Pattern Formation in Chemically Interacting Active Rotors with Self-Propulsion

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We demonstrate that active rotations in chemically signalling particles, such as autochemotactic E. coli close to walls, create a route for pattern formation based on a nonlinear yet deterministic instability mechanism. For slow rotations, we find a transient persistence of the uniform state, followed by a sudden formation of clusters contingent on locking of the average propulsion direction by chemotaxis. These clusters coarsen, which results in phase separation into a dense and a dilute region. Faster rotations arrest phase separation leading to a global travelling wave of rotors synchronized to a positive feedback (Fig. 1). Such a chemical ‘signalling’ can induce spontaneous aggregation through a positive feedback (Fig. 1a): an initial positive chemical density fluctuation attracts microorganisms, which produce further chemicals; this amplifies the initial fluctuation supporting the recruitment of further microorganisms, which eventually condense into a macroscopic cluster coexisting with a dilute microbial bath (Fig. 1b). Quantified by the Keller-Segel (KS) model in the 1970s 5, 6, this signalling-route to phase separation now serves as a prototypical example for self-organization in nonequilibrium 7. It is crucial for the life cycle of Dicty 4; it is also one possible mechanism to explain pattern formation in bacterial colonies within agar 1, 2, alongside an alternative explanation based on arrested motility induced phase separation 8.

Currently, the KS instability is attracting renewed attention in active synthetic Janus colloids. These particles catalyse reactions within a chemical bath, yielding gradients which drive self-propulsion via diffusiophoresis or a similar mechanism. Importantly, other colloids may feel rotational torques caused by the same long-ranged gradients and respond to them by adapting their swimming direction, thereby providing a synthetic analog of chemotactic signalling 9–11. The same KS equations can therefore be transferred from the microbiological world to phoretic colloids 10, 12, 14. Here, either the KS instability, or instabilities based on chemorepulsion 15 may help explain the still elusive dynamic clustering observed in experiments 10, 16, 17.

In all these cases chemotactic instability relies on the ability of weak chemical fluctuations around the uniform density to align microswimmers up (or down) chemical gradients. However, under many circumstances microswimmers vary their swimming direction autonomously of chemical cues: this occurs, e.g., for bacteria swimming clockwise close to a glass wall 18, or anti-clockwise near an oil-water interface 19 (see Electronic Supplementary Information (ESI) for a discussion on parameters). Synthetic examples of active signalling rotors with self-propulsion (sometimes called ‘circle swimmers’) may be realised with L-shaped phoretic swimmers 20, 21, or with active particles with dipole moments 16, 22, 23 which will track the rotation of applied external fields 16, 24 and can interact via self-produced phoretic fields 16.

We should expect that, close to uniform states, finite active torques as caused by intrinsic rotations will generally outcompete chemotactic torques which are proportional to the chemical gradient, when it comes to determining swimming direction: hence, they might generically lead to a breakdown of the linear KS instability. Does this rule out the possibility of phase separation and pattern formation in signalling rotors? This could have profound consequences for systems like thin (bio)films of chemotactic bacteria 25 or actuated phoretic particles.

Here, we propose a generic model to study active signalling rotors. As a key result we find that even weak active rotations suppress the linear KS instability. However, we identify an instability mechanism which is distinct from the KS mechanism and creates a nonlinear, but deterministic, route to pattern formation.

For small rotation frequency, this route generates macroscopic phase separation: a fraction of the rotors condenses into a dense phase, separated from a dilute gas by a quasi-stationary interface where chemotaxis suppresses rotations and ‘locks’ the average swimming direction onto the upward density gradient. Away from the interfaces, rotations persist in both phases and lead to dynamic patterns, such as spots or moving stripes and spirals (Fig. 2a, i). This combination of phase separation with pattern formation within both phases represents a novel type of hierarchical structure formation. When increasing the rotation frequency above a certain threshold, strikingly, the growing clusters do not coarsen any more but form a global pattern of travelling stripes with...
self-limiting size, suggesting that active rotations can be used to control coarsening in suspensions of self-propelled particles (Fig. 2).

We describe active particles which self-propel with velocity $v$ and rotate with frequencies $\omega_i > 0$ at a coarse grained level in two-dimensions. A smooth velocity $v$ of particles (Fig. 2l) is used to control coarsening in suspensions of self-propelling self-limiting size, suggesting that active rotations can be used to control coarsening in suspensions of self-propelled particles (Fig. 2).

Thus, in absence of signalling, $p$ rotates with a collective frequency $\omega$. Generally however, our rotors produce signalling molecules with a local rate $k_0\rho$; hence $p$ also responds, via chemotaxis, to gradients of the resulting chemical field $c(x, t)$, which decays with a rate $k_d$.

This yields a competition of intrinsic active rotations and chemotactic alignment which determines the behaviour of the signalling rotors at large scales. Allowing for finite diffusion of the chemical and colloidal density fields with coefficients $D_c$ and $D_\rho$, we describe signalling rotors phenomenologically by:

$$
\dot{\rho} = -v_0 \nabla \cdot (\rho p) + D_\rho \nabla^2 \rho + K \nabla^2 \rho^3 \quad (1)
$$

$$
\dot{\phi} = \omega + \beta \nabla c; \quad p = (\cos \phi, \sin \phi)^T \quad (2)
$$

$$
\dot{c} = k_0 \rho - k_d c + D_c \nabla^2 c + \epsilon (c_0 - c)^3. \quad (3)
$$

Here, we used the notation $a \times b = a_1 b_2 - a_2 b_1$ and introduced a chemotactic coupling strength $\beta$, as well as an isotropic short range repulsion among colloids of strength $K > 0$ and a cubic reaction term with rate coefficient $\epsilon$ leading to the saturation of any unstable mode [20]. This phenomenological model can be justified microscopically and is studied here as a minimal description of chemotactic rotors (see ESI †), which focuses on the competition between chemotaxis and active rotations. Its parameter space can be reduced to five dimensions including the (initial) density: only three of these enter the linear dynamics close to the uniform state. We choose experimentally sensible values for these parameters (ESI †) and introduce the dimensionless number $\Delta = \omega v_0/(k_0 k_d \beta)$ as a measure of the relative importance of active rotations and chemotaxis.

We now explore the competition of active rotations and chemotaxis by solving Eqs. (1-3) for different $\Delta$ on a square lattice with $L_x \times L_y$ grid points and periodic boundary conditions using finite difference methods. As an initial state, we choose a small and random perturbation of the spatially uniform and coherently rotating state $(\rho, \phi, c) = (\rho_0, \omega t, (k_0/k_d) \rho_0)$ which solves Eqs. (1-3). In absence of rotations ($\Delta = 0$) we observe clusters growing out of the uniform state (Fig. 2b), which colocalise with chemical density maxima and coarsen at long times (Fig. 2b), yielding one dense macroscopic cluster coexisting with a dilute gas (Fig. 2f). Here, instability of the uniform state is expected due to the positive feedback loop of particle aggregation and chemical production we discussed in the introduction and in [15].

For $\Delta = 5 \times 10^{-3}$ this picture changes dramatically. Now the uniform state persists for a certain duration (see Videos 1,2 in ESI): we call this the initial lag regime. Then, almost suddenly, fluctuations ‘awake’ and grow to create clusters (Fig. 2l). These clusters coarsen into a relatively dense phase separated from a dilute rotor gas (Fig. 2c, d) by a slowly-moving interface where rotations are suppressed and particles swim, on average, up the chemical gradient (Video 2). Away from the interface, all colloids perform a ‘stop-and-go’ rotation, which is associated with dynamic short-lived density dips and peaks (white spots and red dips in yellow region in Figs. 2e,f). Choosing stronger rotations ($\Delta = 0.024$) we observe a similar suppression of rotations at the interfaces, but within both phases stripe and spiral patterns form (Fig. 2k-i and Video 2).

For $\Delta = 0.044$ complexity suddenly breaks down: after a long initial lag regime and an intermediate regime where short-lived dynamic clusters continuously emerge and decay, we find a travelling wave made of straight par-

![Fig. 1. Cartoons showing the basic setup and the locking instability (see text).](image-url)
FIG. 2. (a-1): Snapshots of density field $\rho$ for different free rotation frequencies $\omega$ as a function of $x,y$ at different times (increasing from left to right). Values for $\omega$ and $\Delta$ are: $\omega = 0; \Delta = 0$ (a-c); $0.25; 5 \times 10^{-3}$ (d-f); $1.2; 0.024$ (g-i) and $2.2; 0.044$ (j-l). (m-t): snapshots of $\beta|\nabla c| - \omega$ corresponding to the density fields of columns 2 and 3 (m belongs to b; n to c etc). Positive values represent locking, negative values rotations. Parameters: $\beta = 5.0, \rho_0 = 5.0, v_0 = 0.5, k_d = k_0 = 1, \epsilon = 10, D_\rho = 0.417; K = 0.025; D_c = 1; \rho_0 = (k_0/k_d)\rho_0; L_x \times L_y = 50 \times 50$ in the first and third row, $40 \times 40$ in the second row and $80 \times 32$ in the fourth row; $0.32\sqrt{D_\rho/k_d}$ is the distance between adjacent gridpoints.

allel stripes (Fig. 2k,l, Video 4) with self-limiting wavelength. This sudden emergence of a length scale upon increasing $\Delta$ allows control of coarsening via active rotations. The length scale of the stripe pattern which decreases as $\omega$ increases and thus can be also controlled.

Rotation-locking transition – To understand the origin of cluster growth for $\Delta > 0$ and the related suppression of rotations at the cluster interfaces, we recast Eq. (2) as

$$\dot{\phi} = \omega + \beta|\nabla c| \sin (\phi + \delta); \quad \delta = \arg(-\partial_x c + i\partial_y c) \quad (4)$$

Assuming a time-independent chemical field this can be read, locally, as the Adler equation of synchronization theory which features a transition from (quasi-)periodic rotations ($\beta|\nabla c| < \omega$) to phase locking ($\beta|\nabla c| > \omega$), see ESI † and [27–29]. Although, for our signalling rotors $\nabla c$, hence also $\delta$, evolve in time, in the parameter regime relevant to our numerical simulations, $p$ responds fast to changes in $c$, hence we expect that the locking transition should still apply. Our simulations confirm this expectation and clearly show a transition from ‘active’ rotations to locking at $\beta|\nabla c| \approx \omega$ (Fig. 2o-r), which explains the suppression of rotations at, and only at, the interface in Fig. 2f.h.i where chemical gradients are strong enough to generate locking. This locking induces a permanent self-advective flux of particles from the dilute phase to the dense phase balancing diffusion in the opposite direction and thus stabilizes dense rotor clusters.

Locking instability – Since the locking mechanism stabilizes clusters, we expect an instability mechanism allowing for the growth of these clusters out of the uniform state. We now resolve this in detail. Working in the adiabatic limit where $p$ responds fast to changes of $c$, Eqs. (1-3) simplify significantly. If $\omega = 0$, they resemble the Keller-Segel model (ESI †), which leads to phase separation through a long wavelength linear instability – essentially this is the positive feedback described in the Introduction. In sharp contrast, we show in the ESI † that active rotations generically suppress linear instability of the uniform phase in Eqs. (1-3). Physically, this is in line with the intuitive expectation that small chemical gradients caused by fluctuations around the uniform state are too weak to fix the orientations of active rotors and cannot promote a linear instability. As a result, the KS instability is ineffective for active signalling rotors.

This raises the question why we could still observe cluster growth in our simulations. The answer is that chemotaxis can abduct the rotors into the nonlinear regime before they complete a full rotation, and this in turn can lead to a nonlinear instability. To understand the underlying physical mechanism, let us reconsider our coherently rotating uniform initial state, where all rotors are in phase (Fig. 1l). While $\rho,c$ remain approximately uniform in the course of the early-stage dynamics, weak fluctuations of $c$ continuously dephase the orientation field $p$ due
to chemotaxis (Fig. [1]). Once the rotors are sufficiently out of phase, they can form, temporarily, aster-like ‘seed’ configurations (Fig. [1]), where all particles in the vicinity of a positive fluctuation of $c$ swim up the chemical gradient. This configuration promotes, temporarily, a growth of the fluctuation via the standard KS feedback loop. If this growth generates a chemical gradient surpassing the locking-threshold $\beta |\nabla c| > \omega$ before the seed decays, then $p$ remains locked in its vicinity – leading to a stable cluster as just discussed \[33\]. Hence, we call our nonlinear instability the ‘locking instability’. Whether the locking instability is effective or not in practice is a matter of competing timescales, between the instantaneous growth rate of chemical gradients and $\omega$ – the former needs to dominate to create locking. Video 5 in the ESI \[9\] shows the early stage dynamics of $c$ and $p$, reflecting that $c$ and its gradients can indeed grow beyond the locking threshold on timescales where $p$ rotates only slightly.

**Breakdown of locking** – Locking gets increasingly difficult as $\Delta$ increases and eventually breaks down, quite suddenly, at $\Delta \approx 0.027$ (for parameters as in Fig. [2]). Beyond this, stable clusters are no longer possible. Specifically, as $\Delta$ increases (i) steeper interfaces are needed to satisfy the locking criterion and (ii) active rotations increasingly dominate the equilibrium between chemotaxis and rotations which leads to a steering of the locked $p$ away from its ‘ideal’ orientation up the gradient. This in turn reduces the advective flux up the density gradient leading to shallower interfaces (see ESI \[9\] and Videos 2,3). Unlike for linear (supercritical) instabilities an instability criterion is ill-defined here, and the breakdown of locking depends on an implicit balance between steepness of interfaces and locking orientation of $p$ far away from the uniform state. Likewise, when comparing the locking instability to nucleation in systems approaching thermal equilibrium, which is also nonlinear and requires a seed configuration, we find again important differences. Crucially, for example, our clusters require steep interfaces to survive, and a dense cluster with shallow interfaces would quickly decay; in contrast, for near-equilibrium systems, once a dense seed has nucleated it will grow, however shallow its interfaces may be.

**Active rotations arrest coarsening** – For large $\Delta$, where rotations are too strong to allow locking, we observed a long period where small and short lived clusters dynamically emerge and decay on timescales $\sim 2\pi/\omega$. These clusters emerge from seeds which promote a growth of $\rho$ and $c$ which is too slow to surpass the locking threshold and hence slows down the rotation of $p$ only temporarily. Strikingly, however, this dynamic clustering does not proceed forever, and eventually some clusters merge (due to diffusion and short ranged repulsions which penalize large interfaces) and synchronize their rotations and translations to form coherent waves (see Video 4 in ESI). Specifically, within clusters, the relative phases (frequencies) of adjacent rotors change continuously until the system eventually reaches a steady state. Here such a steady state is possible in the form of a stripy pattern (Fig. [2]), which moves precisely by one spatial period during one rotational cycle of $p$. In a frame comoving with the pattern, $p$ is then fixed (Fig. [2b]) and the pattern, a periodic non-uniform state, is an attractor for the dynamics of the system. Hence, merging and approaching this steady state represents a second route allowing short lived clusters to avoid decay.

This process of forming short lived clusters which prevent their decay by merging and moving into a collective direction (spontaneous symmetry breaking) constitutes a second nonlinear instability mechanism of the uniform state that is independent of the locking mechanism. In contrast with the latter, this mechanism works at all frequencies, but is comparatively slow since it involves the coordination of several clusters. Hence this second route to structure formation in signalling rotors is particularly relevant in parameter regimes where the locking mechanism is ineffective, i.e., for large $\omega$, but applies similarly to the moving stripes formed within the dense and the dilute phase in Fig. [2].

As its most remarkable feature and contrasting the locking instability, this route to pattern formation introduces a length scale in steady state. Within a large region of parameter space, this length scale increases approximately as $v_0/\omega$. Hence, for synthetic rotors this length scale can be tuned via the frequency of an applied rotating field. We emphasize that this length scale is determined purely dynamically and cannot be calculated via the standard tools for linear (supercritical) instabilities like amplitude equations \[30\].

**Non-identical frequencies** – We finally discuss signalling rotors with non-identical frequencies (e.g., a population of bacteria close to a wall). In absence of alignment effects these rotors quickly dephase and destroy the local polar order. However, hydrodynamic \[31\] or steric alignment interactions can ‘synchronize’ individual rotations. Assuming, e.g., short ranged polar alignment interactions with alignment rate $(g/\rho^2) \sin(\theta_j - \theta_i)$ between particles $i, j$ (polar coordinates) if their distance $|\mathbf{r}_i - \mathbf{r}_j|$ is smaller than a cutoff radius $r_c$ and ignoring chemo-
taxis (which supports alignment), the local orientation of a rotor follows
\[ \dot{\theta}_i = \omega_i + \sqrt{2D_r \xi_i(t)} + (g/r_c^2) \sum \sin(\theta_j - \theta_i) \] (5)

Here we sum over all \( N_c \approx r_c^2 \rho(r_c) \) particles with distance less than \( r_c \) to the \( i \)th particle and \( \xi_i(t) \) represents unit-variance Gaussian white noise. Remarkably, for dense systems, Eq. (5) can be identified (locally) as the Kuramoto model with noise [32] which shows a phase transition from the incoherent uniform phase to frequency locking if \( g \rho_0 \) is a few times larger than \( D_r \) and the standard deviation \( \Delta_\omega \) of the distribution function of the individual frequencies [32]. Thus, within an ensemble of non-identical rotors, a strong enough alignment interaction causes a macroscopic fraction of them to rotate in phase with an average frequency, so that our analysis for identical rotors still largely applies. The main caveat is that synchronization of the individual rotations takes place locally, so the average rotation may vary in space and time. We thus performed simulations of Eqs. (13) with additional frequency noise \( \omega_\xi(x, t) \) in Eq. (2): the results resemble those in Fig. 2 as long as \( \Delta_\omega < \omega \) and the correlation time of \( \omega_\xi \) is short compared to \( 1/\omega \). Note that the ESI† material provides an alternative and direct microscopic justification of Eqs. (13) for rotors with non-identical frequencies.

In conclusion, although even weak active rotations linearly stabilise the uniform phase in ensembles of auto-chemotactic particles, they generate a nonlinear route to structure formation. This route creates novel patterns including hierarchically organized states combining phase separation and pattern formation. It also allows for features which would be impossible to achieve in linear instability scenarios, such as a delayed onset of patterning whose lag time can be programmed via the initial conditions. More generally, we showed that rotations provide a versatile new tool to design self-assembly and collective behaviour of active matter, for example to control coarsening.

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Note that in contrast to asters, 'anti-asters' with $p$ pointing outwards from a center where $\rho, c$ show minima, suffer from the fact that both $\rho, c$ cannot decay below zero. This limits the maximal possible steepness of chemical gradients to values which are typically insufficient to generate locking.

E. M. Izhikevich, *Dynamical Systems in Neuroscience* (MIT press, Cambridge, Massachusetts, 2007).