Hydrodynamically interrupted droplet growth in scalar active matter

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Suspensions of spherical active particles often show microphase separation. At a continuum level, coupling their scalar density to fluid flow, there are two distinct explanations. Each involves an effective interfacial tension: the first mechanical (causing flow) and the second diffusive (causing Ostwald ripening). Here we show how the negative mechanical tension of contractile swimmers creates, via a self-shearing instability, a steady-state life cycle of droplet growth interrupted by division whose scaling behavior we predict. When the diffusive tension is also negative, this is replaced by an arrested regime (mechanistically distinct, but with similar scaling) where division of small droplets is prevented by reverse Ostwald ripening.

Active matter continuously dissipates energy locally to perform mechanical work. In consequence, the dynamical equations of coarse-grained variables, such as particle density, break time-reversal symmetry. Examples include suspensions of spherical autophoretic colloids, which self-propel due to self-generated chemical gradients at their surfaces [1]. Experiments on several such suspensions have observed activity-induced phase separation that arrests at a mesoscopic scale [2–5]. A generic understanding of such nonequilibrium microphase separations can be sought at the level of continuum equations for a diffusive scalar concentration field, coupled to incompressible fluid flow. Such an approach is complementary to more detailed mechanistic modelling, in which particle motion and/or chemical fields are modelled explicitly [6–9]. By sacrificing detail, the resulting active field theory allows maximal transfer of ideas and methods from equilibrium statistical mechanics. Each distinct mode of activity can be modelled in a minimal fashion, allowing the competition between them to be studied. This knowledge base can inform the design of novel functional materials and devices with tunable properties [10].

Bulk active phase separation can arise through attractive interactions, as in the passive case [11], or, even for repulsive interactions, be motility-induced [12, 13]. At continuum level, when there is no orientational order in bulk, the only order parameter required for the particles is their scalar concentration $\phi(r,t)$. For 'wet' systems, with a momentum-conserving solvent rather than a frictional substrate, this is coupled to a fluid velocity field $\mathbf{v}(r,t)$ [14]. Operationally, the field theory of active scalar phase separation starts from the long-studied passive case, whose stochastic equations of motion are constructed phenomenologically, respecting symmetries and conservation laws, truncated at some consistent order in the fields and their gradients [11]. If $\phi$ is measured relative to the critical point for phase separation, this gives to leading order a symmetric free energy functional $\mathcal{F}[\phi] = \mathcal{F}[-\phi]$. The outcome is Model B (dry) or Model H (wet) [11, 15].

To make these theories active, we add a small number of leading-order terms, each of which breaks time-reversal symmetry via a distinct channel. The first adds to the chemical potential $\delta \mathcal{F}/\delta \phi$ a piece that is not the derivative of any free energy functional $\mathcal{F}$ (this is the $\lambda$ term in (2) below). The deterministic diffusive current then breaks detailed balance but remains curl-free. This channel is known to alter phase boundaries, but cannot arrest phase separation [16]. A second channel (the $\zeta$ term in (2) below) enters at the same order, but in the diffusive current directly. This can arrest phase separation, by creating an effectively negative interfacial tension in the diffusive sector [17], throwing the process of Ostwald ripening, whereby large droplets grow at the expense of small ones, into reverse. These two channels are, for dry systems, captured in Active Model B+$\lambda$ [17].

The third active channel, present for wet systems only, is the mechanical stress arising from self-propulsion. This is a bulk stress in systems with orientational order (where it leads to bacterial turbulence [14]), but for a scalar it is quadratic in $\nabla \phi$ (the $\tilde{\kappa} - \kappa$ term in (3) below), just like the passive thermodynamic stress for Model H [18]. It likewise drives fluid motion via interfacial curvature.

So far, the resulting 'Active Model H' has been explored only in the absence of the other two active channels, without noise, and for systems at the critical density, $\phi_0 \equiv \langle \phi \rangle = 0$ [19]. Under these conditions, activity can arrest separation, giving a dynamically fluctuating bicontinuous state. The effect of active stresses on droplet states, as might describe the cluster phases seen experimentally [1–4], remains unclear. Notably though, in passive systems, the interfacial stress rapidly becomes unimportant on moving away from bicontinuity, since well separated droplets recover spherical symmetry which precludes incompressible fluid flow. Wet and dry dilute passive droplets then behave similarly [15].

In this Letter we first extend the study of Active Model H, with noise, to off-critical quenches ($\phi_0 \neq 0$), where droplets or bubbles arise, and there address the competition between mechanical ($\tilde{\kappa}$) and diffusive ($\lambda, \zeta$) activity channels. We find that, contrary to the passive case, the mechanical stress plays a crucial role in droplet (or bubble) evolution; when sufficiently contractile, it can halt phase separation, causing large droplets to...
split in smaller ones. This balances the diffusive droplet growth due to Ostwald ripening, giving a steady state with a distinctive droplet life cycle. This represents ‘interrupted’ rather than ‘arrested’ phase separation, because the steady state is highly dynamic, and continues unchanged after all noise is switched off. This contrasts with the microphase separation that results from reverse Ostwald ripening, where switching off noise leads to a fully arrested, static assembly of monodisperse droplets [17].

Crucially, the droplet life-cycle just described for the case of hydrodynamic interruption requires splitting to be balanced by forward Ostwald ripening, which sustains a stationary droplet number. In contrast, we find that for \( \zeta < 0 \) only bubbles for \( a < 0 \), with \( \kappa > 0 \) for stability [18, 22].

The terms in \( \zeta \) and \( \lambda \) in (2) break time-reversal symmetry at \( O(\phi^2, \nabla^4) \) in (1). These terms also break the \( \phi \rightarrow -\phi \) symmetry of the passive limit; however the full system of equations remains invariant under \( (\phi, \lambda, \zeta) \rightarrow (\phi, \lambda, -\zeta, -\phi) \). This means that all statements made below about droplets also apply to the phase-inverted case of bubbles, so long as \( \lambda \) and \( \zeta \) are also changed in sign. Notably, the reverse Ostwald process stabilizes only droplets for \( \zeta < 0 \) and only bubbles for \( \zeta > 0 \) [17].

The fluid flow, in the limit of low Reynolds number (as applicable to microswimmers), is obtained from the solution of the Stokes equation: \( \nabla \cdot \sigma = -f \), where \( f = \nabla \cdot (\Sigma^A + \Sigma^E) \) is the force density on the fluid, \( \sigma = -pI + \eta(\nabla v + (\nabla v)^T) \) is the Cauchy fluid stress, \( \eta \) is viscosity, \( I \) is the identity tensor, and \( p \) is the pressure field which contains all isotropic terms and ensures incompressibility \( (\nabla \cdot v = 0) [23] \). We neglect noise in these flow equations since this would involve the thermal temperature \( T \) which is vastly smaller than \( D \) for active swimmers.

There exists a standard procedure [24] to derive the deviatoric stress \( \Sigma^E \) in equilibrium systems using the free energy \( F \). This stress, retaining all isotropic terms, satisfies \( \nabla \cdot \Sigma^E = -\partial \Sigma^E/\partial E \), which is the thermodynamic force density on the fluid due to gradients of the concentration field \( \phi [18] \). The deviatoric stresses \( \Sigma^E \) and \( \Sigma^A \) are then, in \( d \)-dimensions, given to the required order as

\[
\Sigma^E = -\kappa S, \quad \Sigma^A = -(\kappa - \kappa)S, \quad (3)
\]

where \( S \equiv (\nabla \phi)(\nabla \phi) - \frac{\lambda}{2} \nabla \phi^2 I \). The mechanical stress \( \Sigma^A + \Sigma^E = -\kappa S \) is not derived from a free energy and breaks detailed balance in general. Its overall coefficient \( \kappa \) can be either positive (for extensile microswimmers) or negative (for sufficiently contractile ones) [19], unlike equilibrium systems where \( \Sigma^A = 0 \).

Equations (1-3) define our Active Model H for the diffusive dynamics of a conserved order parameter with momentum conservation. The numerical method we use to integrate these equations is described in [20]. Having neglected inertia, they reduce to an effective dynamics for \( \phi \) alone, which reads (with \( M = 1 \))

\[
\dot{\phi} = \nabla^2 \mu^\text{eff} - \nabla \phi(r) \cdot \int G(r-r') \cdot f(r') \, dr' - \nabla \cdot \nabla \mu^\Lambda. \quad (4)
\]

Here \( G(r) \) is the Oseen tensor, \( \mu^\Lambda = \sqrt{2DA} \), and \( \mu^\text{eff} = \mu^c + \mu^\Lambda + \mu^\kappa \) an effective chemical potential. This has a part \( \mu^\Lambda \), constructed via Helmholtz decomposition of the active current term in (2), as \( \zeta(\nabla^2 \phi) \nabla \phi = -\nabla \mu^c + \nabla \times \mu^\kappa + \nabla \times A \). The \( \nabla \times A \) term is divergenceless, and so cannot contribute to \( \phi \) in (4), allowing it to be ignored [17].

Three tensions: At large scales, the dynamics of an interface between phases is controlled by its curvature and its interfacial tension. Without activity, there is only one such tension, \( \gamma_0 = \sqrt{-8\kappa/9b^2} \), governing both diffusive and mechanical sectors; curvature then drives diffusive currents and/or fluid flow via Laplace pressure [15, 18].

With activity, two further tensions enter [17, 19]. First, the chemical potential term \( \mu^c \) is in general nonlocal, and acquires a step-discontinuity across a curved interface that is cancelled by a counter-step in \( \mu^E \). The result is a discontinuity in \( \phi \) different from the equilibrium one. This is captured by a ‘pseudotension’ \( \gamma_\phi \) that replaces \( \gamma_0 \) when calculating diffusive fluxes between droplets, and becomes negative for sufficiently negative \( \zeta, \lambda \). Crucially, negative \( \gamma_\phi \) does not make interfaces locally unstable; spherical droplets stay spherical. Rather, its effect is to drive the system towards states of globally uniform curvature (monodisperse drops) whereas positive tension promotes curvature differences by shrinking small droplets and growing large ones (the Ostwald process) [17].
The third tension arises from the active stress at interfaces, where swimmers align parallel or antiparallel to the surface normal. In either case, contractile swimmers pull fluid inward along the normal direction and expel it in the interfacial plane, causing stretching, whereas extensile swimmers do the opposite. The resulting mechanical tension is \(\gamma_s\), which is negative for sufficient contractility; this is known to interrupt phase separation by destabilizing interfaces \([19]\). We find next that it also does so for droplets, by destabilizing interfaces locally (in contrast with negative \(\gamma_s\); see above).

**Self-shearing instability:** For simplicity we first consider active mechanical stress alone, setting \(\lambda = \zeta = 0\) so that \(\gamma_o = \kappa\gamma_0 / \kappa\), which is negative for sufficient contractility; this is known to interrupt phase separation for bicontinuous regimes \([19]\). We find next that it also does so for droplets, by destabilizing interfaces locally (in contrast with negative \(\gamma_o\); see above).

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**Scaling of droplet size:** In Fig.1C, we show that the droplet size obeys \(\bar{R} \sim |\gamma_v/\gamma_0|^{-0.52}\) (best fit exponent), where, in these simulations, \(\gamma_0 = \gamma_o > 0\). We now argue on simple grounds for a negative one half exponent whenever \(\gamma_v < 0\) and \(\gamma_0 > 0\). From the mechanical tension \(\gamma_v\) and fluid parameters we can construct just one quantity with the dimensions of velocity: \(\gamma_v = \gamma_v / \eta\). This is the familiar coarsening rate \(\dot{L}\) for systems with bicontinuous domains of size \(L\), in the so-called ‘viscous hydrodynamic’ regime where curvature drives fluid motion and diffusive fluxes are negligible \([24]\). For a droplet of size \(\bar{R}\) with negative \(\gamma_v\), one thus expects the time between scission events to scale as \(\tau = -\dot{R}/\gamma_v\). The Ostwald process gives another speed, which is the rate of change of the mean droplet size \(V_o(\bar{R}) = -\dot{\bar{R}} \propto M \gamma_o / \phi^2 B \bar{R}^2\), where \(\phi_B\) is the binodal density \([17, 18]\). Balancing a positive Ostwald speed \(V_o(\bar{R})\) with a negative hydrodynamic speed \(\gamma_v\) gives the promised scaling law

\[
\bar{R} \sim \left(\frac{\gamma_v \phi^2 B}{\eta M \gamma_o}\right)^{-1/2} \sim |\gamma_v/\gamma_o|^{-1/2}.
\]

The predicted one-half exponent is in excellent agreement with Fig.1C. Unlike the simulations, our scaling argument does not include noise, which appears inessential.
to the steady state, at least in the parameter range investigated here. We checked this by explicitly switching off noise once the steady state is achieved and found that it continues to exist, with essentially the same droplet life-cycle and mean size. While noise is needed initially to start the phase separation, once enough droplets are present the contractile active stress can maintain the dynamics of scission and coarsening indefinitely. In addition to splitting and ripening, we observe splitting-induced coalescence mediated by fluid flow. This resembles the coalescence-induced coalescence mechanism seen for semidilute passive droplets in two dimensions [25]. However, this cannot change the scaling because its physics is also governed by $\gamma_\phi$.

**Competing activity channels:** We next consider the full dynamics of (4), where both the active tensions can change sign, and enumerate the possible steady-states. The resulting phase diagram, in the $(\gamma_\phi, \gamma_v)$ plane, is shown in Fig.(3A-B). In panel A, we show time evolution of two bubbles of unequal sizes and their final steady state. The two bubbles disproportionate if both tensions are positive; the corresponding outcome in the many-state. The two bubbles disproportionate if both tensions are positive; the corresponding outcome in the many-state. The three activity parameters $\kappa, \lambda, \zeta$ in this region serve merely to renormalize the passive behavior. In contrast, when $\gamma_v > 0$ and $\gamma_\phi < 0$, we recover the result of [17], with reverse Ostwald ripening arresting phase separation of droplets (see panel C(ii)). As in the forward case, the reverse Ostwald regime entails little or no fluid motion, so the results given in [17] apply even for the wet systems studied here, with physics controlled by $\gamma_\phi$ and no role for $\gamma_v$. Thirdly, the case $\gamma_v < 0$ and $\gamma_\phi > 0$ is governed by our interrupted steady state with the splitting/coarsening life-cycle described above; here $\lambda$ and $\zeta$ enter only via $\gamma_\phi$. Because the steady state balances mechanical and diffusive processes, both tensions enter the mean droplet size via (5).

In the final regime, both tensions are negative. Here, because negative $\gamma_\phi$ reverses Ostwald ripening, the balance of splitting and ripening embodied in (5) cannot be maintained. Were splitting to continue, the number of droplets would increase forever, with the reverse Ostwald process driving these towards uniformity in size but not allowing the number to reduce by evaporation. Accordingly, for negative $\gamma_\phi$, no matter how small its magnitude, the steady state *must* consist of almost static droplets with no splitting, and this is indeed what we observe.

Despite this complete change of mechanism, we now argue that the steady-state droplet size is still governed by $\bar{R} \sim |\gamma_v|^{-1/2}$ as in (5). This is because the reverse Ostwald process drives the system towards uniformity of curvature which, for a single droplet, directly opposes the self-shearing instability. Indeed, for an amplitude $A \sim \epsilon R$ of the lowest deformation mode in a droplet of radius $R$, one expects on dimensional grounds that, up to prefactors, $A \sim \epsilon (\gamma_v + \gamma_\phi(R))$. Here the first term is the self-shearing instability and the second is stabilizing for negative $\gamma_\phi$. Stability is restored for droplets smaller than $\bar{R}$ given by (5), which thus again gives the scaling of steady-state droplet size, albeit by a completely different mechanism of arrest rather than interruption. This scaling is consistent with simulations, see Fig.(3D).

All the above arguments apply equally to bubbles as to droplets via invariance under $(\phi, \lambda, \zeta) \rightarrow -(\phi, \lambda, \zeta)$.

**Conclusion:** In wet active systems containing droplets or bubbles, microphase separation can replace bulk phase separation by three distinct mechanisms. These are reverse Ostwald ripening (also present in the dry limit, and caused by negative pseudotension $\gamma_\phi$); hydrodynamic interruption by self-shearing (caused by negative mechanical tension $\gamma_v$); and a peculiar combination of the two when both of tensions are negative. In contrast to reverse Ostwald ripening, where the size of droplets or bubbles in the arrested steady state is selected by noise (or, in its absence, initial conditions) [17], our self-shearing mechanism continues to operate even if the noise is switched off after the steady-state is reached. The droplet size is then selected by a balance between droplet splitting and forward Ostwald ripening. This balance is unsustainable when $\gamma_\phi$ is also negative, and so is replaced by a new one whereby the reverse Ostwald process suppresses the self-shearing instability for small droplets, leading to static arrest rather than dynamic interruption.

These results point to a subtle interplay of causes behind the phenomena of active microphase separation. We
content that they cannot be understood without first clearly enumerating the relevant activity channels, and then studying their interaction; this is best done within the minimalist framework of an active field theory as done here. A possible generalization of (4) would allow for boundary conditions imposed on the fluid flow via a nearby wall; these are known to play a crucial role in active phase separation [26–28] but with no clear understanding, so far, of the microphase-separated case.

Our findings should complement more detailed work that could connect our activity channels to microscopic interactions [6–9]. They may also be relevant to continuum models of multiple species (some non-conserved) that were used recently to study microphase separations used by cells to create cytoplasmic and nucleoplasmic organization [29, 30]. Fluid motion is often neglected in such studies but we have shown that it brings new features, suggesting exciting directions for future work.

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SUPPLEMENTAL INFORMATION (SI)

Appendix A: Methodology

In this section, we explain the numerical methods used to study the field theory of an active scalar field with mass and momentum conservation. We first explain the solution of the flow equations and then given details of the simulation and provide the system of parameters.

1. Stokes solver

At low Reynolds number, the fluid flow satisfies the Stokes equation

\[-\nabla p + \eta \nabla^2 v = -f, \quad \nabla \cdot v = 0.\]  

(A1)

(A2)

Here,

\[f = \nabla \cdot (\Sigma^A + \Sigma^E),\]

is the force density on the fluid. We now show how to determine flow given the force density and detail the implementation of incompressibility in the flow. We use Fourier transforms to obtain a solution of the above equations. We define the Fourier transform of a function \(\varphi(r)\)

as

\[\hat{\varphi}(k) = \mathcal{F}[\varphi(r)] = \int \varphi(r) e^{-ik \cdot r} \, dr, \quad (A3a)\]

\[\varphi(r) = \mathcal{F}^{-1}[\hat{\varphi}(k)] = \frac{1}{(2\pi)^d} \int \hat{\varphi}(k) e^{ik \cdot r} \, dk. \quad (A3b)\]

We now Fourier transform (A1) to obtain

\[-ik \hat{p} - \eta k^2 \hat{v} = -\hat{f}, \quad (A4a)\]

\[ik \cdot \hat{v} = 0. \quad (A4b)\]

The above equations can then be used to obtain the Fourier transform of the pressure field

\[\hat{p} = -ik \cdot \hat{f} / k^2. \quad (A5)\]

The above expression of the pressure is then used in (A4a) to obtain the solution of the fluid flow, with built-in incompressibility, given as

\[\hat{v} = \hat{G}(k) \cdot \hat{f}, \quad \hat{G}(k) = \frac{1}{\eta} \left( \frac{I}{k^2} - \frac{kk}{k^4} \right). \quad (A6)\]

Here \(\hat{G}(k)\) is the Fourier transform of the Oseen tensor \(G(r)\) [32]. The explicit forms of the Oseen tensor, in three dimensions, is

\[G(r) = \frac{1}{8\pi \eta} \left( \frac{I}{r} + \frac{rr}{r^3} \right). \quad (A7)\]

The above is the Green’s function of Stokes equation in an unbounded domain and captures the long range interactions at low Reynolds number [35]. It should be noted that we are confining ourselves to low Reynolds number by not solving the full Navier-Stokes equation. Thus, our theory is mainly applicable to active matter systems of microorganisms [34] and synthetic microwimmers [1], where the Reynolds number is known to be very small.

2. Simulation details

The simulations reported in this manuscript are performed by the numerical integration of Eq.(1-3) of the main text. Eq.(A6) is the solution for the fluid flow, which we use for numerical computations. The solution, by construction, satisfies incompressibility exactly. The Fourier solution of the fluid flow is then used in Eq.(1) to numerical update the order parameter field. The terms in the mass conservations equations are computed using a pseudospectral method (involving Fourier transforms and 2/3 dealiasing procedure) [35, 36]. The spectral solver for the fluid flow and order parameter is numerically implemented using standard fast Fourier transforms (FFTs) and by virtue of these Fourier transforms, periodic boundary conditions are automatically ensured in the system. We evolve the system in time using the explicit Euler–Maruyama method [37]. We provide the parameters used in generating all the figures of the manuscript in Table I.
Table I. Simulation parameters used to study the field theory of an active scalar with mass and momentum conservation. Throughout the paper, the following parameters have been kept fixed \( a = -0.25 \), \( b = 0.25 \), \( \kappa = 1 \), \( M = 1 \), and \( D = 0.02 \). The value of \( \tilde{\kappa} \) has been varied in Fig.(1C) and Fig.(3D) to show the scaling of the length scale with contractile activity. We start with an elliptic droplet in 2A and 2B. The parameters for activity in mass conservation (\( \lambda \), \( \zeta \)) and momentum conservation (\( \tilde{\kappa} \)) equations has been varied in Fig.(3A-B) to enumerate distinct steady states. All the simulations reported are in two space dimensions, while the predictions of the manuscript would be maintained in higher dimensions.

### Appendix B: Supplemental movies

In this Appendix, we describe the supplemental movies which are complementary with the main text. These movies are available online at [this URL](#).

**Movie I:** The movie shows the dynamics of Fig.1A in the main text, which is the self-shearing instability in the bubble phase for contractile active stress (\( \tilde{\kappa} < 0 \)) with no activity in the diffusive sector. Starting from a uniform phase, droplets nucleate due to the noise and they grow in size until interrupted by the contractile active stress. The parameters are \( a = -0.25 \), \( b = 0.25 \), \( \kappa = 1 \), \( \tilde{\kappa} = -0.1 \), \( \lambda = 0 \), and \( \zeta = 0 \). The initial global density \( \phi_0 = 0.6 \) and the system size is \( 256 \times 256 \).

**Movie II:** Same as in movie I but with \( \phi_0 = -0.4 \). It corresponds to Fig.(1B) of the main text.

**Movie III:** The movie shows the fluid flow and dynamics of an initial elliptical interface, as shown in Fig(2A) of the main text, for an extensile active stress (\( \tilde{\kappa} > 0 \)). It is shown that an extensile flow makes the interface circular, and thus, stabilizes it. The parameters are \( a = -0.25 \), \( b = 0.25 \), \( \kappa = 1 \), \( \tilde{\kappa} = 1.1 \), \( \lambda = 0 \), \( \zeta = 0 \). The system size used for this simulation is \( 256 \times 256 \).

**Movie IV:** Same as in movie III, but with \( \tilde{\kappa} = -0.1 \). It corresponds to Fig(2B) of the main text.

**Movie V:** Nucleation and growth of bubbles when activity is present in both the mechanical and diffusive sectors. A snapshot from the steady state is in Fig(3C-(ii)) of the main text. The parameters are \( a = -0.25 \), \( b = 0.25 \), \( \kappa = 1 \), \( \tilde{\kappa} = 1.2 \), \( \lambda = 1 \), \( \zeta = 2 \). The initial global density \( \phi_0 = 0.6 \) and the system size is \( 128 \times 128 \).

**Movie VI:** Same as in movie V but with \( \tilde{\kappa} = -0.1 \). It corresponds to Fig.3D in the main text. The fluid due to contractile stress has the effect of stretching the interface, which is the precursor to the self-shearing instability as the system approaches the steady state. It should be noted that there is no self-shearing in the steady state - this is because the reverse Ostwald process drives the system towards uniformity of curvature which directly opposes the self-shearing instability.