The Phase Synchronized State of Oriented Active Fluids

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We present a theory for self-driven fluids, such as motorized cytoskeletal extracts or microbial suspensions, that takes into account the underlying periodic duty cycle carried by the constituent active particles. We show that an orientationally ordered active fluid can undergo a transition to a state in which the particles synchronize their phases. This spontaneous breaking of time-translation invariance gives rise to flow instabilities distinct from those arising in phase-incoherent active matter. Our work is of relevance to the transport of fluids in living systems, and makes predictions for concentrated active-particle suspensions and optically driven colloidal arrays.

The field of broken-symmetry hydrodynamics [1] is seeing increasing success in describing the large-scale behavior of spatially ordered active systems [2–6]. In this Letter we explore the consequences of the spontaneous breaking of time-translation invariance [7, 8] for orientationally ordered fluids, uncovering the existence of a phase coherent state of active matter, which we expect to be seen in experiments and to be relevant to fluid transport in biological systems [9].

Time-periodicity in a macroscopic system can arise either through large-scale spontaneous wavelike motion in a system without distinct local oscillators [10, 11] or via phase-locking of particle-scale periodic processes [4, 8, 12, 13], the case with which this Letter is concerned. Active particles [14], such as motor proteins [15, 16], motile organisms [17], or non-living imitations thereof [18–21] convert energy from a reservoir into mechanical work. This energy conversion process generally involves a periodic duty cycle (Fig. 1), quantified by a phase variable \( \theta \in [0, 2\pi] \) and is responsible for the force- and torque-free motion of an active particle. Hydrodynamic theories for fluid or liquid-crystalline phases of active particles – active fluids, for short – have been developed and predict that these materials can flow spontaneously if the stresses generated by the active particles are large enough [22–30]. Many biological materials are active fluids [31, 33]. The emergence of local oscillators in active systems was treated in [34, 35]. It is natural to ask how synchronization physics [7, 8, 12] enriches active-matter hydrodynamics, usually presented in terms of the force and torque dipoles that emerge from an incoherent time-average of the underlying duty cycles. In this Letter we explore the broken-symmetry hydrodynamics of a phase-coherent active fluid [36].

We present a generic hydrodynamic theory of active particles in a fluid medium, accounting for their phases and thus extending earlier theories for phase-incoherent active fluids. To this end, we reformulate low-Reynolds-number active-matter hydrodynamics in terms of phase-dependent force and torque dipoles and obtain generic expressions for their phase dynamics using the tools of nonequilibrium thermodynamics [37]. Our work generalizes the theory of hydrodynamic synchronization [12] to oriented and spatially extended systems. Within this framework we study the stability of the phase-incoherent state of a suspension of active particles and identify conditions under which the particles synchronize. We consider: (i) active shakers which produce periodic contributions to the symmetric part of the active stress; (ii) active polar rotors which produce periodic contributions to the antisymmetric part of the active stress; and (iii) active nematic rotors which produce periodic contributions to the angular momentum flux (Fig. 2). We find that all three systems can synchronize by hydrodynamic interactions (Fig. 3). We find, however, that the fully synchronized homogeneous steady state is generically unstable at zero wavenumber and conclude that the broken time-translation symmetry gives rise to a class of flow instabilities absent in the phase-incoherent state of active matter (Fig. 4). We expect that the phenomena we predict will be seen in experiments on active-particle fluids at high concentrations, such as realizations of the oscillating active filaments of [35], and optically driven arrays of colloidal oscillators or rotors [38, 39].

Consider a suspension of active particles which carry a phase variable. In the absence of external forces and torques, the linear and angular momentum balance equa-

![FIG. 1: Duty cycles of active processes. Sketch of (a) the swimming stroke of the alga chlamydomonas and (b) a molecular motor moving along a cytoskeletal filament.](image-url)
tions of a low Reynolds number fluid are given by

\[ \partial_\beta (\sigma_{\alpha\beta} + \sigma_{a\beta}^a + \sigma_{\alpha\beta}^a) = 0 \]  
(1)

\[ \frac{1}{2} \partial_\epsilon (M_{\alpha\beta\gamma} + M_{\epsilon\beta\gamma}) = \sigma_{\alpha\beta}^a , \]  
(2)

where \( \sigma_{\alpha\beta} \) and \( \sigma_{a\beta}^a \) are the symmetric and antisymmetric parts of the deviatoric stress, respectively, and \( M_{\alpha\beta\gamma} \) is the deviatoric angular momentum flux. Furthermore, \( \sigma_{\alpha\beta}^a \) and \( M_{\epsilon\beta\gamma}^a \) are the hydrostatic contributions to the stress and angular momentum flux, respectively, see [30, 40, 41].

The deviatoric stress and angular momentum flux consist of contributions stemming from the fluid (labelled \( f \)) and the particles (labelled \( act \)): \( \sigma_{\alpha\beta}^f = \sigma_{\alpha\beta}^{af} + \sigma_{\alpha\beta}^{act} \) and \( M_{\alpha\beta\gamma}^f = M_{\alpha\beta\gamma}^{af} + M_{\epsilon\beta\gamma}^{act} \). In the following we consider an incompressible fluid which obeys the constitutive equations, \( \sigma_{\alpha\beta}^f = 2\eta \sigma_{\alpha\beta}^f \) and \( M_{\alpha\beta\gamma}^f = 2\eta \alpha\beta \Omega_{\alpha\beta}^f \), where \( \eta, \eta' \) are viscosities and \( \kappa \) is the internal rotational friction of the fluid. Here we introduced the strain rate \( u_{\alpha\beta} = (\partial_i v_\beta + \partial_\beta v_i)/2 \), the vorticity \( \omega_{\alpha\beta} = (\partial_i v_\beta - \partial_\beta v_i)/2 \), where \( v \) is the center of mass velocity of the fluid, and \( \Omega_{\alpha\beta}^f \) is the intrinsic ‘spin’ rotation rate of the fluid \( \Omega_{\alpha\beta}^f \).

The active symmetric stress generated by a collection of \( N \) particles is

\[ \sigma_{\alpha\beta}^{act} = \sum_{i=0}^{N} D_{\alpha\beta}^{i} \delta (\mathbf{r} - \mathbf{r}^{(i)}) , \]  
(3)

where \( \mathbf{r}^{(i)} \) is the position of the \( i \)-th particle and \( D_{\alpha\beta}^{i} \) is its intrinsic force dipole \[41, 43\], generated by an underlying periodic process. We define the associated phase variable \( \theta_0^{(i)} \) by

\[ D_{\alpha\beta}^{i} = s_0 (\theta_0^{(i)}) \hat{\theta}_0^{(i)} Q_{\alpha\beta}^{(i)} , \]  
(4)

where \( s_0 (\theta_0^{(i)}) \) is the phase dependent action of the swimming stroke. The axis of the dipole is given by the nematic tensor \( Q_{\alpha\beta}^{(i)} = p_\alpha^{(i)} p_\beta^{(i)} - \delta_{\alpha\beta}/d \), where the unit vector \( p^{(i)} \) denotes the particle orientation and \( d \) is the number of spatial dimensions. See Fig. 2.a.

Similarly the antisymmetric active stress and the active angular momentum flux generated by a collection of \( N \) particles are given by

\[ \sigma_{a\beta}^{act} = \sum_{i=0}^{N} \tau_{a\beta}^{(i)} \delta (\mathbf{r} - \mathbf{r}^{(i)}) , \]  
(5)

\[ M_{\alpha\beta\gamma}^{act} = \sum_{i=0}^{N} T_{\alpha\beta\gamma}^{(i)} \delta (\mathbf{r} - \mathbf{r}^{(i)}) , \]  
(6)

where \( \tau_{a\beta}^{(i)} \) and \( T_{\alpha\beta\gamma}^{(i)} \) are the internal torque and the torque dipole generated by the \( i \)-th particle, respectively \[41, 43\]. In parallel to Eq. 4 we define the phases \( \phi_1^{(i)} \) and \( \theta_1^{(i)} \) of the periodic processes generating active internal torques and angular momentum fluxes, respectively:

\[ \tau_{\alpha\beta}^{(i)} = s_1 (\theta_1^{(i)}) \hat{\theta}_1^{(i)} m_{\alpha\beta\gamma}^{(i)} p_{\gamma}^{(i)} , \]  
(7)

\[ T_{\alpha\beta\gamma}^{(i)} = s_2 (\hat{\phi}_2^{(i)}) \hat{\theta}_2^{(i)} n_{\alpha\beta\gamma}^{(i)} , \]  
(8)

where \( n_{\alpha\beta\gamma}^{(i)} = \epsilon_{\alpha\beta\gamma} \rho_i^{(i)} p_{\gamma}^{(i)} \), with corresponding phase dependences carried by functions \( s_1 \) and \( s_2 \). See Fig. 2.b, c.

We obtain dynamical equations for the particle phase variables following the logic outlined in [11, 25, 41]. The power dissipated by a single active particle embedded in a fluid at temperature \( T \) is

\[ T \Phi = 8 s_0 \theta_0^{(i)} Q_{\alpha\beta}^{(i)} u_{\alpha\beta} + s_1 \theta_1^{(i)} m_{\alpha\beta\gamma}^{(i)} \partial_\gamma \Omega_{\alpha\beta} \]  
+ \[ s_2 \hat{\phi}_2^{(i)} \epsilon_{\alpha\beta\gamma} \rho_i^{(i)} (\Omega_{\alpha\beta} - \omega_{\alpha\beta}) + r^{(i)} \Delta \mu \]  
(9)

where \( r^{(i)} \) denotes the rate at which the \( i \)-th particle consumes fuel from its energy reservoir and \( \Delta \mu \) is the amount of free energy carried by one unit of fuel. Note that in cases where the \( \theta_1^{(i)} \) originate from a common periodic process, additional passive couplings amongst them need to be considered. We ignore these, and the corresponding contributions to \( \Phi \) and \( T \) below, for simplicity. We write dynamic equations for the phase variables by expanding the thermodynamic fluxes \( s_0 \theta_1, s_1 \theta_1, s_2 \hat{\phi}_2 \) and \( s_2 \hat{\phi}_2 \) in terms of the thermodynamic forces \( Q_{\alpha\beta} u_{\alpha\beta}, \epsilon_{\alpha\beta\gamma} \rho_i^{(i)} (\Omega_{\alpha\beta} - \omega_{\alpha\beta}) \) and \( m_{\alpha\beta\gamma} \partial_\gamma \Omega_{\alpha\beta} \). We work to linear order in the forces, yielding

\[ s_A \hat{\theta}_A^{(i)} = \zeta_A (\Delta \mu + X_{A0} u_{\alpha\beta} Q_{\alpha\beta}^{(i)} + X_{A2} m_{\alpha\beta\gamma}^{(i)} \partial_\gamma \Omega_{\alpha\beta} ) \]  
+ \[ X_{A1} \epsilon_{\alpha\beta\gamma} \rho_i^{(i)} (\Omega_{\alpha\beta} - \omega_{\alpha\beta}) + s_A \xi_A(t) \]  
(10)

where \( A \) ranges over 0, 1, 2 and Onsager’s reciprocity principle \[41\] constrains \( X_{AB} \) to be symmetric. In general the phenomenological \( X_{AB} \) and \( \zeta_A \) are periodic functions of the phases \( \theta_A \). In Eqs. \[10\], \( \xi_A(t) \) are phenomenological Gaussian white noise sources.

Finally, to fully specify the system, we provide dynamic equations for the particle positions and directors

\[ \partial_t \mathbf{p}^{(i)} = \mathbf{v} + \zeta_r(t) \]  
(11)

\[ \partial_t \mathbf{p}^{(i)} = \mathbf{h}^{(i)} + \Omega \cdot \mathbf{p}^{(i)} + \nu_1 \mathbf{u} \cdot \mathbf{p}^{(i)} + \nu_2 \mathbf{p}^{(i)} \cdot \mathbf{u} \cdot \mathbf{p}^{(i)} \]  
+ \[ \nu_3 \mathbf{p}^{(i)} \cdot \mathbf{u} \]  
(12)

where the coefficients \( \nu_1 \) to \( \nu_4 \) describe the particles’ tendency to align with shear and rotational flow, respectively, and \( \mathbf{h}^{(i)} \) is the orientational molecular field aligning the \( i \)-th particle with its neighbours \[8\]. The functions \( \zeta_r(t) \) and \( \xi_\Omega(t) \) are phenomenological white noise sources.

We next seek to understand if active particles can synchronize their phases by hydrodynamic interactions. For simplicity, we restrict our analysis to the case where global orientational order is not perturbed, i.e. \( p_\alpha^{(i)} = p_\alpha \), \( Q_{\alpha\beta}^{(i)} = Q_{\alpha\beta} \) and \( m_{\alpha\beta\gamma} = m_{\alpha\beta\gamma} \) are imposed and held.
constant. Such a description will apply on scales smaller than the length \( \sqrt{K/\zeta_0 \Delta \mu} \) beyond which the active Fredericksz instability \[22\ 23\ 17\] sets in, where \( K \) is a typical Frank \[40\] elastic constant.

![FIG. 2: Sketches of active shakers (i), polar rotors (ii) and nematic rotors (iii). These objects exert periodic forces (blue arrows) and torques (black curved arrows) on the fluid. Note that the total torque and force exerted by (i), (ii) and (iii) is zero at each instant in time.](image)

We consider three different cases: (i) a suspension of shakers which generate a periodic force dipole only, i.e. \( \zeta_1 = \zeta(\theta_0) \), \( X_{00} = X_{00}(\theta_0) \) and all other \( X \) and \( \zeta \) are zero, (Fig. 2 (i)); (ii) a suspension of polar rotors which produce an intrinsic force only, i.e. \( \zeta_1 = \zeta(\theta_1) \), \( X_{11} = X_{11}(\theta_1) \) and all other \( X \) and \( \zeta \) are zero, (Fig. 2 (ii)); and (iii) a suspension of nematic rotors which produce a torque dipole only, i.e. \( \zeta_2 = \zeta(\theta_2) \) and \( X_{22} = X_{22}(\theta_2) \) and all other \( X \) and \( \zeta \) are zero, (Fig. 2 (iii)).

We start by investigating case (i). The phase of the \( i \)-th shaker, which we shall simply call \( \theta_i \), obeys

\[
\dot{\theta}_i = \omega(\theta_i) + X(\theta_i)u_{\alpha\beta}Q_{\alpha\beta} + \zeta_0(t),
\]

where \( X = X_{00}/s_0 \) and \( \omega = \zeta_0 \Delta \mu/s_0 \), and the active stress \( \sigma_{act}(\theta_i) = s_0(\theta_i)\dot{\theta}_i Q_{\alpha\beta} \).

To describe synchronization in a continuum theory we apply the treatment of [24] to the distribution in position and phase-angle space, through the Fourier components \( Z_n = \sum_i \exp(n\theta_i)\delta(r - r_{\theta(i)}) \). Here \( Z_0(r) \) is the shaker concentration and \( Z_1(r) \) is the complex Kuramoto order parameter of the suspension. Note that we use \( i \) to denote \( \sqrt{-1} \) to avoid confusion with the index \( i \).

The dynamic equations for \( Z_n \), obtained from (10) and (11), are

\[
(\partial_t + v_r \partial_r - D\Delta + n^2 D_\theta)Z_n = i(n\omega_m + X_m u_{\alpha\beta}Q_{\alpha\beta})Z_{n+m},
\]

where \( D = -\xi_0^2 \) and \( D_\theta = -\xi_0^2 \) are translational and phase-rotational diffusivities, which encode the dephasing effects of the noise terms in (11) and (10). We have expressed the phase-dependent phenomenological coefficients \( \omega \), \( X \) and \( s \) in terms of their Fourier coefficients:

\[
\omega(\theta) = \sum_n \omega_n \exp(n\theta),
\]

and similarly for \( s \) and \( X \).

The active stress produced by a suspension of shakers can then be rewritten as

\[
\sigma_{act} = Q_{\alpha\beta} \sum_{n,m} s_n(\omega_m + X_m u_{\alpha\beta}Q_{\alpha\beta})Z_{n+m} - iQ_{\alpha\beta} \sum_{n} n s_n D_\theta Z_n.
\]

In this framework, we analyse the linear stability of a fully phase-disordered suspension around the quiescent homogeneous steady state \( v = 0 \), \( \Omega_{\alpha\beta} = 0 \), \( Z_0 = 0 \), and all other \( Z_n = 0 \). We truncate Equations (14) or analogous equations for polar and nematic rotors, for \( |\gamma| \geq 2 \) in a manner analogous to [48]. We find that the fully desynchronized state of the suspension has a linear instability. If we impose for simplicity \( s(\theta) = S \cos(\theta) \) and \( \omega(\theta) = \omega_0 \), the stability criterion reads

\[
c_\omega S A^{(i)}(q) \text{Im}(-X_1) \geq 2Dq^2 + D_\theta \left[ 2 + cSA^{(i)}(q)\text{Re}(X_1) \right],
\]

where \( X_1 \) is the first Fourier component of \( X(\theta(i)) \), and the superscript \( i \) = \( 1 \), \( ii \), \( iii \) with

\[
A^{(i)}(q) = \frac{q_\perp^2 q_\parallel^2}{q^2 \left( \eta + \eta' \frac{q^2 \ell^2}{q^2 \ell^2 + 1} \right) - 2cS q_\parallel q_\perp \text{Re}(X_1)},
\]

for shakers, where \( q^2 = q_\perp^2 + q_\parallel^2 \) with \( q_\perp \) denoting the \( q \) directions perpendicular and parallel to the polarity vector \( \mathbf{p} \), respectively;

\[
A^{(ii)}(q) = \frac{q_\perp^2 q_\parallel^2}{q^2 \left( \eta + \eta' \frac{q^2 \ell^2}{q^2 \ell^2 + 1} \right) - 2cS q_\parallel q_\perp \text{Re}(X_1)},
\]

for polar rotors, and

\[
A^{(iii)}(q) = \frac{q^2}{q^2 (q^2 \ell^2 + 1) \left( \eta + \eta' \frac{q^2 \ell^2}{q^2 \ell^2 + 1} \right) - 2cS q_\parallel q_\perp \text{Re}(X_1)}
\]

for nematic rotors, respectively.
Thus, at large enough driving frequency $\omega_0$ and particle density $c$, the fully asynchronous states of suspensions of all three types of particles is linearly unstable, provided $\text{Im}(-X_1) > 0$, for the choice $s(\theta) = S \cos \theta$, the case treated here. In general the synchronization criterion involves phase-dependent stresses and angular-momentum fluxes $s_A$, and the phase relation between the forcing $\omega$ and the feedback $X$ from the fluid. The growth rate of the instability is nonzero for $q \ell \to 0$ for (i) and (iii) and $O(q^2 \ell^2)$ for case (ii), where $\ell = \sqrt{\kappa/(2\eta)}$ is an inherent length scale which is in general microscopic \cite{11, 22}. Fig. 3 shows the intricate dependence of the synchronization threshold value of $c\omega_0$ on the direction of the perturbation wave vector.

We next investigate the linear stability of the homogeneous phase-coherent suspension. We define the coarse-grained phase $\Theta(r) = (1/N) \sum_i X^{(i)}(r - r^{(i)})$ which, in the case (i) of active shakers, obeys

$$\partial_t \Theta + v_0 \partial_r \Theta = D \Delta \Theta + \omega(\Theta) + X(\Theta)u_{\alpha\beta} Q_{\alpha\beta},$$

with active stress

$$\dot{\sigma}_{\alpha\beta}^{act} = cs(\Theta) Q_{\alpha\beta} \dot{\Theta},$$

where $c$ is the particle concentration field.

We perturb the homogeneously synchronized quiescent state $\Omega_{\alpha\beta} = 0$ and $\mathbf{v} = 0, \Theta = \Theta_0(t), c = \text{constant}$, and ask whether the flows set up by the resulting stresses drive the system further from the unperturbed state. As the reference state is time-periodic, our result is expressed in cycle-averaged terms. We find that the system is linearly unstable if

$$A^{(i)}(q) c \overline{X|s_X|} > D q^2,$$

where the bar denotes averaging over time and the prime denotes a derivative with respect to the argument $\Theta$. Thus, both asynchronous states and global synchrony are unstable at strong enough activity, suggesting that the likely long-time behavior consists of complex spatially patterned states. Stability criteria completely analogous to \cite{22} can also be derived for (ii) and (iii). In Fig. 4 we plot phase diagram for solutions for shakers and nematic rotors. Independent of the precise numerical values of parameters, the topology of the phase diagram is a robust testable result. Further, we predict that the zone where only the phase-incoherent state is stable shrinks with decreasing $D_\theta$. Interestingly, the instability condition Eq. \cite{22} is always satisfied when \cite{17} holds. Thus a spatially unbounded oriented active fluid that synchronizes will do so in a spatially inhomogeneous manner.

In summary, we have developed a theory for bulk active fluids that accounts for the underlying periodicity of active processes and displays an instability towards phase synchronization via a purely hydrodynamic mechanism. We have mapped out the wave vector dependence of the onset of this synchronization instability. \cite{Fig. 3}. For shakers and nematic rotors, the growth rate of the instability is non-vanishing for wavenumber $q \to 0$, presumably because of long-range hydrodynamic interactions carried by the velocity field. In contrast the growth rate of the synchronization instability of polar rotors vanishes as $q \to 0$, suggesting that the more rapidly decaying spin rotation rate is responsible. We then showed that the globally synchronized quiescent state is also generically unstable (Fig.4). Thus spontaneously broken time-translation invariance ultimately leads to spatial pattern formation and spontaneous flow in bulk active fluids, via gradients in the Kuramoto phase, even while global nematic order is maintained. The topology of the resulting phase diagram (Fig.4) is a robust experimentally testable result. We also find that the width of the region in which the fully phase-incoherent state alone is stable decreases as the phase-rotational diffusivity decreases. These ideas should be testable in a variety of systems \cite{35, 38, 39}.

In order to focus on general principles we have worked with spatially unbounded ordered systems, but generalization to thin-film or wall-bounded geometries \cite{12, 13, 39, 49, 50} is straightforward. Moreover, a complete treatment must allow distortions of the state of orientational order. Such a study would bring out the interplay between the spontaneous flow instabilities \cite{5} of asynchronous active liquid crystals and the new mechanisms outlined in this work. Finally, large-scale numerical studies will ultimately be required in order to go beyond the linear stability analysis presented here.

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