-range interactions require additional constraints for the existence of equilibrium states [44], e.g., overall charge neutrality for Coulomb interactions, since otherwise the free energy density becomes extensive with system size. The general condition $\int d\mathbf{x} g[\bar{\phi}] = 0$ is satisfied by our choice $g[\phi] = \phi - \bar{\phi}$, but it forbids strictly positive or negative $g[\phi]$, e.g., for gravitational forces. Equilibrium states with long-range interactions generally involve screening [44], i.e., a positive $g[\bar{\phi}]$ region shelled by a negative $g[\bar{\phi}]$ region, resulting in regular patterns arranged in a hexagonal grid (in $d = 2$). The typical pattern length scale is governed by

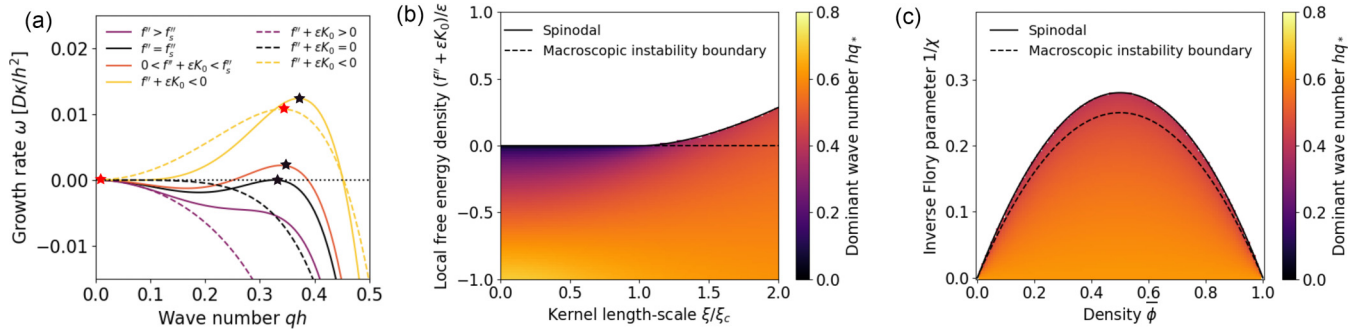


FIG. 1. Short-range interactions induce transition from homogeneous phase coexistence to regular patterns. (a) Dispersion relation $\omega(q)$ as a function of wave number q given by Eqs. (3) and (5) for $\xi < \xi_c \equiv h\sqrt{\kappa/\epsilon}$ (dashed) and $\xi > \xi_c$ (solid) for various f'' putting the system outside, at, and inside the spinodal boundary (colors). The dominant wave number q_* (stars, black for $\xi > \xi_c$ and red otherwise) is finite inside the spinodal region; at the spinodal boundary (black lines) $q_* = q_s$ continuously increases from $q_s = 0$ to $q_s \neq 0$ as ξ crosses ξ_c . (b) Dominant wave number q_* (Appendix A) as a function of ξ/ξ_c and f'' for fixed $\lambda/h = 1.4$. Along the spinodal (solid black line), q_s transitions continuously from $q_s = 0$ to $q_s > 0$ at $\xi = \xi_c$. Macroscopic modes with $q \rightarrow 0$ are unstable below the dashed line, corresponding to the yellow lines in panel (A). (c) q_* as a function of overall density ϕ and interaction strength χ for $\xi/\xi_c = 1.6$ and f given by Eq. (7) with $\kappa = \chi$. (a)–(c) $\lambda/h = 1.4$.

the dominant spinodal mode $q_s = (n\epsilon/2h^2\kappa)^{1/(n+2)}$ following from Eq. (4b). Inserting q_s into Eq. (4a) yields the spinodal condition $f_s'' = -h^2\kappa q_s^2(n+2)/n$ showing that long-range interactions always stabilize homogeneous states since $f_s'' < 0$. In summary, long-range interactions require stronger attractive forces to induce phase separation, and once patterns form they arrest at a finite length scale due to screening.

We next study short-range interactions, which are described by kernels $K_{sr}(|x|)$ that decay exponentially for large distance. Examples include nonlocal elasticity [17,20,21], viscoelastic fluids [18], bilayer membranes [33], and other forms of differential forces [45]. Since the Fourier transform of such interaction kernels is analytic [38], we describe them by a generic Taylor expansion around $q = 0$,

$$\hat{K}_{sr}(q) \simeq \epsilon[K_0 + \xi^2 q^2(-1 + \lambda^2 q^2)], \quad (5)$$

where we truncate the expansion at fourth order of q . The length scales ξ and λ characterize spatial dependencies of the interaction, ϵ sets the energy scale, and the constant K_0 controls whether nonlocal interactions are overall attractive ($K_0 < 0$) or repulsive ($K_0 > 0$). We consider kernels that decay at intermediate distances ($\xi > 0$) and are positive for $q \rightarrow \infty$ to stabilize homogeneous regions ($\lambda > 0$).

To see whether short-range interactions exhibit finite pattern sizes, we analyze the spinodal mode q_s by inserting Eq. (5) into Eq. (4). We find that Eq. (4b) always permits the solution $q_s = 0$, but $q_s \neq 0$ is also possible. The stability of the system is controlled by the mode with the largest growth rate ω , so we insert both solutions into Eq. (4a) and determine the one with maximal f_s'' . This approach yields the spinodal conditions

$$f_s'' = \epsilon[-K_0 + \xi^2 \lambda^2 q_s^4], \quad q_s^2 = \begin{cases} \frac{\xi^2 - \xi_c^2}{2\lambda^2 \xi^2}, & \xi > \xi_c, \\ 0, & \xi \leq \xi_c, \end{cases} \quad (6)$$

where $\xi_c = h\sqrt{\kappa/\epsilon}$ emerges as a critical length, which reveals a competition between the short-range kernel and interface properties. For $\xi \leq \xi_c$, we find $q_s = 0$, implying that F has a (local) minimum for $q = 0$, so coarsening will always

stabilize the system and macroscopic modes ($q \rightarrow 0$) dominate, similar to the case without nonlocal interactions. In contrast, for $\xi > \xi_c$ the free energy F exhibits a minimum at finite q_s , suggesting that coarsening is suppressed. The transition at $\xi = \xi_c$ is continuous since q_s increases continuously from $q_s = 0$. In summary, we find that systems with short-range interactions exhibit a finite spinodal mode q_s for sufficiently large ξ .

To learn more about the two qualitatively different regimes identified in Eq. (6), we next analyze the dispersion relation $\omega(q)$ given by Eq. (3). We first consider the case $\xi < \xi_c$, where stability is governed by macroscopic modes with $q \rightarrow 0$ and the spinodal is given by the condition $\omega'(0) = 0$ [dashed lines in Fig. 1(a)]. Inside the spinodal region, the fastest-growing mode is finite, but arbitrarily small modes ($q \rightarrow 0$) are unstable, so coarsening will take place, similar to systems without nonlocal interactions.

In the contrasting case $\xi > \xi_c$, stability is governed by the finite mode q_s given by Eq. (6). This finite mode corresponds to a local maximum in the dispersion relation $\omega(q)$, which is already present even when the system is stable [violet line in Fig. 1(a)]. The length scale corresponding to this local maximum might be detectable in the fluctuation spectrum of this stable system. For $f'' < f_s''$, the system becomes unstable ($\omega(q_*) > 0$), but if $f'' > \epsilon K_0$, macroscopic modes are still stable ($\omega'(0) < 0$). This implies a band of unstable modes (orange line), suggesting that patterns with finite length scale form. In contrast, macroscopic modes become unstable ($\omega'(0) > 0$) for $f'' < \epsilon K_0$, implying that coarsening could take place (yellow line). However, the spinodal mode q_s is still finite, suggesting patterns of a finite length scale will persist since this mode also satisfies $\partial_q(\delta^2 F/\delta\phi^2)|_{q=q_s} = 0$ and therefore is a local minimum of the free energy. We thus expect that an initial instability leads to patterns corresponding to the dominant wave number q_* , which then coarsen until a length scale governed by q_s . Note that the transition at $\xi = \xi_c$ qualitatively influences the spinodal mode, but it does not affect q_* [Fig. 1(b)]. We therefore hypothesize that the effects of the transition at ξ_c will become evident only at

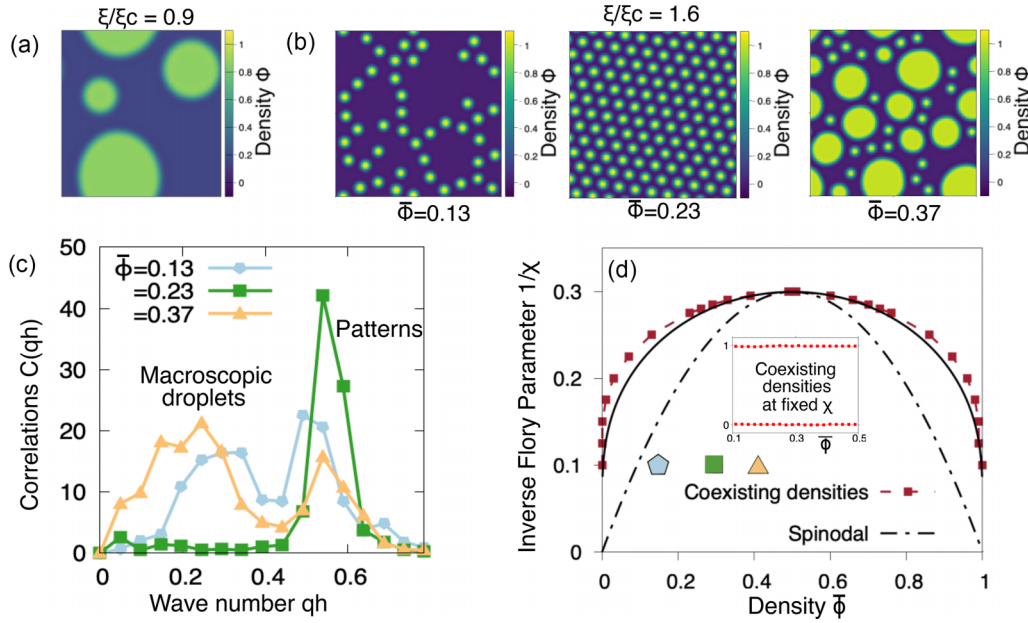


FIG. 2. Patterned phase can coexist with homogeneous phase for short-range interactions. (a), (b) Density fields $\phi(x)$ from 2D numerical integration (Appendix B) of Eqs. (1) and (2) with f given by Eq. (7) with $1/\kappa = 1/\chi = 0.1$, and short-range interactions given by Eq. (5) with $\lambda/h = 1.4$ and (a) $\xi/\xi_c = 0.9$ and (b) $\xi/\xi_c = 1.6$. (c) Density correlations in Fourier space for different wave numbers q for each of the three snapshots in panel (b). (d) Coexisting densities (red symbols) as function of $\bar{\phi}$ and χ are approximated by effective binodal (black line, without nonlocal interactions), indicating a first-order phase transition in two dimensions. Inset shows that changing $\bar{\phi}$ at fixed $\chi = 0.1$ does not affect coexisting densities. Symbols indicate values of $\bar{\phi}$ in panels (b) and (c). In all panels, we have used $\Lambda = \bar{\phi}(1 - \bar{\phi})$ (see Appendix B for details).

late times. Our results based on the analysis of the linearized equation suggest that systems with $\xi < \xi_c$ essentially behave like standard phase-separating systems without nonlocal interactions. In contrast, systems with $\xi > \xi_c$ seem to exhibit a finite wave length akin to pattern formation. One interesting property of the model with short-range interactions is thus a transition between pattern formation and phase separation at $\xi = \xi_c$.

To analyze the interplay between phase separation and nonlocal interactions further, we next consider the full nonlinearities dominating the late-time dynamics. To assess our hypothesis that ξ determines the long-time behavior and test the influence of short-range interactions in more detail, we investigate equilibrium configurations for the particular choice of a Flory-Huggins free energy [10,11],

$$f[\phi] = \frac{k_B T}{v} \left[\phi \ln \phi + (1 - \phi) \ln(1 - \phi) - \frac{\chi}{2} \phi^2 \right], \quad (7)$$

which is commonly used in the context of phase separation. The first two terms on the right represent translational entropy, while the term proportional to the nondimensional parameter χ accounts for pairwise interactions with $k_B T$ the thermal energy and v the relevant molecular volume. In addition, we consider the case where the short-range kernel given by Eq. (5) vanishes for large q for simplicity ($K_0 = 0$). In this case, the overall effect of short-range interactions is to destabilize the system and shift the spinodal to higher values of $1/\chi$ [Fig. 1(c)], implying phase separation takes place at weaker attractive interactions χ when short-range nonlocal interactions are present.

To go beyond the linear regime, we numerically evolve the dynamics given by Eqs. (2), (5), and (7); see Appendix B. We find that full coarsening only takes place for $\xi < \xi_c$ [Fig. 2(a)], whereas all examples with $\xi > \xi_c$ exhibit somewhat regular patterns [Fig. 2(b)], consistent with the finite spinodal mode q_s in this case. However, these numerical results indicate that patterns often contain more than one dominant length scale: At low overall density $\bar{\phi}$ [left panel in Fig. 2(b)], we observe the coexistence of a dilute homogeneous phase with patterns. For slightly larger $\bar{\phi}$, we observe a pure patterned phase with one dominant length scale (middle panel). At even larger $\bar{\phi}$, the system exhibits coexistence of dense homogeneous phases with patterns (right panel). These visually apparent transitions are corroborated by the correlations of the density fields, which exhibit two peaks for the complex patterns, whereas the pure patterned phase exhibits a single peak [Fig. 2(c)]. Note that when two peaks exist, only the peak at higher q corresponds to a length scale emerging from nonlocal interactions. In contrast, the peak at lower q corresponds to macroscopic phase separation, which is visible in the large circular regions in Figs. 2(a) and 2(b). We observe multiple distinct regions of the macroscopic phase in our simulations since the coarsening dynamics are very slow and we are limited by computation times. However, we checked that the peak corresponding to the low q gradually moves to lower values (Appendix C), indicating coarsening, and we predict that it would continue to do this until limited by system size. Taken together, our numerical simulations indicate that equilibrium states with short-ranged interactions exhibit patterned regions for $\xi > \xi_c$, which can sometimes coexist with homogeneous phases.

To characterize the patterns further, we next analyze the average densities of low-density and high-density regions (see Appendix B for details). The inset of Fig. 2(d) shows that these densities are independent of the total density $\bar{\phi}$, similar to coexisting densities in phase separation without nonlocal interactions. This is surprising since individual droplets in the patterned phase are not thermodynamic phases, so arguments from the thermodynamics of phase coexistence need not apply. Rather, all droplets together form the thermodynamic patterned phase and only the average density in this phase is thermodynamically constrained. Inspired by this observation, we next ask how measured densities vary with the interaction parameter χ at fixed overall density $\bar{\phi}$. Figure 2(d) shows that measured densities get closer to 0.5 for larger χ^{-1} , similar to coexisting densities in systems without nonlocal interactions. In fact, the family of densities (red dots) is well described by the binodal (black line) of an effective system without nonlocal interactions, but with a shifted interaction parameter $\tilde{\chi} = \chi + f_s''$. This observation is consistent with our previous conclusion that nonlocal short-range interactions can promote ($f_s'' > 0$) or suppress ($f_s'' < 0$) phase separation. This analysis suggests that the system exhibits a first-order phase transition, indicating that suddenly a patterned phase with finite amplitude emerges when χ is increased (except at the critical point).

The phases we observe for $\xi > \xi_c$ are similar to those observed in the conserved Swift-Hohenberg model, e.g., used to describe crystal growth [46–48]. In fact, the free energy given by Eq. (1) with the generic short-range kernel given by Eq. (5) can be mapped directly to a Swift-Hohenberg-like free energy (Appendix D),

$$F_{\text{SH}} = \int d\mathbf{x} \left\{ f[\phi(\mathbf{x})] - f_s'' \frac{\phi^2(\mathbf{x})}{2} \right\} + \frac{\epsilon \xi^2 \lambda^2}{2} \int d\mathbf{x} \{ \phi(\mathbf{x}) (\nabla^2 + q_s^2)^2 \phi(\mathbf{x}) \}, \quad (8)$$

where we have assumed that $\xi > \xi_c$, implying $q_s > 0$. Consequently, modes with $q = q_s$ minimize the nonlocal part of the free energy (second line), suggesting that patterns with such a length scale could be stable. However, the detailed equilibria depend on all terms in Eq. (8) and thus the precise form of the local free energy f .

The minima of F_{SH} have been studied for the case where $f[\phi]$ is a polynomial in ϕ , e.g., in the context of phase-field models of crystal growth [46–48]. References [46–50] contain detailed calculations of the corresponding phase diagram, which shows the same qualitative behavior described above, including coexistence of homogeneous phases and patterns. Moreover, Ref. [50] also shows that fixing the density $\bar{\phi}$ but changing a control parameter analogous to $1/\chi$ leads to a continuous phase transition in one spatial dimension, but a discontinuous transition in two or higher dimensions, the latter being consistent with our observations in two dimensions. The similarity in the phase diagrams suggests that results from phase-field descriptions of crystal growth can be transferred to models describing phase separation with short-range interactions, where f given by Eq. (7) is more natural. In particular, we expect that the amplitude equations derived in Ref. [50]

also hold for f given by Eq. (7) when χ and $\bar{\phi}$ are sufficiently close to the critical point. Similarly, finite systems should exhibit a series of bifurcations, one for each droplet that is formed/removed from the system as density is changed [50]. Finally, disturbances in these bifurcations at sufficiently low $1/\chi$ are expected to be directly related to the emergence of macroscopic droplets that we describe here [47,48]. Taken together, the mapping to the conserved Swift-Hohenberg model explains similar behavior observed across different systems that couple phase separation with effective short-range interactions [17,18,20,21,33,45,51].

Discussion. We have explored how nonlocal interactions affect phase separation. Similar to classical systems, the homogeneous state is stable if the driving force toward phase separation is weak (large f''). However, if phase separation takes place, nonlocal interactions affect the system qualitatively. In particular, long-range (algebraic) interactions always form regular patterns due to screening. In contrast, short-range interactions only slightly modify the classical behavior if they are weak ($\epsilon \xi^2 < \kappa h^2$). However, strong short-range interactions exhibit patterns, which can coexist with homogeneous macroscopic phases, depending on the overall composition. In this case, the free energy can be mapped to a conserved Swift-Hohenberg model, revealing how different short-range interactions exhibit universal behavior.

Our generic analysis provides a guideline to interpret results from different phase-separating systems with complex interactions. For instance, the mapping to the Swift-Hohenberg model resolves a discrepancy observed in models of phase separation with nonlocal elasticity with a Gaussian kernel [20,51]. While Ref. [51] found a first-order transition in three dimensions, Ref. [20] discovered a continuous transition in one dimension, both consistent with the Swift-Hohenberg model. In contrast, experiments suggest a continuous phase transition in three-dimensional elastic gels [19,52], suggesting that neither theory captures all relevant details. More generally, we expect that our results provide a suitable classification for many complex phase-separating systems.

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Data availability. Source code for running the simulations is available in Ref. [53].

Appendix A: Fastest-growing mode. A necessary condition for the fastest-growing mode q_* is $\omega'(q_*) = 0$, where $\omega(q)$ is given by Eq. (3). Hence,

$$q_* \left(f''(\bar{\phi}) + 2h^2 \kappa q_*^2 + \hat{K}(q_*) + \frac{q_*}{2} \hat{K}'(q_*) \right) = 0. \quad (A1)$$

For the short-range interactions given by Eq. (5), this becomes

$$q_*^2 = \frac{\xi^2 - \xi_c^2}{3\lambda^2 \xi^2} \left[1 \pm \sqrt{1 - \frac{3(f'' + \epsilon K_0)\lambda^2 \xi^2}{\epsilon(\xi^2 - \xi_c^2)^2}} \right], \quad (A2)$$

which reduces to Eq. (6) for q_s when $f'' = f_s''$. Here, $\pm$ represent the solutions for $\xi > \xi_c$ (+) and for $\xi < \xi_c$ (−).

Appendix B: Simulation details. We integrate Eq. (2) numerically in Fourier space using a pseudospectral method

[54]. To avoid numerical problems with the logarithm in the free energy given by Eq. (7), we regularize $\ln(\phi)$ as

$$\tilde{\ln}(\phi) = \begin{cases} \ln(\phi), & \text{if } \phi > \delta, \\ \ln(\delta) - \frac{\phi - \delta}{\delta} + \frac{(\phi - \delta)^2}{2\delta^2}, & \text{if } \phi \leq \delta. \end{cases} \quad (\text{B1})$$

To recover Fickian diffusion for $\phi \rightarrow 0$ and to preserve symmetry with respect to $\phi = \frac{1}{2}$, we use the mobility

$$\Lambda(\phi) = D \left\{ \frac{\partial^2}{\partial \phi^2} [\phi \tilde{\ln}(\phi) + (1 - \phi) \tilde{\ln}(1 - \phi)] \right\}^{-1}, \quad (\text{B2})$$

where D is a constant. This recovers the usual expression $\Lambda = D\phi(1 - \phi)$ for $\delta < \phi < 1 - \delta$ and guarantees that the diffusivity $\Lambda(\phi)f''$ remains constant and equal to D for ideal systems ($\chi = 0$) for all ϕ . Similar relations are used in macroscopic fluctuation theory [55], but this aspect is often overlooked in the literature on Cahn-Hilliard models with a regularized logarithm, which could result in unphysical discontinuities in the diffusivity. We use $\delta = 10^{-2}$ in all our simulations, and we verified that lowering δ to 10^{-4} did not change any results significantly.

We extract coexisting densities inside and outside droplets for large times by identifying the two dominant peaks in the density histograms. To do so, we bin the density space $\phi \in (0, 1)$ into intervals of width 0.01 and compute corresponding histogram. We then identify the maxima in the histogram in the low-density ($\phi < 0.5$) and high-density ($\phi > 0.5$) regions, respectively, which is the data reported in Fig. 2(d). In the patterned phase, the area occupied by interfaces between microscopic droplets is comparable to the area occupied by their interiors and, together with the observation that the density profile does not change monotonically as an interface is crossed, our method to determine the coexisting densities leads to small deviations of the true peaks toward slightly higher values (e.g., when compared to the density at the center of the droplet) or slightly lower values (i.e., at the middle point between two microscopic droplets). This deviation is

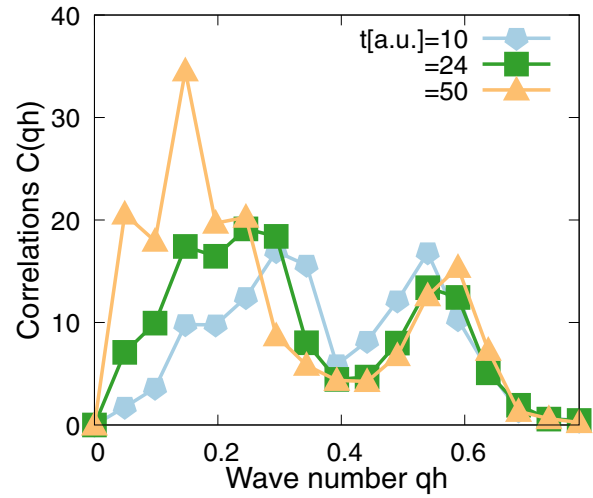


FIG. 3. Macroscopic droplets coarsen to system size. Structure factor C as a function of wave number q at density $\bar{\phi} = 0.37$ for three different times t . The peak at smaller q shifts toward lower q , indicating coarsening of macroscopic droplets. Parameters are as in Fig. 2.

expected, since microscopic droplets do not form a thermodynamic phase, meaning “interface” contributions do not vanish for large systems.

Appendix C: Coarsening of macroscopic droplets. In the main text, we identified a coexistence of a macrophase with a patterned phase. However, the numerical evidence for this situation is complicated by finite-size effects. For instance, the structure factor shown in Fig. 2(c) exhibits two peaks. To show that these peaks correspond to the patterned phase (at large wave number q) and macrophases (at small q), respectively, we here present evidence for the coarsening of macroscopic: Figure 3 shows that the structure factor for a fixed density $\bar{\phi} = 0.37$ at different times clearly shifts toward lower q for the putative macrophase, whereas the peak corresponding to the patterned phase remains stable.

Appendix D: Mapping to Swift-Hohenberg free energy. The expansion of the Fourier-transformed short-range kernel given by Eq. (5) corresponds to a gradient expansion in real space, $K_{\text{sr}}(|\mathbf{r}|) \simeq \epsilon[K_0\delta(|\mathbf{r}|) + \xi^2\nabla^2 + \xi^2\lambda^2\nabla^4]$. We thus consider the full nonlocal free energy given by Eq. (1) for a generic isotropic kernel expanded as $K(|\mathbf{x} - \mathbf{x}'|) = \epsilon K_0 + \sum a_n \nabla^{2n} \delta(\mathbf{x} - \mathbf{x}')$,

$$F = \int d\mathbf{x} \left\{ f[\phi(\mathbf{x})] + \frac{h^2\kappa}{2} |\nabla\phi(\mathbf{x})|^2 \right\} + \frac{1}{2} \int d\mathbf{x} g[\phi(\mathbf{x})] \left[\epsilon K_0 + \sum_{n=1} a_n \nabla^{2n} \right] g[\phi(\mathbf{x})]. \quad (\text{D1})$$

Integrating the κ term by parts gives

$$F = \int d\mathbf{x} \left\{ f[\phi(\mathbf{x})] - \frac{h^2\kappa}{2} \phi \nabla^2 \phi \right\} + \frac{1}{2} \int d\mathbf{x} g[\phi(\mathbf{x})] \left[\epsilon K_0 + \sum_{n=1} a_n \nabla^{2n} \right] g[\phi(\mathbf{x})], \quad (\text{D2})$$

where we assume that boundary terms vanish, e.g., because of periodic boundary conditions or because $g[\phi]$ vanishes at the system boundary. Truncating at $n = 2$ and letting $g[\phi(\mathbf{x})] = \phi(\mathbf{x}) - \bar{\phi}$,

$$F = \int d\mathbf{x} \left\{ f[\phi(\mathbf{x})] + \frac{\epsilon K_0 \phi^2(\mathbf{x})}{2} \right\} + \int d\mathbf{x} \left\{ \frac{\phi(\mathbf{x})}{2} [(a_1 - h^2\kappa)\nabla^2 + a_2\nabla^4] \phi(\mathbf{x}) \right\}. \quad (\text{D3})$$

Completing the square in the square bracket and identifying $q_s^2 = (a_1 - h^2\kappa)/(2a_2) > 0$,

$$F_{\text{SH}} = \int d\mathbf{x} \left\{ f[\phi(\mathbf{x})] + \left(\epsilon K_0 - \frac{q_s^4 a_2}{2} \right) \frac{\phi^2(\mathbf{x})}{2} \right\} + \frac{a_2}{2} \int d\mathbf{x} \phi(\mathbf{x}) (\nabla^2 + q_s^2)^2 \phi(\mathbf{x}), \quad (\text{D4})$$

gives Eq. (8) with $a_1 = \epsilon \xi^2$ and $a_2 = \epsilon \xi^2 \lambda^2$.

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