Letter

Diversity of phase transitions and phase separations in active fluids

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Active matter is not only indispensable to our understanding of diverse biological processes, but also provides a fertile ground for discovering novel physics. Many emergent properties impossible for equilibrium systems have been demonstrated in active systems. These emergent features include motility-induced phase separation, a long-ranged ordered (collective motion) phase in two dimensions, and order-disorder phase coexistences (banding and reverse-banding regimes). Here, we unify these diverse phase transitions and phase coexistences into a single formulation based on generic hydrodynamic equations for active fluids. We also reveal a novel comoving coexistence phase and a multicritical point.

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Active matter refers to many-body systems in which each volume element can generate its own mechanical stresses [1–3]. As the fluctuation-dissipation relation is broken at the microscopic level, active matter can be viewed as an extreme form of far-from-equilibrium systems. Given the relevance of active matter to nonequilibrium physics and biophysics, the subject area has been rapidly expanding and many approaches have been used to study this diverse class of nonequilibrium, many-body systems. Arguably, the most generic way to investigate the emergent properties of an active matter system is to first formulate a model based solely on the underlying symmetries and conservation laws of the system [4].

This is what was done in the case of active fluids—a class of active matter in which translation invariance holds—in the seminal work by Toner and Tu [5–8]. Motivated by the simulation study by Vicsek *et al.* [9], Toner and Tu provided the generic equations of motion (EOM) for polar active fluids and demonstrated the existence of the polar ordered, or collective motion, phase using a renormalization group analysis. Subsequently, a coexistence regime consisting of the ordered and disordered phases was also found, which generically separates the disordered phase and the ordered phase in typical polar active fluid models [10–15]. Numerous studies have also confirmed the Toner-Tu EOM for polar active fluids using formal coarse-graining strategies that link microscopic models of self-propelled particles and hydrodynamic level equations [16–22].

Concurrently, dense collections of active particles without aligning interactions were shown to spontaneously phase separate in the presence of purely steric interactions. This phenomenon is now known as *motility-induced phase separation* (MIPS) [23]; it was first predicted theoretically and simulationally [24–26], and then experimentally observed [27,28]. Scalar field theories, typically based on the density field of the particles, have also been formulated to describe this process, e.g., the so-called *Active Model B* [29,30]. In terms of symmetries and conservation laws, MIPS and polar active fluids are completely identical. It is therefore natural to view the emergence of the ordered phase and MIPS as properties of the same class of active systems. Through scattered efforts, recent studies have attempted to gain insight into the competition between Vicsek-like aligning interactions and steric repulsion in experiments [31] and in models [32–37] of active particles. Nevertheless, our understanding of the connections between the emergence of collective motion and phase separation is still crucially lacking. In this Letter, we elucidate the interplay between these phenomena; specifically, we unify these diverse types of phases and phase coexistences in a single formulation based on generic hydrodynamic EOM for active fluids. In the process, we also uncover a novel coexistence regime and a multicritical point.

Conservation law and symmetries.— Our model EOM are based on the conservation law and symmetries in the system. Specifically, mass conservation leads to the continuity equation

$$\partial_t \rho + \nabla \cdot \mathbf{J} = 0, \tag{1}$$

where the total flux $\mathbf{J} = \mathbf{p} - \eta \nabla \rho$ is composed of an *active flux* $\mathbf{p}$ and a *gradient term* (leading to a diffusive term in the continuity equation).

For the EOM of the active flux $\mathbf{p}$, following Refs. [5–8], we impose temporal, translation, rotation, and chiral invariances to obtain

$$\partial_t \mathbf{p} + \lambda(\mathbf{p} \cdot \nabla)\mathbf{p} = \mu \nabla^2 \mathbf{p} - \kappa(\rho)\nabla\rho + \alpha(\rho)\mathbf{p} - \beta p^2 \mathbf{p}, \tag{2}$$

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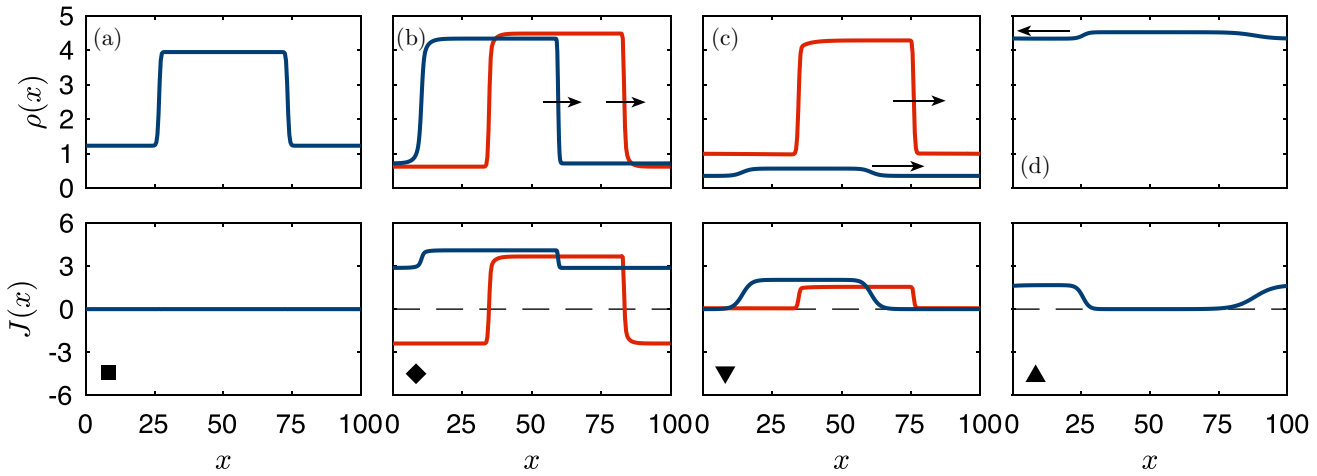

FIG. 1. Phase coexistence in a one-dimensional active fluid. The mass density ρ (upper panels) and total flux J (lower panels) profiles of the four distinct types of phase coexistences: (a) a dilute (**d**) and disordered (**D**) phase coexisting with a condensed (**c**) and disordered (**D**) phase, denoted as **dD-cD**; (b) two possible **dO-cO** coexistences where condensed and dilute phases are transported in the same direction (blue) or in opposite directions (red); in both cases shown here, the condensed ordered phase is moving to the right as the magnitude of the flux in the condensed phase $|J_c|$ is higher than that in the dilute phase $|J_d|$; (c) two examples of **dD-cO** coexistence, where in both cases, the condensed ordered phase is moving to the right; and (d) **dO-cD** coexistence, the condensed disordered phase is here moving to the left due to differential adsorption at the two interfaces. The profiles shown here correspond to the stationary states of the hydrodynamic EOM. In the lower panels, the symbols correspond to the symbols shown in Fig. 2. Movies of these cases can be found in the Supplemental Material [38].

where we have only retained the terms crucial to our discussion here. We have also emphasized the density dependency of the "compressibility" coefficient κ and that of the "order-disorder" control parameter α in the above equations.

Note that our EOM differ from the Toner-Tu EOM in our choice of hydrodynamic variables and the imposition of the diffusive term in the EOM of ρ. Indeed, while ρ describes our active fluid mass density, $\mathbf{p}$ denotes here an active flux and can only formally be identified with the momentum density when the diffusive term vanishes ($\eta \to 0$), in which case our EOM reduce exactly to the reduced Toner-Tu EOM. The presence of this diffusive term facilitates our numerical analyses of the EOM and was commonly adopted in previous studies [25,33,39,40]. However, since we recover diverse salient features known in polar active fluids, we are confident that the findings in our Letter remain valid for generic active fluids as described by the Toner-Tu EOM. In particular, we show in the Supplemental Material [38] that our results are qualitatively unchanged by the presence of the diffusive term.

Diversity of phase separations.— Phase separation occurs in systems with a conserved quantity. Here, the conserved quantity is the total mass and so a phase coexistence consists of one condensed density phase (denoted by **c**) and one dilute density phase (denoted by **d**). At the same time, the Toner-Tu model allows for two distinct spatially homogeneous phases, the disordered (**D**) and the ordered (**O**) phases, characterized by whether or not the nonconserved order parameter $|\langle \mathbf{p} \rangle|$ is zero. We therefore generically expect four possible phase coexistences: (i) **dD-cD** (i.e., a dilute disordered phase coexisting with a condensed disordered phase), (ii) **dD-cO**, (iii) **dO-cD**, and (iv) **dO-cO** (see Fig. 1). Indeed, three out of these four coexistences have already been demonstrated: (i) corresponds to MIPS, (ii) corresponds to the banding regime, and (iii) corresponds to the recently uncovered reverse-banding regime [41–43]. Type (iv) coexistence still needs to be demonstrated; here, we first predict analytically and then confirm its existence numerically (see Fig. 1) in a particular model. To that end, we will first describe how a generic phase diagram can be constructed approximately following a linear stability analysis.

Linear stability and phase separation.— In thermal phase separation, a linear stability analysis of the dynamical equation of a phase separating system (e.g., Cahn-Hilliard equations) can reveal the spinodal decomposition region of the phase diagram of the system [44]. Furthermore, a signature of phase separation is that the most unstable mode from the linear instability analysis corresponds to the $k \to 0$ mode where k is the wave number. We will use these criteria as our guiding principles in constructing an approximate phase diagram for a particular hydrodynamic model. Specifically, we will perform a linear stability analysis on the EOM and focus on the $k \to 0$ limit. Furthermore, since in the disordered phase, the instability has no direction dependency, while in the ordered phase, the most unstable direction is longitudinal to the direction of the collective motion, the initial perturbation in our stability analysis is taken to be along the direction of the ordered state [18,19]. We note that all known examples of phases and phase coexistences in polar active fluids can be qualitatively understood in quasi-one-dimensional (1D) geometries, from long-ranged collective motion in ordered homogeneous phases to the one-dimensional bands observed in phase coexistences. We therefore believe that our one-dimensional analytic treatment of this problem is sufficient to capture the nature of phase coexistences in general polar active fluids, even in higher dimensions.

As an example, in the disordered case ($\alpha < 0$), we expand around the homogeneous disordered state with $\rho = \rho_0 + \delta\rho \exp[st - ikx]$, $p = \delta p \exp[st - ikx]$, where we have

arbitrarily chosen the x direction to be the direction of interest. We note that in the Toner-Tu theory, symmetries generically allow all the phenomenological coefficients appearing in Eq. (2) to be functionally dependent on the density ρ. We thus proceed to Taylor expand κ and α in (2) as follows:

$$\kappa(\rho) = \sum_{i=0}^{\infty} \kappa_i \delta\rho^i \quad \text{and} \quad \alpha(\rho) = \sum_{i=0}^{\infty} \alpha_i \delta\rho^i. \tag{3}$$

To linear order, the EOM read

$$\begin{aligned} \partial_t \delta\rho &= -\partial_x \delta p + \eta \partial_x^2 \delta\rho, \\ \partial_t \delta p &= \mu \partial_x^2 \delta p - \kappa_0 \partial_x \delta\rho - |\alpha_0| \delta p. \end{aligned} \tag{4}$$

Solving for s and focusing on the hydrodynamic limit ($k \to 0$), we have

$$s = \begin{cases} -|\alpha_0| + \left(\frac{\kappa_0}{|\alpha_0|} + \mu\right)k^2 + \mathcal{O}(k^3), \\ -\left(\frac{\kappa_0}{|\alpha_0|} + \eta\right)k^2 + \mathcal{O}(k^3). \end{cases} \tag{5}$$

The first eigenvalue ($-|\alpha_0|$) corresponds to the fast relaxation when the active flux deviates from the mean-field value $p_0 = 0$ in the absence of spatial variations. The second eigenvalue quantifies when the instability sets in, which happens whenever $\kappa_0 + \eta|\alpha_0|$ becomes negative. Since the system is in the disordered regime, within this instability region, the system exhibits **dD-cD** coexistence as shown in Fig. 1(a).

The analysis in the ordered regime ($\alpha > 0$) follows the exact same procedure; the full details of the linear stability analysis can be found in the Supplemental Material [38]. Here, we just recall that when $\kappa - \eta\alpha > 0$, the homogeneous disordered phase will generically be separated from the homogeneous ordered phase through phase separation. We will now present a particular model that illustrates the diversity of phase transitions and phase coexistences possible in active fluids.

A model with all four phase coexistences.—The linear stability analysis can be applied straightforwardly once $\kappa(\rho)$ and $\alpha(\rho)$ in (2) are explicitly defined. Here, we consider the following minimal model,

$$\alpha(\rho) = -A + 18\rho - 10/3\rho^2, \tag{6}$$

$$\kappa(\rho) = 140 - 145\rho + 30\rho^2, \tag{7}$$

with $\eta = 2$, $\lambda = 1$, $\beta = 0.5$, and $\mu = 1$. A microscopic-level (particle based) system that realizes this model could for instance be a polar active fluid with contact inhibition of alignment (e.g., as discussed in Ref. [42]) such that its equation of state dictates that it can also phase separate within the homogeneous ordered phase.

For large enough values of A, α remains negative, and so the system remains in the disordered phase. In this case, instability occurs if $(\kappa - \eta\alpha) < 0$, and we expect **dD-cD** coexistence in this parameter range. On the other hand, as A decreases, the range of densities ρ for which the system is in the ordered phase gets wider, and, importantly, is separated from the disordered phase by two instability regions: A **dD-cO** coexistence to the left and a **dO-cD** coexistence to the right. Simultaneously, $(\kappa - \eta\alpha)$ remains negative around $\rho \sim 2.5$. We therefore expect an interesting interplay of distinct phase separations.

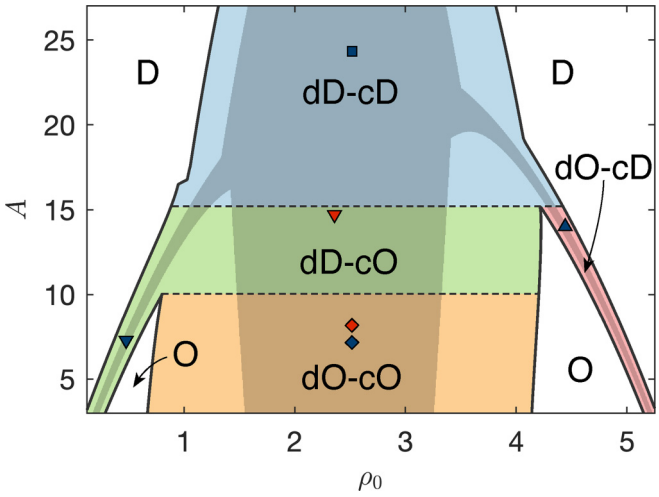

FIG. 2. Phase diagram of a one-dimensional active fluid. The shaded area corresponds to the instability regime obtained from the linear stability analysis of the hydrodynamic model (6) and (7). The edges of the instability region correspond to the *spinodal* lines. Black lines correspond to the *binodal* lines (or coexistence lines), which were obtained via direct numerical simulations of the one-dimensional hydrodynamic model in (6) and (7). Along with the homogeneous disordered (**D**) and homogeneous ordered (**O**) phases, we observe four coexistence regions which are delimited by the binodal lines (and horizontal dashed lines): A coexistence of a dilute disordered phase with a condensed disordered phase (**dD-cD**, blue), a coexistence of a dilute disordered phase with a condensed ordered phase (**dD-cO**, green), a coexistence of a dilute ordered phase with a condensed disordered phase (**dO-cD**, red) and a coexistence of a dilute ordered phase with a condensed ordered phase (**dO-cO**, orange). The symbols (and their colors) denote the locations in phase space from which we extracted the profiles shown in Fig. 1.

In Fig. 2, the instability regions resulting from our linear stability analysis are shown as the shaded area, while the homogeneous disordered (**D**) and ordered (**O**) regions are shown in white. We equate the instability region to be within the phase separating region, but to which of the four possible types of phase coexistences?

Since the conserved quantity here is the total mass, ρ can be redistributed as long as the overall density remains the same. Therefore, we can characterize the phases as follows: Given any starting point on the phase diagram within the instability region (shaded area in the phase diagram), we extend a horizontal line from that point; the first homogeneous phase encountered to the right (respectively, to the left) will describe the nature of the condensed (respectively, dilute) phase (**D** or **O**).

Using the above construction, we see that this particular model contains all four variations of the phase coexistences (Fig. 2). As aforementioned, we report the existence of a phase coexistence in which both the dilute and condensed phases are ordered. *A priori*, these two comoving phases can move either in the same direction or in opposite directions. By directly solving the hydrodynamic EOM numerically [38], we find that both scenarios are possible, as evidenced in the steady-state profiles shown in Fig. 1(b). Besides this particular model, we note that a further diversity of phase diagrams is rendered

possible by varying the specific definitions of $\alpha(\rho)$ and $\kappa(\rho)$; we discuss other interesting cases in the Supplemental Material [38].

Instability versus phase separation.—The instability region obtained from a linear stability analysis does not correspond exactly to the whole phase separation region. Indeed, as in thermal phase separation, the instability region in fact corresponds to the spinodal decomposition region, which is always flanked by the so-called nucleation and growth regions on either side [44,45]. This is no different here: The actual phase separation boundaries encapsulate the instability regions (Fig. 1). Of course, while the phase separation boundaries (i.e., the binodal lines) for thermal systems in equilibrium can be obtained by analyzing the free energy, e.g., by using the Maxwell tangent method, no free energy exists in our nonequilibrium systems and so the phase separation boundaries will instead be given by the appropriate boundary conditions obtained from the stable steady-state solution of the actual hydrodynamic EOM. This is exactly what we did to obtain the profiles shown in Fig. 1. Specifically, the locations of the binodal lines correspond to the density values of the stationary regions of the condensed and dilute phases (see Supplemental Material [38] for further details). Finally, we note that both density and active flux profiles obtained numerically display a characteristic fore-rear asymmetry with a steeper fore-front (see Fig. 1). This asymmetry was already observed and discussed in both simulations of microscopic Vicsek-like and active Ising spins models, and their continuum counterparts [13,46,47].

A multicritical point.—Besides uncovering the **dO-cO** coexistence regime, our analysis also reveals the existence of a multicritical point pertaining to a potentially new universality class. To illustrate this (Fig. 3), we consider a polar active fluid system in which there are two generic parameters K_1 and K_2 that control the phase behavior of the system. Specifically, the system undergoes **dD-cD** phase separation at high K_1 while at small K_1 the system is in the ordered phase. In addition, we assume that the second parameter K_2 controls the threshold level K_1 at which the distinct phase separations happen (Fig. 3). In other words, instead of having an additional phase separation due to a negative κ inside the homogeneous ordered phase as in the previous example, we have here a **dD-cD** phase coexistence in the homogeneous disordered phase instead.

Now, **dD-cD** phase separation at criticality belongs to the Ising universality class [48] (but see also Refs. [49,50]). In terms of our hydrodynamic EOM, this critical point corresponds to having $\alpha > 0$, $\kappa_0 = \kappa_1 = 0$ in (3). On the other hand, the order-disorder critical point that accompanies critical **dD-cO** and **dO-cD** phase separations belongs putatively to a different universality class ($\kappa > 0$, $\alpha_0 = \alpha_1 = 0$) [42]. Therefore, by fine tuning κ_0, κ_1, α_0, and α_1 to zero (indicated by the yellow ball in Fig. 3), these two distinct critical points coincide and the resulting multicritical point is likely to correspond to yet a distinct universality class for the following reasons: At the linear level around this critical point, the EOM of the active flux $\mathbf{p}$ is completely decoupled from that of the density field ρ. Specifically, the linear EOM are

$$\partial_t \delta\rho + \nabla \cdot \mathbf{p} = \eta \nabla^2 \delta\rho, \quad \partial_t \mathbf{p} = \mu \nabla^2 \mathbf{p} + \mathbf{f}, \tag{8}$$

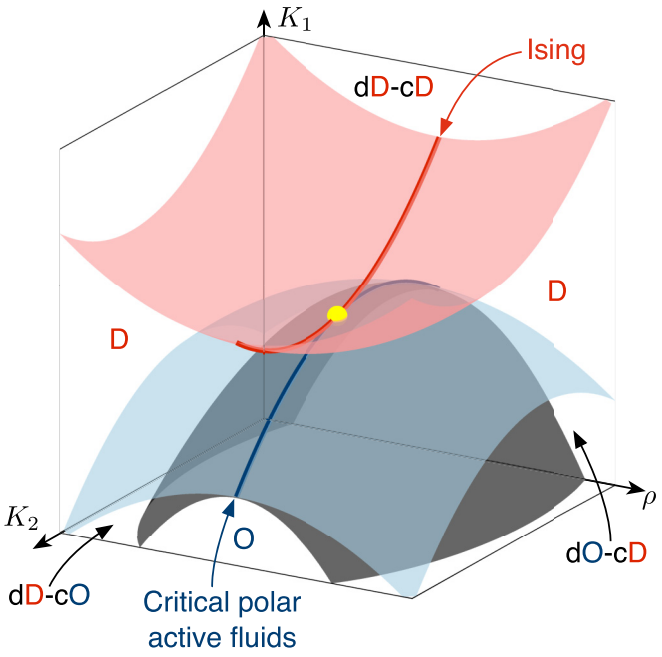

FIG. 3. Multicritical point. In this schematic, the **dD-cD** coexistence (or MIPS) occurs above the red surface (at high K_1), and the corresponding critical phase separation (indicated by the red line) belongs to the Ising universality class [48] (but see also Refs. [49,50]). In contrast, an ordered phase coexists with a disordered phase (**dD-cO** or **dO-cD**) between the blue and gray surfaces, and a spatially homogeneous ordered phase (**O**) exists below the gray surface. The corresponding order-disorder critical line is indicated by the blue line [42]. When the blue line and the red lines are tangent to each other (indicated by the yellow dot), a putatively novel multicritical point emerges.

where $\mathbf{f}$ is a Gaussian noise term with a nonzero standard deviation. The fact that $\delta\rho$ does not feature in the linear EOM of $\mathbf{p}$ is distinct from all known active fluids at the order-disorder critical transition [42,51–57].

Using the linear theory above, we can also identify some interesting features of this critical point. To do so, we first perform the following rescalings:

$$\mathbf{r} \mapsto e^{\ell}\mathbf{r}, \quad t \mapsto e^{z\ell}t, \tag{9}$$

$$\delta\rho \mapsto e^{\chi_\rho \ell}\delta\rho, \quad \mathbf{p} \mapsto e^{\chi_p \ell}\mathbf{p}. \tag{10}$$

We can then conclude that the following choice of scaling exponents leave the linear EOM invariant [38]:

$$z = 2, \quad \chi_p = \frac{2-d}{2}, \quad \chi_\rho = \frac{4-d}{2}. \tag{11}$$

Applying these linear exponents to the generic nonlinear EOM then indicates that (i) the upper critical dimension d_c is 6 and (ii) the first two nonlinear terms that become relevant right below d_c are $\delta\rho^2\mathbf{p}$ and $\nabla(\delta\rho^3)$ in the EOM of $\mathbf{p}$ [38].

Summary and outlook.—Starting from generic hydrodynamic EOM of polar active fluids, we have unified existing phase transitions and phase separations into a single formulation. In particular, we showed that there are generically four distinct types of phase separations, and illustrated

them with a particular model. In doing so, we exhibited a novel coexistence regime: The coexistence of a dilute ordered phase and a condensed ordered phase. We expect that this phase coexistence will be observed in a microscopic model of active Brownian particles with steric repulsion and velocity-alignment interactions. The numerical study of such a microscopic model and its coarse graining to link microscopic parameters to the phenomenological coefficients appearing in our EOM is of great interest and will be the subject of further studies. Moreover, we also revealed the existence of a critical behavior pertaining putatively to a new universality class. Our work highlights the richness of generic polar active fluid models. The phase behavior can be further enriched by considering variations in other parameters in the EOM. For instance, patterns other than phase separation have been observed when the coefficient μ in the EOM of the momentum density field (2) becomes negative [58]. We believe that elucidating these diverse phase behaviors will be a fruitful research direction in the future.

Note added.—After our paper was submitted, Jentsch and Lee have used functional renormalization group analysis to show that the multicritical point uncovered here indeed constitutes a distinct universality class [59].

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