

Advection selects pattern in multistable emulsions of active droplets

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(Received 30 September 2025; accepted 4 January 2026; published 30 January 2026)

Controlling the size of droplets, for example, in biological cells, is challenging because large droplets typically outcompete smaller droplets due to surface tension. This coarsening is generally accelerated by hydrodynamic effects, but active chemical reactions can suppress it. We show that the interplay of these processes leads to three different dynamical regimes: (1) Advection dominates the coalescence of small droplets, (2) diffusion leads to Ostwald ripening for intermediate sizes, and (3) reactions finally suppress coarsening. Interestingly, a range of final droplet sizes is stable, of which one is selected depending on initial conditions. Our analysis demonstrates that hydrodynamic effects control initial droplet sizes, but they do not affect the later dynamics, in contrast to passive emulsions.

DOI: [10.1103/hdls-nzyc](https://doi.org/10.1103/hdls-nzyc)

I. INTRODUCTION

Emulsions of multiple droplets provide spatial structures in biological cells [1–4] and synthetic applications [5,6]. In these examples, effective patterning requires controlled droplet sizes and positions. These aspects are affected by physical processes such as diffusion, advection, and chemical reactions [4]. In particular, diffusion generally causes droplet coarsening, either because molecules diffuse from smaller to larger droplets (Ostwald ripening [7,8]) or because droplet diffusion leads to coalescence [9]. Hydrodynamic advection generally accelerates coarsening [9–11], whereas driven chemical reactions can suppress coarsening [12–16]. However, how the interplay of these processes organizes emulsions, and in particular selects length scales, is poorly understood.

II. RESULTS

A. Chemically active droplets couple diffusion, advection, and reactions

To unveil the interplay between diffusion, advection, and reactions, we study a minimal model of an emulsion described by an isothermal fluid comprising two chemical species A and B. We assume incompressibility, so that the system's state is described by only the number concentration $c(\mathbf{r}, t)$ of species A, which evolves as

$$\partial_t c + \mathbf{v} \cdot \nabla c = \Lambda \nabla^2 \mu - k(c - c_0), \quad (1)$$

where the left-hand side describes the material derivative with velocity field $\mathbf{v}$, whereas the right-hand side accounts

for diffusion with constant mobility Λ and a linear reaction with rate k describing the conversion between A and B. This conversion reaction describes a driven reaction since it is not derived based on thermodynamic principles [4,17,18]. In particular, the activity-dependent reaction equilibrium at $c = c_0$ is distinct from the thermodynamic equilibrium described by the free energy F of the system. In contrast, the diffusive flux is driven by gradients in the exchange chemical potential $\mu = \delta F / \delta c$, implying that it biases the system toward the free energy minimum. To analyze the interplay of passive diffusion and active reactions, we consider a free energy of the Ginzburg-Landau form [19]

$$F = \int dV \left[\frac{a}{2} c^2 \left(1 - \frac{c}{c_{\text{in}}} \right)^2 + \frac{\kappa}{2} |\nabla c|^2 \right], \quad (2)$$

where a sets the energy scale, c_{in} denotes the concentration difference between coexisting phases, and κ penalizes gradients, implying a surface tension of $\gamma = \frac{1}{6} c_{\text{in}}^2 \sqrt{\kappa a}$ and an interfacial width of $w = \sqrt{\kappa / a}$ [19]. The concentrations of coexisting equilibrium phases are thus $c = 0$ and $c = c_{\text{in}}$, which are distinct from the reaction equilibrium at $c = c_0$ since the reactions are active. This imbalance drives continuous fluxes and flows in the system, which we analyze in small systems with low velocities, implying that inertial effects are negligible. Consequently, the velocity field $\mathbf{v}$ is governed by the Stokes equations,

$$\eta \nabla^2 \mathbf{v} - \nabla p = c \nabla \mu, \quad \nabla \cdot \mathbf{v} = 0, \quad (3)$$

describing balanced viscous stresses proportional to viscosity η , hydrostatic pressure p , and equilibrium stresses.

We consider periodic boundary conditions since we are interested in internally created flows and neglect external influences. To judge the relative magnitude of the terms in Eq. (3), we use the interfacial width w as a relevant length scale and estimate the associated velocity scale as $v \sim \gamma / \eta$, implying the Péclet number $\text{Pe} = w^2 a c_{\text{in}}^2 / (D \eta)$,

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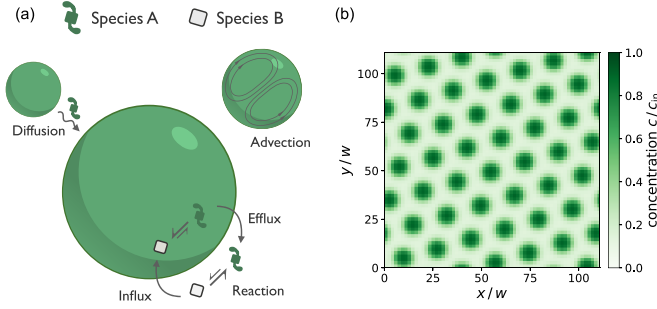


FIG. 1. Turnover causes hexagonal droplet pattern. (a) Schematic of our model involving two species that interconvert and phase separate from each other. (b) Simulation snapshot showing $c(\mathbf{r})$ at stationarity for $Pe = 0$, $c_0 = 0.35 c_{in}$, $k = 0.02/\tau$, $\tau = w^2/D$, and $w = \sqrt{\kappa/a}$.

where $D = \Lambda a$ is the relevant diffusivity and we ignored a proportionality factor of $\frac{1}{6}$ to arrive at simple nondimensional equations (see Sec. I of the Supplemental Material [20]). To estimate realistic values for Pe , we consider molecules of sizes comparable to the interfacial width w , concentration $c_{in} \sim w^{-3}$, and interaction energy $ac_{in} \sim k_B T$, where $k_B T$ is the thermal energy. Using the Stokes-Einstein relation to obtain $D \sim k_B T / (6\pi\eta w)$, we find $Pe \sim 19$. Since Pe is not small compared to 1, advection is likely relevant in our system.

B. Chemical reactions arrest droplet coarsening

1. Stationary droplets are larger for weaker reactions

We start by investigating the interplay between diffusion and reactions, neglecting hydrodynamics for now ($Pe = 0$, implying $\mathbf{v} = \mathbf{0}$). Numerical simulations in two dimensions suggest that reactions suppress coarsening, so that droplets form a hexagonal pattern [Fig. 1(b)]. To understand how chemical reactions alter material fluxes to suppress coarsening, we map the active system described by Eq. (1) to an equivalent passive system with nonlocal interactions. In particular, we rewrite the dynamics using a surrogate free energy [21,22]

$$\tilde{F} = F + \frac{k}{2\Lambda} \int dV \psi(c - c_0), \quad (4)$$

where the potential ψ is governed by the Poisson equation

$$\nabla^2 \psi = -(c - c_0). \quad (5)$$

This potential captures the effect of reactions as nonlocal interactions, so that the dynamics $\partial_t c = \Lambda \nabla^2 \delta \tilde{F} / \delta c$ are identical to Eq. (1) when $\mathbf{v} = \mathbf{0}$. Equations (4) and (5) reveal that the reactions are analogous to an electrostatics problem, where $(c - c_0)$ acts as a charge density creating an electrostatic potential ψ . Droplets can thus be interpreted as positively charged disks, surrounded by a negative charge cloud extending over the reaction-diffusion length $\xi = \sqrt{D/k}$ [4,23]. If the average concentration of the system reached its stationary state value c_0 , the mathematical mapping to the equilibrium system exhibits the same dynamics as the original system given by Eq. (1). Consequently, minimizing $\tilde{F}$ allows us to identify stationary states of the original system. Our numerical minimization described in Sec. II B of the Supplemental

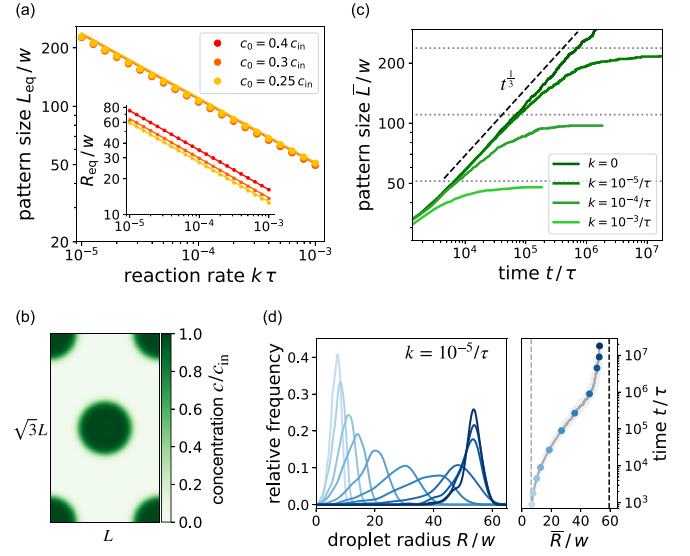


FIG. 2. Reaction rate controls stationary pattern size. (a) Pattern size L_{eq} , determined numerically (symbols) and predicted by Eq. (6) (lines), as a function of reaction rate k for various average compositions c_0 . Inset shows corresponding radii. (b) Concentration profile $c(\mathbf{r})$ for $k = 10^{-3}/\tau$ in a unit cell with adjustable length L . (c) Average pattern size $\bar{L}$ as a function of time for various k , where $\bar{L} = 2\pi^{-1/2} (V_{sys}/N)^{1/2}$ from droplet count N and system size V_{sys} . Numerical data are compared to L_{eq} from Eq. (6) (gray dotted lines) and a $t^{1/3}$ power law (black dashed line). (d) Distribution of droplet radii R (left) and the average radius $\bar{R}$ (right) for various time points for $k = 10^{-5}/\tau$. Numerical data are compared to the prediction from spinodal decomposition (gray dashed line) and $R_{eq} = \sqrt{c_0/4c_{in}} L_{eq}$ (black dashed line). (a)–(d) Model parameters are $c_0 = 0.25 c_{in}$, $\tau = w^2/D$, and $w = \sqrt{\kappa/a}$.

Material [20] reveals that the pattern size, quantified by the center-to-center distance L , decreases with higher reaction rates k [Fig. 2(a)], consistent with previous work [22,24–26].

We next derive an analytical estimate of the pattern size L by focusing on a single droplet of radius R and approximating its environment as a spherically symmetric shell with radius $\frac{1}{2}L$ (see Sec. II C of the Supplemental Material [20]). To determine the relation between R and L , we focus on two-dimensional systems and approximate the concentration profile as $c(r) = c_{in} \Theta(R - r)$, which describes droplets without reactions and surface tension effects. Since the average concentration is c_0 in steady state, we conclude that $R/L = \sqrt{c_0/4c_{in}}$, based on a spherically symmetric system. Inserting this together with the solution of Eq. (5) into Eq. (4) leads to an expression for $\tilde{F}$ that only depends on L . Minimizing $\tilde{F}$ with respect to L yields (see Sec. II C of the Supplemental Material [20])

$$\frac{L_{eq}}{w} = \frac{4}{6^{1/3}} \left(\frac{c_{in}}{c_0} \right)^{1/2} \left[\frac{c_0}{c_{in}} - 1 - \ln \left(\frac{c_0}{c_{in}} \right) \right]^{-1/3} \left(\frac{w^2 k}{D} \right)^{-1/3}, \quad (6)$$

consistent with our data [Fig. 2(a)]. This expression is proportional to Eq. (58) in Ref. [22], but our prefactor approximates the data better. In any case, material turnover leads to stationary states with controlled droplet sizes with $L_{eq}, R_{eq} \propto k^{-1/3}$.

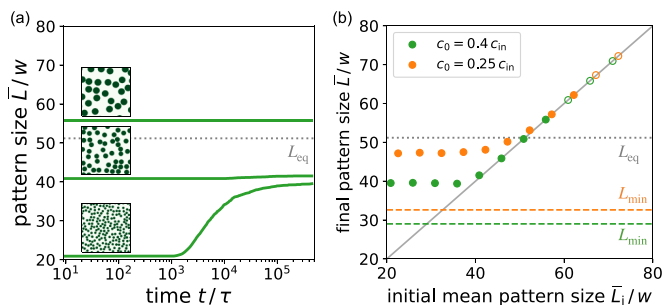


FIG. 3. Many stationary states are stable. (a) Pattern size $\bar{L}$ as a function of time t for three initial conditions with $c_0 = 0.4 c_{in}$ and differing droplet size (see snapshots and Sec. I A of the Supplemental Material [20]). (b) Final pattern size $\bar{L} = 2/\sqrt{\pi}(V_{sys}/N)^{1/2}$ as a function of the initial pattern size $\bar{L}_i$ for various c_0 . Colored dashed lines indicate minimal stable size L_{min} obtained from stability analysis. Open circles indicate shape instabilities. (a), (b) Gray dotted lines mark free energy minimum for $c_0 = 0.4 c_{in}$ given by Eq. (6). Model parameters are $k = 10^{-3}/\tau$, $\tau = w^2/D$, and $w = \sqrt{\kappa/a}$.

2. Chemically active droplets coarsen until their stationary size

We next analyze the approach toward stationary state. To analyze generic situations, we start with a slightly perturbed homogeneous state and simulate spinodal decomposition (see Sec. I A of the Supplemental Material [20]). Once proper droplets with a narrow size distribution form, we switch to an effective simulation method to cover long timescales [27]. The average pattern size $\bar{L}$ generally increases over time and saturates at a level that depends on k [Fig. 2(c)]. During the coarsening phase, we find $\bar{L} \propto t^{1/3}$ [Fig. 2(c)], consistent with Lifshitz-Slyozov-Wagner theory [28,29]. Moreover, our data suggest that the droplet size distribution is similar to the expected universal one during coarsening [28,29], but it becomes much narrower close to the stationary state [Fig. 2(d)] [16]. Interestingly, the pattern size saturates below the length scale predicted by Eq. (6) [Fig. 2(c)].

3. Chemical reactions introduce multistability

To understand the discrepancy between the predicted pattern size and the final state of the simulation, we next analyze more diverse initial conditions. Specifically, we vary the initial average droplet size, compensating the droplet count to keep the overall amount of material the same. Figure 3(a) shows that coarsening occurs if the initial droplets are small, whereas large droplets remain basically unchanged. This implies that there are many different (meta)stable stationary states with various pattern sizes above a minimal size. Our numerical analysis suggests that this minimal size depends on the average concentration c_0 [Fig. 3(b)].

To understand the minimal size of patterns, we next analyze their stability. Our simulations indicate that patterns coarsen by dissolving droplets, whereas coalescence is negligible in the late stage of coarsening. To determine the smallest droplet that can be stable, we consider a system comprising a droplet of radius R_1 , surrounded by droplets of radii R_2 at distance L , assuming spherical symmetry (see Fig. S3 of the Supplemental Material [20]). The dynamics of R_1 are governed by $\partial_t R_1 = (j_{in} - j_{out})/c_{in}$, where j_{in} and j_{out} are

the diffusive fluxes inside and outside of the interface [19]. These fluxes can be determined analytically assuming quasistationarity, implying that $\partial_t R_1$ can be expressed as a function of R_1 , R_2 , and L . A stability analysis for small perturbations around $R_1 = R_2 = R_*$ allows us to determine the minimal stable radius, R_{min} (see Sec. III of the Supplemental Material [20]). The corresponding predicted minimal pattern size L_{min} underestimates the smallest observed patterns [Fig. 3(b)], but it reveals the correct trends: Droplets can be smaller if there is more material (larger c_0). The dependence of L_{min} on c_0 is stronger than for L_{eq} , but both quantities scale as $k^{-1/3}$ (see Fig. S4B of the Supplemental Material [20] and Ref. [26]). The stability analysis predicts that patterns with droplets above the minimal size R_{min} are stable.

We next ask whether there is also a maximal pattern size L_{max} . Generally, large chemically active droplets exhibit shape instabilities [30], but these are not captured by the simulation method used in Fig. 3(a) [27]. To test whether instabilities would occur, we investigate the final states of our effective simulation using detailed simulations of Eq. (1) (see Sec. IV of the Supplemental Material [20]). Indeed, we find that large droplets exhibit instabilities [open symbols in Fig. 3(b)], suggesting that such systems would evolve toward smaller droplets.

In summary, we find that chemically active emulsions can in principle exhibit a range of pattern sizes, distributed around the theoretical prediction of L_{eq} . The observed pattern size thus depends on the initial condition. The surrogate equilibrium model given by Eq. (4) suggests that this multistability can be interpreted as a kinetic arrest, and noise would allow the system to explore various states. This implies that the droplet count varies over time by nucleation, dissolution, coalescence, and splitting of droplets. These effects are generally driven by molecular diffusion and Brownian motion of entire droplets, which depends on hydrodynamic effects.

C. Hydrodynamic advection accelerates coarsening by inducing coalescence

Finally, we ask how hydrodynamics affects the coarsening of chemically active droplets by considering $Pe > 0$. We perform numerical simulations of Eqs. (1)–(3), where we solve the Stokes equation using a pseudospectral method (see Sec. I of the Supplemental Material [20]). To form droplets effectively, we consider the spinodal decomposition regime. We find that hydrodynamics generally accelerates coarsening for all reaction rates (Fig. 4), consistent with literature [9–11]. This acceleration is less pronounced for a smaller average fraction c_0 (see Fig. S1 of the Supplemental Material [20]), indicating that it relies on a large droplet density.

For passive mixtures [$k = 0$; Fig. 4(a)], our data at early times are consistent with the theoretically expected scaling of $\bar{L} \propto t$ resulting from “coalescence-induced coalescence” in the viscous regime [9,10,31,32]. In contrast, we observe a transition to Ostwald ripening at large length scales and timescales, where the mean pattern size obeys $\bar{L} \propto t^{1/3}$. Interestingly, we observe a plateau region between these two regimes where $\bar{L}$ hardly changes, suggesting that Ostwald ripening is delayed. Note that the transition between the two regimes is opposite to what has been described for

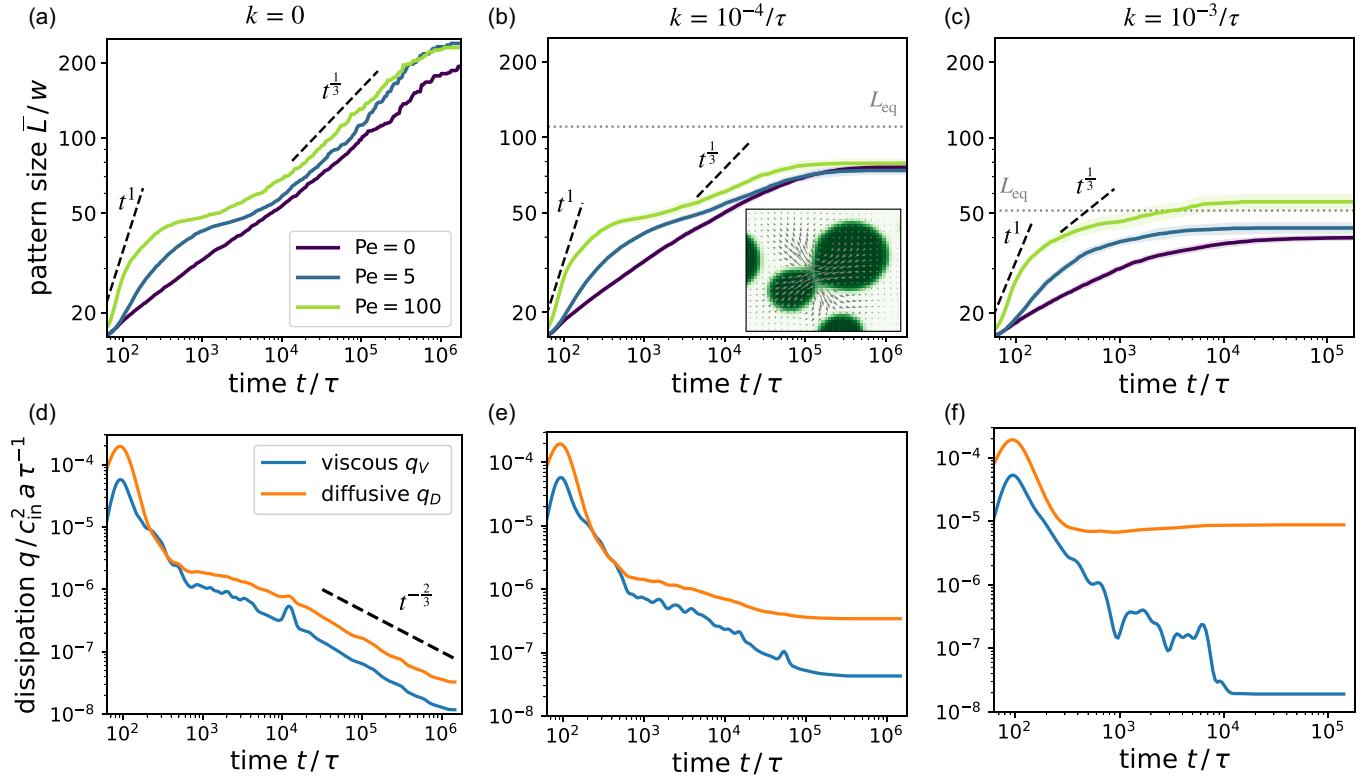


FIG. 4. Advection accelerates coarsening. Pattern size $\bar{L}$ (shaded area indicates standard deviation, $n = 20$) as a function of time t for various Péclet numbers $Pe = w^2 a c_{in}^2 / (D\eta)$ and reaction rates k (across panels). Black dashed lines show power laws $\bar{L} \propto t$ (left) and $\bar{L} \propto t^{1/3}$ (right). Gray dotted lines show L_{eq} calculated from Eq. (6). Inset in panel (b) shows the velocity field generated by two colliding droplets (gray arrows) for $k = 10^{-4}/\tau$ and $Pe = 100$. (d)–(f) Volume averaged viscous (blue) and diffusive (orange) dissipation corresponding to panels (a)–(c) for $Pe = 100$ (see Sec. V of the Supplemental Material [20]). The curves were smoothed by convolution with a Gaussian kernel with a standard deviation of two data points to make the plot more readable. Black dashed line in panel (d) shows a power law $\propto t^{-2/3}$. (a)–(f) Model parameters are $c_0 = 0.4 c_{in}$, $\tau = w^2/D$, and $w = \sqrt{\kappa/a}$.

bicontinuous structures in equivalent systems, where diffusion dominates at small scales (implying $\bar{L} \propto t^{1/3}$) and advection becomes important later (leading to $\bar{L} \propto t$) [33]. This difference might be caused by the fact that advection is less important in our system as judged by the smaller viscous dissipation compared to diffusive dissipation [Fig. 4(d)]. However, the identical scaling of the dissipations indicates that both advection and diffusion are relevant for the entire coarsening process of passive mixtures.

When mixtures are active ($k > 0$), coarsening must eventually cease since reactions limit the pattern sizes $\bar{L}$. For weak reactions, the initial coarsening dynamics are virtually identical to the passive case, including the ballistic regime, a transient plateau, and a phase involving Ostwald ripening [Fig. 4(b)]. As expected, $\bar{L}$ eventually reaches a stable pattern size, which is independent of Pe for weak reactions. Note that the Ostwald ripening regime does not quite reach the expected scaling $\bar{L} \propto t^{1/3}$, but we hypothesize that this would be the case for even smaller reaction rates, which are difficult to simulate. In contrast, the Ostwald ripening regime is completely absent for stronger reactions [Fig. 4(c)]. Concomitantly, we observe that the final pattern size depends on Pe (and on initial condition, evident from larger standard deviation), consistent with the multistability described above. Note that advection generally opposes shape deformations [34], suggesting larger maximal stable pattern sizes L_{max} . Indeed, we generally ob-

serve larger final pattern sizes for larger Pe if reactions are sufficiently strong [Fig. 4(c)]. This is likely caused by strong advection leading to fewer and larger droplets in the initial phase, which then remain in the final state. Taken together, advection selects the initial pattern size, essentially independent of k , but it does not affect the further coarsening significantly, so that the final pattern size is governed by the results shown in Fig. 3(b).

The relative importance of advection and diffusion can be quantified by the viscous and diffusive dissipation, respectively. Figures 4(d)–4(f) show that advection is relevant early, but decays with time, essentially independent of k [blue lines are similar in panels (d)–(f)]. The diffusive dissipation also peaks early, and decays similarly to the viscous dissipation during droplet coarsening. Interestingly, both dissipations are similar during the time where we observe a plateau in the pattern size [Figs. 4(a)–4(c)]. Beyond this point, diffusive dissipation dominates, and it plateaus at a much larger value compared to the viscous dissipation. This behavior indicates that energy induced by reactions leads to diffusive fluxes, whereas advection is negligible in the stationary state. This might be surprising since diffusive fluxes and the velocity field are driven by gradients in chemical potential. However, the fluid is incompressible ($\nabla \cdot \mathbf{v} = 0$), so that chemical potential gradients get projected onto a solenoidal subspace. Therefore, osmotic pressure gradients are balanced by gradients in

hydrostatic pressure. Accordingly, the velocity field remains small compared to diffusive fluxes for moderate advection strength (see Fig. S6 of the Supplemental material [20]). In contrast, large advection (large Pe) causes a dynamic instability leading to spatiotemporal chaos [35].

III. DISCUSSION

We showed that a chemical conversion reaction can arrest coarsening at various length scales. Generally, stronger conversion (larger k) leads to smaller length scales, although a range of scales are stable for each value of k . The final length scale thus also depends on initial conditions, and particularly the number of droplets that are present. We find that this initial droplet count depends strongly on advection, whereas advection is negligible in later stages.

Our work shows how regular patterns of droplets can be controlled by an interplay of chemical conversion and advection. These processes are directly relevant for synthetic systems of chemically active droplets [5,14], and they might affect biomolecular condensates in cells. We thus speculate that cells exploit the discussed mechanism to control their condensates. However, cellular systems are much more complex, and it is thus likely that additional processes affect condensates [4]. In particular, cellular flows, e.g., driven by

molecular motors, could further affect coarsening [36–38], and inertia could be important on larger length scales [39,40]. Moreover, cellular droplets exhibit significant thermal fluctuations, which would not only be relevant for nucleation [41], but could particularly allow the system to transition from one metastable state to another. In this case, we hypothesize that the most likely state exhibits a length scale close to L_{eq} given by Eq. (6). The multistability should then be interpreted as kinetic traps, potentially leading to glassy dynamics [42].

ACKNOWLEDGMENTS

We thank Noah Ziethen and Riccardo Rossetto for helpful discussions. We gratefully acknowledge funding from the Max Planck Society and the European Research Council (ERC, EmuSim, 101044662). S.K. acknowledges funding through a fellowship of the IMPRS for Physics of Biological and Complex Systems. S.K. acknowledges support by the study program “Biological Physics” of the Elitenetzwerk Bayern.

DATA AVAILABILITY

The data that support the findings of this article are openly available [43].

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