

Nonreciprocal Interactions between Condensates in Chemically Active Mixtures

Jacopo Romano^{1,2}, Martin Kjøllesdal Johnsrud¹, Benoît Mahault¹, and Ramin Golestanian^{1,3}

¹Max Planck Institute for Dynamics and Self-Organization (MPI-DS), 37077 Göttingen, Germany

²SISSA—International School for Advanced Studies and INFN, via Bonomea 265, 34136 Trieste, Italy

³Rudolf Peierls Centre for Theoretical Physics, University of Oxford, Oxford OX1 3PU, United Kingdom

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We study the behavior of catalytically active droplets in multicomponent conserved mixtures affected by noise. Working in the thin interface limit, we analytically determine the state diagram of the system, characterized by multiple dynamical regimes, and verify our findings using numerical simulations. In particular, we show the emergence of a nonreciprocal, chemically mediated interaction between the droplets, which leads to the formation of (meta)stable clusters of droplets of different species. We find that the clusters can display self-propulsion in a large part of the parameter space, including regions where the nonreciprocal interactions between the droplets are purely attractive. This surprising feature arises from the nonlocal nature of the chemical interactions, and points to locality violations as a general mechanism for energy dissipation and emergence of out-of-equilibrium steady states in active matter.

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The distinguishing feature of an active system is the continuous energy consumption of its microscopic constituents, which drives the system out of equilibrium leading to complex and novel interactions. In multiple experimental systems—both biological and artificial—such interactions are due to the production and consumption of chemicals, which in turn affect the dynamics of the species constituting the system [1–5]. Examples include large collections of catalytically active colloids [1,6–8], bacterial colonies and collections of cells [9–13], as well as metabolically active condensates [14–20], and suspensions of enzymes [21–23]. Chemical activity of enzymes manifests in a variety of nonequilibrium behavior including growth and division of droplets [24–29]. Such mixtures are naturally present in the cytoplasm of cells and it is believed that their activity is crucial in allowing them to perform their biological functions [14,30].

When multiple chemically active species are present in a mixture, the effective interactions mediated by the chemical often break action-reaction symmetry [31–36]. This has been studied in droplets [37], mixed bacterial colonies [38–40], and catalytic colloids [41–44], showing the formation of self-propelled clusters. The formation of multicomponent, multiphase mixtures has been observed also *in vivo* in embryo cells [45]. The physical mechanisms

describing how the chemicals affect the species are not fully understood, and are likely system-specific, especially for phase separating biopolymers [27]. It has been argued that the enzymes in these mixtures display an effective chemotactic behavior [46], leading to a mass current of the

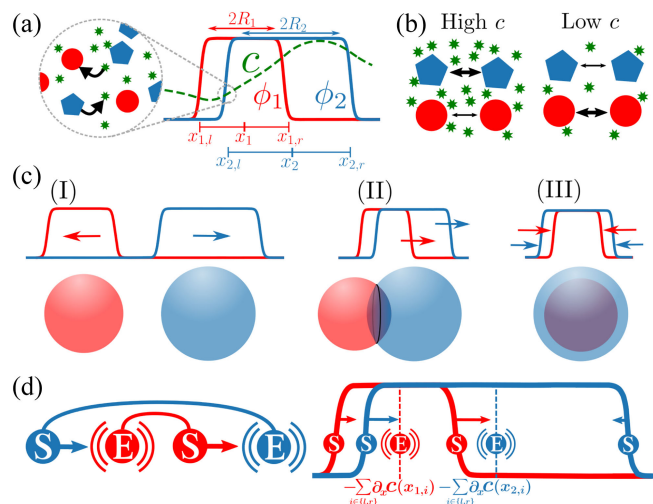


FIG. 1. (a) The species gather into droplets due to their interaction. Spatial variations in the densities of the species (ϕ_1 and ϕ_2) create a spatially varying chemical field (c , in green) as they produce and consume the chemical. (b) The chemical affects the intraspecies interaction and mediates effective inter-species interactions. (c) Configurations of droplets: (I) disjoint, (II) partially overlapping, and (III) fully overlapping. (d) Attractive interactions give rise to chasing behavior. The center of the droplet is an emitter (E) of a chemical signal and the interfaces are sensors (S). An example where purely attractive interactions lead to chasing (left) and its implementation with two droplets (right).

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species proportional to the chemical gradients [19,47–50]. Moreover, it has been shown in multiple experiments on phase separating droplets [15–17,51–53] that the chemicals affect the species by altering the interspecies interactions, which determine the phase separation, effectively changing the critical point of the mixture. These observations cannot be fully accounted for by chemotactic currents, and suggest that the free energy of the mixture depends on the local chemical concentrations, as illustrated in Figs. 1(a) and 1(b).

In this Letter, we explore the dynamics of a multi-component mixture in the presence of a chemical that affects its material properties and is in turn produced and consumed by the components of the mixture. We elucidate how such dependencies lead the individual droplets to respond to chemical gradients, reminiscent of phoretic motion. We then demonstrate how this property gives rise to effective nonreciprocal interactions between droplets in a way that can be controlled via material parameters and geometric characteristics. We find the emergence of (meta)stable clusters of droplets of different species, displaying different mechanisms of self-propulsion. The first mechanism is chasing of droplets due to nonreciprocal, attractive-repulsive interactions, similarly to what is observed in [31,38,40,42]. The second mechanism is nonlocality-induced chasing of mutually attractive droplets, defying the expectation that self-propulsion requires attractive-repulsive interactions. Finally, the third mechanism is due to the interplay between nonreciprocal couplings and fluctuations in deterministically static configurations.

Theoretical framework—To describe the system's dynamics, we model the evolution of N density fields for $\phi_a(\mathbf{r}, t)$, $a \in \{1, 2, \dots, N\}$, which are governed by a free energy $\mathcal{F}[\{\phi_a\}, c]$ that depends on a chemical field c that the species produce or consume, e.g., through dependence of the material properties on the chemical. We neglect advective transport, which is appropriate for low-dimensional or sufficiently small systems [54,55] and qualitatively justified in regimes exhibiting ripening phenomenology [53,56,57]. We thus consider a generalization of model B in d dimensions [58] as follows:

$$\partial_t \phi_a = \nabla \cdot \left[M_a \nabla \frac{\delta \mathcal{F}[\{\phi_b\}, c]}{\delta \phi_a} \right] + \sqrt{2M_a T} \nabla \cdot \mathbf{\Lambda}_a, \quad (1a)$$

$$-D_c \nabla^2 c + \gamma c = \sum_a r_a \phi_a, \quad (1b)$$

where M_a 's are the corresponding mobilities, $\mathbf{\Lambda}_a$'s are Gaussian stochastic currents with zero mean and correlations $\langle \Lambda_{a,i}(\mathbf{r}, t) \Lambda_{b,j}(\mathbf{r}', t') \rangle = \delta_{ij} \delta_{ab} \delta^d(\mathbf{r} - \mathbf{r}') \delta(t - t')$ [59], and T is the temperature. In Eq. (1b), r_a 's are the production (consumption) rates of the chemical by the two species, D_c is the diffusion coefficient of the chemical, and $\gamma \equiv D_c \kappa^2$ is its decay rate, which introduces a decay length scale κ^{-1} .

The dynamics of c is assumed faster than that of the two species; in this limit, temporal evolution and fluctuations of c can be neglected [13,60], leading to (1b). For simplicity, we also assume that the free energy can be decomposed into independent parts: $\mathcal{F}[\{\phi_b\}, c] = \sum_a \mathcal{F}_a[\phi_a, c]$. In Appendix A, we show in that our results are not fundamentally modified by the presence of (weak) interspecies interactions. We then consider: $\mathcal{F}_a = \int d^d \mathbf{r} [\frac{1}{2} K_a(\phi_a, c) (\nabla \phi_a)^2 + U_a(\phi_a, c)]$, where the U_a terms are double-well potentials (see Appendix B) promoting phase separation in both species leading to the formation of droplets of the dense phase coexisting with a dilute background.

In the sharp interface limit [61], the densities in and out of the droplets are given by the two minima of the field potentials $U_a(\phi_a)$, which we denote by $\bar{\phi}_{a,+}$ for the dense phases and $\bar{\phi}_{a,-}$ for the dilute phases, leading to density contrasts of $\Delta \bar{\phi}_a = \bar{\phi}_{a,+} - \bar{\phi}_{a,-}$ across the droplet interfaces. The dynamics of the droplets is then controlled by c -dependent surface tensions $\sigma_a(c)$, which can be calculated in terms of U_a and K_a , as well as $\bar{\phi}_{a,\pm}(c)$ [54,62]. We consider the linear response regime, in which $\sigma_a(c) = \sigma_a(0) + (\partial \sigma_a / \partial c) c$ with the derivative evaluated at vanishing concentration, and neglect the c dependence of $\bar{\phi}_{a,\pm}$ (see Supplemental Material [55]).

Our Letter reveals that the coupling constant,

$$C_{a \rightarrow b} \equiv -r_b \frac{M_a}{\Delta \bar{\phi}_a^2} \frac{\partial \sigma_a}{\partial c}, \quad (2)$$

specifies the strength of the interaction experienced by a toward b , with the convention that for positive $C_{a \rightarrow b}$ species a is attracted to species b . This is because a droplet of species a climbs (or descends) chemical gradients at a rate proportional to $-(M_a / \Delta \bar{\phi}_a^2) (\partial \sigma_a / \partial c)$, while it senses a chemical gradient proportional to the production rate r_b of the other droplet [see Eq. (1b)]. As in general we have $C_{a \rightarrow b} \neq C_{b \rightarrow a}$, the interactions are nonreciprocal, similar to what is observed in phoretic active systems [31,42]. These droplets, however, present remarkable additional features in comparison with phoretic colloids, as their effective nonreciprocal interactions can be controlled by tuning their geometric features and the screening length, which is afforded by the emergent nontrivial size dependence of the effective mobilities (see below). Moreover, the droplets can penetrate each other and lead to a richer set of configurations for active complexes that form due to nonreciprocal interactions (see Fig. 1). We note that the nonreciprocity, which manifests itself both in strength and sense of the interactions, cannot be simply represented by an antisymmetric matrix of coupling constants due to the nontrivial dependence on the droplet sizes and screening length.

To elucidate this phenomenology, we focus on the long-time behavior of the system, which is characterized by the presence of spherical droplets of radii R_a (one for each

species), with volume $\mathcal{V}_a = V_d R_a^d$ and surface area $S_a = dV_d R_a^{d-1} = d\mathcal{V}_a/R_a$, where $V_d = \pi^{d/2}/\Gamma(1+d/2)$. We can use the dynamical equation for the concentration fields (1) to extract effective stochastic equations of motion for the center of mass of the droplet of species a , which we denote by $\mathbf{x}_a$ (see Fig. 1), as follows [54]:

$$\dot{\mathbf{x}}_a = -\mathcal{M}_a S_a \frac{\partial \sigma_a}{\partial c} \nabla \bar{c}_{s,a}(\mathbf{x}_a) + \sqrt{2\mathcal{M}_a T} \boldsymbol{\eta}_a(t), \quad (3)$$

where $\bar{c}_{s,a}(\mathbf{x}_a) \equiv \langle c \rangle_{S_a} = (1/(dV_d)) \int d\hat{\mathbf{n}}_a c(\mathbf{x}_a + R_a \hat{\mathbf{n}}_a)$ with $\hat{\mathbf{n}}_a$ representing the radial unit vector describing the points on the boundary S_a of droplet a , $\mathcal{M}_a = M_a d/(\mathcal{V}_a \Delta \bar{\phi}_a^2)$ is an effective mobility coefficient, and $\boldsymbol{\eta}_a$'s are Gaussian white noise terms with zero mean and correlations $\langle \eta_{a,i}(t) \eta_{b,j}(t') \rangle = \delta_{ij} \delta_{ab} \delta(t-t')$. We note that Eq. (3) is independent of the details of the free energy, and can be regarded as a generalized (diffusio)phoretic transport mechanism resulting from interfacial stresses [63]. Note that Eq. (3) describes the dynamics of droplets even when they overlap. Within the thin interface approximation, it is possible to derive exact expressions for $\bar{c}_{s,a}(\mathbf{x}_a)$ in different dimensions (see Appendix C).

When the droplets do not overlap, we obtain a closed-form expression for the surface-averaged concentration and use it to express the drift velocity as follows:

$$\dot{\mathbf{x}}_a = -\frac{d^2 M_a}{\Delta \bar{\phi}_a^2} \frac{\partial \sigma_a}{\partial c} \frac{\mathcal{I}_d(\kappa R_a)}{R_a} \nabla c(\mathbf{x}_a) + \sqrt{2\mathcal{M}_a T} \boldsymbol{\eta}_a(t), \quad (4)$$

where $\mathcal{I}_d(b)$ is defined in Appendix C, which details the derivation. To demonstrate the emergence of nonreciprocal droplet interactions and the resulting phenomenology, we focus on the two-body dynamics of two droplets of different species. We define the relative position of the two droplets as $\mathbf{x} = \mathbf{x}_2 - \mathbf{x}_1$ and their midpoint as $\mathbf{X} = \frac{1}{2}(\mathbf{x}_1 + \mathbf{x}_2)$. The dynamics of these coordinates will take the general form

$$\dot{\mathbf{x}} = -\mathcal{M} \nabla_{\mathbf{x}} W_{-}(\mathbf{x}) + \sqrt{2\mathcal{M}T} \boldsymbol{\eta}_x(t), \quad (5a)$$

$$\dot{\mathbf{X}} = \mathbf{v}(\mathbf{x}) + \sqrt{\mathcal{M}T/2} \boldsymbol{\eta}_X(t), \quad (5b)$$

where $\boldsymbol{\eta}_x(t)$ and $\boldsymbol{\eta}_X(t)$ are two independent Gaussian white noises with zero mean and unit variance, $\mathcal{M} = \mathcal{M}_1 + \mathcal{M}_2$, and $\mathbf{v}(\mathbf{x}) = -\frac{1}{2} \mathcal{M} \nabla_{\mathbf{x}} W_{+}(\mathbf{x})$, with the potentials $W_{\pm}(\mathbf{x}) = (\mathcal{M}_2/\mathcal{M}) S_2 (\partial \sigma_2 / \partial c) \bar{c}_{s,2}(\mathbf{x}) \pm (\mathcal{M}_1/\mathcal{M}) \times S_1 (\partial \sigma_1 / \partial c) \bar{c}_{s,1}(\mathbf{x})$ depending only on $\mathbf{x}$ due to translational invariance of the system. We start with $d = 1$, where a droplet of species a is characterized by the positions $x_{a,l} = x_a - R_a$ and $x_{a,r} = x_a + R_a$ of its left and right interfaces, respectively. Consequently, we can obtain a simple expression for the surface-averaged concentration as $\bar{c}_{s,a}(x_a) = [c(x_a - R_a) + c(x_a + R_a)]/2$. Note, however, that this

choice is made for simplicity of the presentation, and that analytical results can be obtained in all dimensions, as shown in Appendix C. From here onward, we set $\bar{\phi}_{1,+} = \bar{\phi}_{2,+} = 1$, $\bar{\phi}_{1,-} = \bar{\phi}_{2,-} = 0$, $D_c = 1$, and $\gamma = 1$, and hence express all length scales in terms of the decay length of the chemical $\kappa^{-1} = \sqrt{D_c/\gamma} = 1$ [55]. Moreover, we assume $R_2 > R_1$ without loss of generality. To close the equations, we need to solve for the concentration field using Eq. (1b), which yields $\partial_x c(x) = \frac{1}{2} \sum_a r_a (e^{-|x-x_a+R_a|} - e^{-|x-x_a-R_a|})$.

Using this expression, we obtain explicit expressions in the form of Eq. (5), with the potentials $W_{\pm}(\mathbf{x})$ given in Appendix D. As anticipated, the expressions for $W_{\pm}(\mathbf{x})$ depend on the material parameters only through the coupling constants $C_{a \rightarrow b}$, highlighting the nonreciprocal nature of the interactions.

Deterministic dynamics—We can now study the behavior of the system as a function of chasing rates [as defined in Eq. (2)] and droplet sizes. In Fig. 2(a), we present a classification of the different states of the dynamics by using the dimensionless parameters $\mathcal{R} \equiv R_1/R_2$ as radius and $\vartheta \equiv \arg(C_{1 \rightarrow 2} + iC_{2 \rightarrow 1})$ as angle (see Appendix B for the details of the rules used in the classification).

Starting our analysis with no noise, we note that Eq. (5) always admits $x = 0$ as a stationary point, corresponding to the symmetric configuration with the small droplet encapsulated inside the large one, i.e., configuration (III) of Fig. 1(c). This fixed point is stable when

$$C_{2 \rightarrow 1} < \frac{R_2}{R_1 \tanh R_1} C_{1 \rightarrow 2}, \quad (6)$$

which means that if Eq. (6) is satisfied, droplets will reach a symmetric configuration of complete engulfment when starting from asymmetric initial conditions

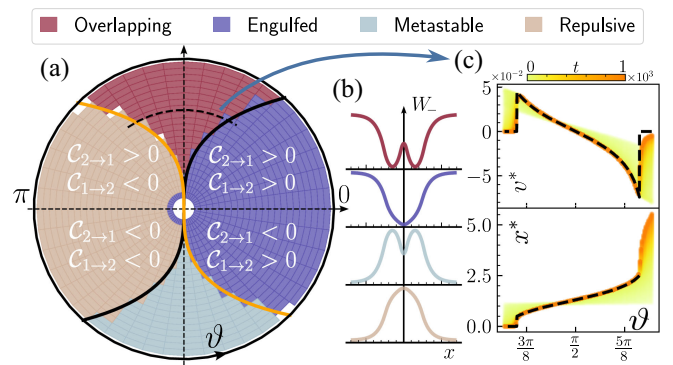


FIG. 2. (a) State diagram from simulations of Eq. (1) presented as a radial plot with radius $\mathcal{R} = R_1/R_2$ and angle $\vartheta = \arg(C_{1 \rightarrow 2} + iC_{2 \rightarrow 1})$. The black and orange lines mark the boundaries of Eqs. (6) and (7), respectively. (b) Effective potentials for $W_{-}(\mathbf{x})$ in the various states. (c) Steady-state velocity and relative position calculated along the dashed line in (a) (dashed black), compared with direct simulations. Color indicates time, with the steady-state shown in orange.

[see Fig. 2(a), black line]. For sufficiently large initial distances [droplets in configuration (I) of Fig. 1(c)], the potential decreases with $|x|$ when

$$C_{2 \rightarrow 1} < -\frac{R_2 \tanh R_2}{R_1 \tanh R_1} C_{1 \rightarrow 2}. \quad (7)$$

In this case, two well-separated droplets experience an effective repulsion (and their distance increases indefinitely). Note that Eqs. (6) and (7), which can be satisfied at the same time, become increasingly symmetric around the $C_{1 \rightarrow 2} = 0$ line [vertical axis in Fig. 2(a)] for large values of R_1 and R_2 . When they are both satisfied, $W_-(x)$ has a metastable minimum at $x = 0$, while disjoint droplets repel each other [see Fig. 2(b)]. Finally, when both Eqs. (6) and (7) are violated, $W_-(x)$ becomes a double-well potential with two minima [see Fig. 2(b)], representing configuration (II) of Fig. 1(c). In this case, the fixed point at $x = 0$ is unstable, while two stable fixed points appear at $\pm x^*$, where $x^* = \frac{1}{2} \ln \{1 + (e^{2R_2} - e^{2R_1})[(C_{2 \rightarrow 1}R_1 - C_{1 \rightarrow 2}R_2)/(C_{2 \rightarrow 1}R_1 + C_{1 \rightarrow 2}R_2)]\}$. This corresponds to the partially overlapping configuration (II) of Fig. 1(c), with the smaller droplet being on the left of the larger one for $x > 0$. The steady-state velocity $v^* = v(x^*)$ of the midpoint of the cluster is then found $v^* = [(\sinh(R_2 - R_1)C_{1 \rightarrow 2}C_{2 \rightarrow 1})/(C_{2 \rightarrow 1}R_1 + C_{1 \rightarrow 2}R_2)]e^{-x^*}$ [see Eq. (5b)]. These predictions are verified by simulating Eq. (1) at negligible noise ($T = 10^{-5}$) to check if the final state is in a disjointed, overlapping, or engulfed configuration [respectively, in configurations (I), (II), or (III) of Fig. 1(c)], and it is indeed observed that the shape of the effective potential W_- determines the behavior; see Figs. 2(a)–2(c).

It is remarkable that self-propelling clusters are observed in the region of fully attractive, albeit nonreciprocal, droplet interactions where $C_{1 \rightarrow 2} > 0$ and $C_{2 \rightarrow 1} > 0$. This may be surprising, as self-propulsion in systems with short-range interactions emerges when the chasing rates have opposite signs. However, since droplets are extended objects, additional opportunities to realize conditions for effective nonreciprocal interactions arise from the fact that the sources and sinks of chemicals are in the bulks of the droplets while the force-sensing operates at their interfaces. The chasing rates determine how the interface of each droplet interacts with the center of the other. This implies that configurations in which the leftmost interface of droplet 2 lies between the leftmost interface and center of mass of droplet 1 can lead to a net repulsive effect on droplet 1, despite all the forces being attractive from the point of view of the respective interfaces; see Fig. 1(d). Note that this effect is only possible in active systems.

As a consequence of the change in self-propulsion mechanism when $C_{1 \rightarrow 2}$ changes sign [corresponding to $\vartheta = (\pi/2)$] the cluster inverts its velocity. While for $C_{1 \rightarrow 2} < 0$ droplet 1 on the left is repelled by droplet 2 that is chasing it, for $C_{1 \rightarrow 2} > 0$ droplet 1 chases droplet 2 due to

the effective nonreciprocity described above. This is shown in Fig. 2(c), where we present the velocities and relative positions for a set of simulations, within, and just outside, the overlapping regime. In both cases, the solutions relax over time toward the analytical predictions.

Stochastic dynamics—We now study the finite temperature case, focusing on the attractive and overlapping regimes. In the overlapping regime of Fig. 2, Eq. (5a) describes a Langevin dynamics in a double-well potential. At sufficiently low temperatures, the dynamics of the relative position is confined in the close neighborhood of the two minima $\pm x^*$, while rare trajectories exhibit barrier crossing, going from one minimum to the other. The mean first-passage time can be calculated from Kramers' law as $\tau_K = (2\pi \exp\{[W_-(0) - W_-(x^*)]/T\})/\sqrt{|W_-''(0)W_-''(x^*)|}$ [64,65]. Each time the relative position passes from one minimum to the other, the cluster midpoint X inverts its direction of motion. This results in an *emergent* run-and-tumble dynamics [41,66], which can be modeled as $\dot{X} = v^*e(t) + \sqrt{MT/2\eta_X(t)}$, where $e(t) = \pm 1$ is a random Poisson process with rate $1/\tau_K$ characterizing the tumbling motion [67]. The probability distributions for the stochastic jump times and relative positions obtained from our simulations of Eq. (1) show good agreement with the prediction from Kramers' theory [see Fig. 3(a)] and the Boltzmann distribution associated with the potential W_- [see Fig. 3(c)]. The run-and-tumble dynamics of the midpoint is then characterized by a transition of the mean-squared displacement (MSD) $\langle X(t)^2 \rangle$ from ballistic to diffusive behavior at τ_K , which is consistent with the behavior observed from Eq. (1), as shown in Fig. 3(b).

In the engulfed state at zero temperature, the steady state is a static configuration with the smaller droplet symmetrically placed inside of the larger one, leaving no sign of the chemical activity in the droplet dynamics at large times. However, when temperature is increased, the relative

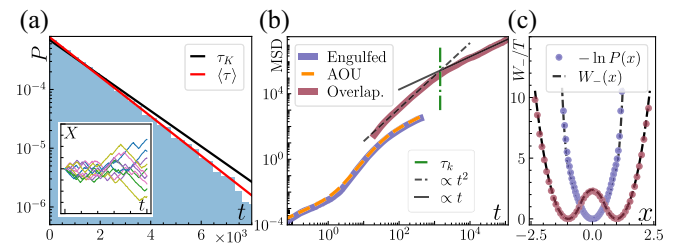


FIG. 3. (a) Jumping times for the relative position in the overlapping regime at $T = 2 \times 10^{-2}$, following a Poisson distribution $\langle \tau \rangle = 1.29 \times 10^3$, compared with $\tau_K = 1.44 \times 10^3$. Inset: center position against time for 10 droplet pairs. (b) MSD of the cluster from simulations of Eq. (1) in the overlapping (red) and engulfed (blue) regimes. The latter follows an AOU process with coefficients obtained from Eq. (5) (dashed orange). (c) Steady-state probability distribution for x . Points show simulations of overlapping (red) and attractive (blue) regimes, compared with Eq. (5) (dashed lines).

position starts to fluctuate around $x = 0$, which in turn affects the dynamics of the midpoint. At small temperatures we can expand Eq. (5b) around $x = 0$, obtaining the equations of motion of an active Ornstein-Uhlenbeck process (AOU) [68,69]. We thus retrieve a signature of the chemical activity of the system in the MSD of the midpoint, and quantify it analytically and via numerical simulations of Eq. (1) [see Figs. 3(b) and 3(c)]. We find that active fluctuations enhance the diffusivity of the engulfed clusters at large times, broadening the range of parameters for which the droplet dimer exhibits self-propulsion. Such active fluctuations can provide the possibility to control the self-organization of these compound active particles [70].

Higher dimensions—We now briefly examine the role of dimensionality to probe the robustness of our findings. Because of rotational symmetry, W_- is a radial function, which can be a attractive, repulsive, or in the form of an (inverted) Mexican hat potential, depending on the radii and chasing rates. When the potential forms a Mexican hat, its degenerate minimum is a circle or sphere of radius x^* . When the potential is strong as compared to the noise, the dynamics is confined to this minimum, such that the relative position satisfies $\mathbf{x}(t) \simeq x^* \hat{\mathbf{e}}(t)$, where $\hat{\mathbf{e}}(t)$ is a unit vector, which evolves according to $\dot{\hat{\mathbf{e}}}(t) = (1/x^*) \sqrt{2\mathcal{M}T} \hat{\mathbf{e}}(t) \times [\hat{\mathbf{e}}(t) \times \boldsymbol{\eta}_x(t)]$. As before, the dynamics of the midpoint reads $\dot{\mathbf{X}} = |\mathbf{v}(x^*)| \hat{\mathbf{e}}(t)$ (ignoring its own noise term). We thus find that the self-propelled cluster behaves effectively like an active Brownian particle [7,71] with angular diffusion coefficient $T\mathcal{M}/(x^*)^2$.

We analytically calculate W_- in $d = 3$ and use it to build the state diagram (see Appendix C and Ref. [55]), which results qualitatively similar to the 1D case. At finite decay rate γ [Fig. 4(a)], nonlocality allows for the formation of self-propelling clusters of mutually attractive droplets, highlighting the general nature of this mechanism and its relevance to the dynamics of multicomponent mixtures. Working in 3D allows us to study Eq. (1) in the case of $\gamma = 0$, which is characterized by the lack of overlapping clusters at $\mathcal{C}_{1 \rightarrow 2}, \mathcal{C}_{2 \rightarrow 1} > 0$ [Fig. 4(a)].

While the same procedure could be carried over also in $d = 2$ (see Appendix C), we choose to take a complementary approach and present a numerical simulation of Eq. (1) in this case. Again, in 2D we find overlapping, self-propelling clusters of droplets with both positive chasing rates [Fig. 4(b)], as well as attractive clusters with AOU behavior. The MSD versus time plot [Fig. 4(c)] shows a transition from ballistic to diffusive motion at a time consistent with our analysis. We also measure the distribution of the separation of the droplets, which in 2D is related to the effective potential as $W_-(\mathbf{x}) \propto -\ln[P(\mathbf{x})/x]$, to estimate the effective potential in the engulfed and overlapping regimes [Fig. 4(d)].

Discussion—We have investigated the dynamics of catalytically active multicomponent mixtures. Our Letter

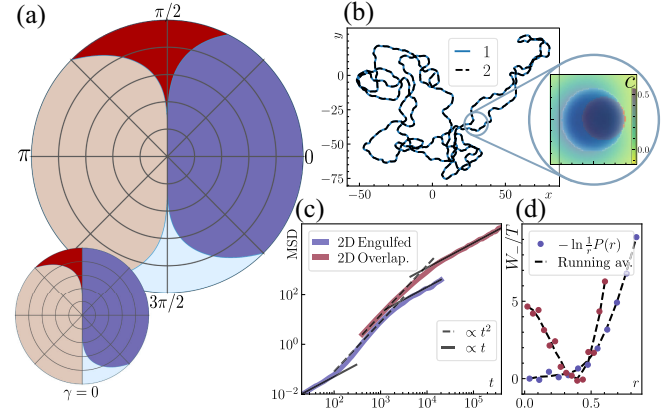


FIG. 4. (a) State diagram at finite (large disk) and vanishing (small disk) decay rate γ in $d = 3$, colors as in Fig. 2. (b) Simulations of Eq. (1) in $d = 2$ at $T = 10^{-4}$, droplet 1 (2) in blue (black). Inset: snapshot of the densities ϕ_a and the chemical field c . (c) MSD for droplets in the attractive (blue) and overlapping (red) configurations. (d) The effective potential for $\mathbf{x}$, inferred from the probability distribution of the relative position. Dashed lines are running averages.

indicates that droplet motion within these mixtures is primarily governed by the properties of their surface tension and reaction kinetics when the interactions causing the phase separation depend on concentrations of chemicals that participate in the catalytic activity. We observe self-propelling clusters in a large part of the state diagram due to the emergence of multiple mechanisms for self-propulsion, which depend on the interplay between the nonlocal nature of the chemical interactions, the sizes of the droplets, and thermal fluctuations. A prominent emerging feature in this system is that the effective coupling constants controlling the nonreciprocal interactions between two droplets depend inversely on the droplet sizes, implying that two droplets can cross the threshold of mutually attractive to chasing interaction simply by ripening. The resulting strong and nontrivial dependence of the properties of self-propulsion on the sizes of the droplets could be of significant help in experimental validation of the aforementioned findings. The self-propulsion induced by chemical activity leads to enhanced diffusion of the droplet clusters, providing a method to probe the chemical activity of cellular subcompartments *in vivo*. We therefore argue our results can help design and analyze chemically active matter, in addition to shedding light on the interplay between activity and nonlocality in extended systems. We note that a direct estimate for the expected magnitude of the droplet velocity will be difficult to surmise in the absence of quantitative information about the values for the field mobility coefficients M_a . However, we can argue that the mechanism that underlies the propulsion is inherently related to the process of droplet coarsening (see the discussion in Supplemental Material [55]), which suggests that the propulsion should be visible in the regimes when the coarsening can be experimentally

probed. Moreover, the scale of propulsion is directly controlled by the rate of chemical activity r_b , which implies that experimental systems with relatively high catalytic rate of reaction such as those involving fast enzymes that produce proton gradients [15] will be the most promising candidates for detecting the proposed phenomenology. In the future, it would be interesting to extend this study to chemically active suspensions with a large number of components [72], incorporate systems with fluid flow to address Marangoni-driven dynamics [73,74], investigate the cases when metabolic fluxes are themselves the drivers of nonequilibrium phase separation [48], and explore possible roles of the emerging phenomenology in origins of life scenarios [21].

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Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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End Matter

Appendix A: The role of interspecies interaction—To test the robustness of our results with respect to the introduction of interspecies interactions, in particular, the existence of the purely attractive chasing states, we add a Flory-Huggins interaction term of the form $\chi\phi_1\phi_2$ to the free energy.

In Fig. 5, we illustrate the effect of χ on the state diagram, as obtained from simulation of Eq. (1) in 1D, together with the self-propulsion velocity. We indeed observe the presence of chasing states at nonzero χ and positive $C_{a \rightarrow b}$. In particular, when $\chi < 0$, the passive interaction between the droplets due to the cross term is also attractive. Nevertheless, simulations show the formation of self-propelling, overlapping states.

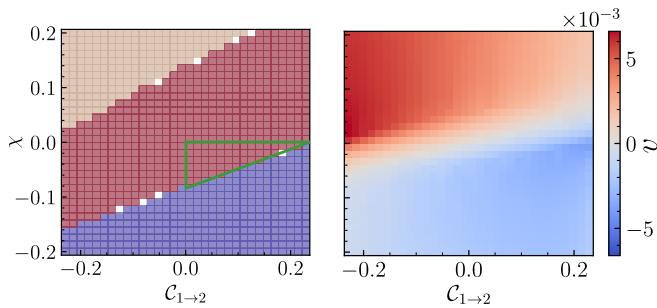


FIG. 5. Simulations with nonzero χ . To the left: state diagram categorizing the behavior of the system, color as in Fig. 2. The green triangle indicates states where all interactions are attractive, but the system still self-propels. To the right: the corresponding steady-state velocity. Parameters: $R_1 = 1$, $R_2 = 1.5$ ($\mathcal{R} = \frac{2}{3}$), and $\partial_c \sigma_2(0) = 0.8$.

Appendix B: Details on numerical simulations—For the numerical integration of the equations of motion, Eq. (1), we have to make a specific choice for free energy. We consider

$$\mathcal{F}_a = \int d^d \mathbf{r} \left[\frac{1}{2} K (\nabla \phi_a)^2 + \frac{1}{4} A_a(c) (\phi_a^2 - \bar{\phi}_a^2)^2 \right], \quad (\text{B1})$$

$$A_a = \left(\sqrt{A_0} + \frac{3}{2\sqrt{2}} \alpha_a c \right)^2, \quad (\text{B2})$$

which yields $A_a(c=0) = A_0$ and $\partial_c \sigma_a(0) = \alpha_a$. This particular choice of the Ginzburg-Landau type potential and function $A_a(c)$ is for convenience, but different choices would lead to the same phenomenology (so long as the system is in the linear response regime for the dependence of the surface tension in the chemical concentration). In simulations, we used $K = 1$, $M_a = 1$, $D_c = 1$, $-r_1 = r_2 = 1$, and $A_0 = 256$. In the simulations for the phase diagrams, velocity, and position, we include a finite but negligible temperature $T = 10^{-5}$ for improved numerical stability. We furthermore set $c(\mathbf{q} = 0) = 0$, as this improves stability without affecting the results.

To classify the phases, we initialize two simulations in the configurations (I) and (III), and check if the final configurations are (I), (II), or (III). This gives us data points such as [(I), (III)] $\rightarrow$ [(I), (I)], which indicate that both simulations evolve to the (I) configuration. This is thus classified as repulsive. The complete classification rules are as follows:

$$\begin{aligned}
 &[(\text{I}), (\text{III})] \xrightarrow{\text{repulsive}} [(\text{I}), (\text{I})], \quad [(\text{I}), (\text{III})] \xrightarrow{\text{engulfed}} [(\text{III}), (\text{III})], \\
 &[(\text{I}), (\text{III})] \xrightarrow{\text{metastable}} [(\text{I}), (\text{III})], \quad \text{and} \quad [(\text{I}), (\text{III})] \xrightarrow{\text{overlapping}} [(\text{II}), (\text{II})].
 \end{aligned}$$

We simulate Eq. (1) using pseudospectral methods with periodic boundary conditions. The system is stiff due to the fourth-order derivative and sharp gradients, which is dealt with by using exponential time stepping [75], as detailed in [76]. For the simulations in 1D, we use a length $L = 16$ and discretize it in $N = 512$ grid points, such that the thickness of the interface is $\xi = \sqrt{2/A_0} = \sqrt{8}dx$. In 2D, we set $N = 64$ and $L = 4$ for the simulation of the attractive cases, and $N = 128$ and $L = 8$ for the overlapping cases.

Appendix C: Surface-averaged concentrations in different dimensions—Here, we present the details of the derivation of the expression for effective phoretic velocity of droplets in arbitrary dimensions. The deterministic part of Eq. (3) reads $\dot{\mathbf{x}}_a = -[(d^2 M_a)/(R_a \Delta \bar{\phi}_a^2)](\partial \sigma_a / \partial c) \times \nabla \bar{c}_{s,a}(\mathbf{x}_a)$, where $\bar{c}_{s,a}(\mathbf{x}_a) = (1/(dV_d)) \int d\hat{\mathbf{n}}_a c(\mathbf{x}_a + R_a \hat{\mathbf{n}}_a)$. We can solve for the chemical concentration by using Eq. (1b) in Fourier space, which yields $c(\mathbf{x}) = \sum_b (r_b/D_c) \int (d^d \mathbf{q}/(2\pi)^d) [\phi_b(\mathbf{q})/(q^2 + \kappa^2)] e^{iq \cdot (\mathbf{x} - \mathbf{x}_b)}$, where $\phi_b(\mathbf{q}) = \int d^d \mathbf{y} \phi_b(\mathbf{y}) e^{-iq \cdot \mathbf{y}} = \int d\hat{\mathbf{n}}_b \int_0^{R_b} r^{d-1} dr e^{-iq \cdot \hat{\mathbf{n}}_b r}$ since $\phi_b(\mathbf{y}) = 1$ for $|\mathbf{y}| \leq R_b$ and $\phi_b(\mathbf{y}) = 0$ for $|\mathbf{y}| > R_b$ in the thin interface limit. Combining these expressions, we obtain

$$\begin{aligned}
 \bar{c}_{s,a}(\mathbf{x}_a) &= \sum_b \left(\frac{r_b}{D_c} \right) \frac{1}{dV_d} \int d\hat{\mathbf{n}}_a \int d\hat{\mathbf{n}}_b \int_0^{R_b} r^{d-1} dr \\
 &\times \int \frac{d^d \mathbf{q}}{(2\pi)^d} \frac{e^{iq \cdot (\mathbf{x}_a - \mathbf{x}_b + \hat{\mathbf{n}}_a R_a - \hat{\mathbf{n}}_b r)}}{q^2 + \kappa^2}. \quad (\text{C1})
 \end{aligned}$$

By introducing the integral $\mathcal{J}_d(p) = (\int_0^\pi d\theta \sin^{d-2} \theta e^{ip \cos \theta}) / (\int_0^\pi d\theta \sin^{d-2} \theta)$, which can be evaluated as $\mathcal{J}_d(p) = 2^{\frac{d}{2}-1} \Gamma(d/2) p^{-\frac{d}{2}} [dJ_{\frac{d}{2}}(p) + pJ_{\frac{d}{2}+1}(p)]$ in terms of the Bessel function J_ν , we can write a closed-form expression for the surface averaged concentration around droplet a as follows:

$$\begin{aligned}
 \bar{c}_{s,a}(\mathbf{x}_a) &= \sum_{b \neq a} \left(\frac{r_b}{D_c} \right) \frac{(dV_d)^2}{(2\pi)^d} \int_0^\infty \frac{dq}{q} \frac{\mathcal{J}_d(qR_a)}{q^2 + \kappa^2} \\
 &\times \mathcal{J}_d(q|\mathbf{x}_a - \mathbf{x}_b|) \int_0^{qR_b} u^{d-1} du \mathcal{J}_d(u) \quad (\text{C2})
 \end{aligned}$$

in any dimension d . The specific forms $\mathcal{J}_1(p) = \cos p$, $\mathcal{J}_2(p) = J_0(p)$, and $\mathcal{J}_3(p) = (\sin p)/p$ can be used to write explicit expressions for $\bar{c}_{s,a}(\mathbf{x}_a)$ in different dimensions, namely

$$\begin{aligned}
 \bar{c}_{s,a}^{(d=1)}(\mathbf{x}_a) &= \frac{2}{\pi} \sum_{b \neq a} \left(\frac{r_b}{D_c} \right) \int_0^\infty \frac{dq}{q} \frac{\cos(qR_a) \sin(qR_b)}{q^2 + \kappa^2} \\
 &\times \cos(q|\mathbf{x}_a - \mathbf{x}_b|) \quad (\text{C3})
 \end{aligned}$$

for $d = 1$,

$$\begin{aligned}
 \bar{c}_{s,a}^{(d=2)}(\mathbf{x}_a) &= \sum_{b \neq a} \left(\frac{r_b R_b}{D_c} \right) \int_0^\infty dq \frac{J_0(qR_a) J_1(qR_b)}{q^2 + \kappa^2} \\
 &\times J_0(q|\mathbf{x}_a - \mathbf{x}_b|) \quad (\text{C4})
 \end{aligned}$$

for $d = 2$, and

$$\begin{aligned}
 \bar{c}_{s,a}^{(d=3)}(\mathbf{x}_a) &= \frac{2}{\pi} \sum_{b \neq a} \frac{1}{|\mathbf{x}_a - \mathbf{x}_b|} \left(\frac{r_b}{D_c R_a} \right) \int_0^\infty \frac{dq}{q^3} \frac{\sin(qR_a)}{q^2 + \kappa^2} \\
 &\times [\sin(qR_b) - (qR_b) \cos(qR_b)] \sin(q|\mathbf{x}_a - \mathbf{x}_b|) \quad (\text{C5})
 \end{aligned}$$

for $d = 3$. These expressions are in agreement with those obtained using a more direct integration method.

In the special case where the droplets do not overlap, we obtain a closed expression in the form of generalized phoresis. To this end, we use a formal representation of the Taylor expansion as $\bar{c}_{s,a}(\mathbf{x}_a) = (1/(dV_d)) \int d\hat{\mathbf{n}}_a e^{R_a \hat{\mathbf{n}}_a \cdot \nabla} c(\mathbf{x}_a)$, which can be written formally as $\bar{c}_{s,a}(\mathbf{x}_a) = \mathcal{I}_d(R_a \nabla) c(\mathbf{x}_a)$, where $\mathcal{I}_d(b) \equiv \mathcal{J}_d(p = -ib)$ is given as $\mathcal{I}_d(b) = 2^{\frac{d}{2}-1} \Gamma(\frac{d}{2}) b^{-\frac{d}{2}} [dI_{\frac{d}{2}}(b) + bI_{\frac{d}{2}+1}(b)]$ in terms of the modified Bessel function of the first kind I_ν . In particular, we have $\mathcal{I}_1(b) = \cosh b$, $\mathcal{I}_2(b) = I_0(b)$, and $\mathcal{I}_3(b) = (\sinh b)/b$. Since we are interested in contributions to c from $b \neq a$ and because the droplets do not overlap, the right-hand (source) side of Eq. (1b) is zero in the region of interest, and we can write $\nabla^{2n} c = \kappa^{2n} c$ for $n \geq 1$. Combining these results, we obtain $\dot{\mathbf{x}}_a = -[(d^2 M_a)/(R_a \Delta \bar{\phi}_a^2)](\partial \sigma_a / \partial c) \mathcal{I}_d(\kappa R_a) \nabla c(\mathbf{x}_a)$. Note that the effective phoretic mobility arising from this mechanism shown in Eq. (4) exhibits a leading order $1/R$ size dependence in any dimension d , which is in sharp contrast with the case of standard diffusiophoresis where the mobility is independent of size [1]. Moreover, in the derivation of this result no thin slip-layer assumption is used [46,63], and near-field effects are taken into consideration [77,78], as represented by the function $\mathcal{I}_d(\kappa R_a)$, which introduces additional d -dependent size dependence.

Appendix D: Effective potentials in 1D—We report here the expression for $W_\pm$ in Eq. (5) for $d = 1$. We consider the three subdomains corresponding to the three possible configurations, as illustrated in Fig. 1(c): (I) disjoint, (II) partially overlapping, and (III) completely overlapping. These correspond to $|x| > R_1 + R_2$, $R_2 - R_1 < |x| < R_1 + R_2$, and $|x| < R_2 - R_1$, respectively.

The effective potentials $W_{\pm}(x)$ are given by

$$W_{\pm}(x) = \frac{1}{\mathcal{M}} \begin{cases} -2^{-\frac{(1\pm 1)}{2}} e^{-|x|} \left[\frac{C_{2\pm 1}}{R_2} \cosh R_2 \sinh R_1 \mp \frac{C_{1\pm 2}}{R_1} \cosh R_1 \sinh R_2 \right] + \text{const}, & \text{(I)} \\ 2^{-\frac{(3\pm 1)}{2}} e^{-(R_1+R_2)} \left[\left(\frac{C_{2\pm 1}}{R_2} \pm \frac{C_{1\pm 2}}{R_1} \right) \cosh x + e^{-|x|} \left(\frac{C_{2\pm 1}}{2R_2} \mp \frac{C_{1\pm 2}}{2R_1} \right) (e^{2R_2} - e^{2R_1}) \right], & \text{(II)} \\ -2^{-\frac{(1\pm 1)}{2}} e^{-R_2} \left[\frac{C_{2\pm 1}}{R_2} \sinh R_1 \pm \frac{C_{1\pm 2}}{R_1} \cosh R_1 \right] \cosh x + \text{const}. & \text{(III)} \end{cases} \quad (\text{D1})$$