

Electronic Supplementary Information to Pattern Formation in Chemically Interacting Active Rotors

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A. Microscopic approach to phenomenological model

1. Rotors with identical frequencies

Here, we derive coarse grained equations of motion for the rotor density and orientation fields. Since we are mainly interested in the competition between active rotations and chemotaxis - and in the corresponding physical mechanism allowing for structure formation - we use a variety of approximations to achieve a ‘minimal’ rather than a rigorous coarse grained description of active rotors which focuses on this competition (and hence only covers a subset of possible phenomena). This procedure leads to Eqs. (1-3) considered in the main text.

Consider an ensemble of N ‘signalling rotors’, which self-propel with constant velocity v_0 along the directions $\mathbf{p}_i = (\cos \theta_i, \sin \theta_i)$. In the uniform state, these directions rotate actively with natural frequencies Ω_i , but generally also respond to chemical gradients, where β_R denotes the ‘chemotactic’ coupling strength. We further assume (steric) alignment interactions between different rotors which are sufficiently short ranged to allow us to replace their spatial dependence by a pseudopotential (a zero ranged ‘ δ ’-interaction). Denoting the rotational diffusion constant by D_r , and by $\xi(t)$ a Gaussian white noise with unit variance, we describe a single chemotactic rotor in 2D via the following Langevin equations

$$\dot{\mathbf{r}}_i = v_0 \mathbf{p}_i \quad (1)$$

$$\dot{\theta}_i = \Omega_i + \beta_R \mathbf{p}_i \times \nabla c + G \sum_{j=1}^N \sin(\theta_i - \theta_j) \delta(\mathbf{r}_i - \mathbf{r}_j) + \sqrt{2D_r} \xi_i(t) \quad (2)$$

where we used the notation $\mathbf{a} \times \mathbf{b} = a_1 b_2 - a_2 b_1$ and $c(\mathbf{x}, t)$ is the chemical field produced by the ensemble of ‘signalling’ rotors with rate k_0 . This field evolves as

$$\dot{c} = k_0 \sum_{i=1}^N \delta(\mathbf{r} - \mathbf{r}_i) - k_d c + D_c \nabla^2 c + \epsilon(c - c_0)^3 \quad (3)$$

where k_d is the decay rate. Here, D_c is the chemical diffusion constant and the term proportional to ϵ prevents unlimited growth of c in case of linear instability.

We now use Itô’s Lemma and follow [1] to derive coarse grained equations of motion for the combined probability density $f_i(\mathbf{r}, \theta) = \delta(\mathbf{r} - \mathbf{r}_i) \delta(\theta - \theta_i)$. For rotors with identical frequencies $\Omega_i \rightarrow \Omega$ we find for $f(\mathbf{r}, \theta) = \sum_{i=1}^N f_i(\mathbf{r}, \theta)$

$$\dot{f} = -v_0 \mathbf{p} \cdot \nabla f - G \partial_\theta \int d\mathbf{r}' d\theta' f(\mathbf{r}', \theta') F(\theta - \theta', \mathbf{r} - \mathbf{r}') f(\mathbf{r}, \theta) + D_r \partial_\theta^2 f - \beta_R |\nabla c| \partial_\theta [f \sin(\theta + \delta)] - \partial_\theta \Omega f - \partial_\theta \sqrt{2D_r} f \xi \quad (4)$$

where $\xi = \xi(\mathbf{r}, \theta, t)$ represents unit-variance Gaussian white noise and $\delta = \text{atan2}(\partial_y c, -\partial_x c)$, with $\text{atan2}(y, x)$ being the (principle value of) the argument function $\arg(x + iy)$ and $F(\theta - \theta', \mathbf{r} - \mathbf{r}') = \sin(\theta - \theta') \delta(\mathbf{r} - \mathbf{r}')$. The term $-\partial_\theta \sqrt{2D_r} f \xi$ describes multiplicative noise, which we neglect in the following because we are interested in the mean field phenomenology.

From here, we follow [2] and expand f in a Fourier series $f(\mathbf{r}, \theta) = \sum_{k=-\infty}^{\infty} f_k(\mathbf{r}) e^{-ik\theta}$ with $f_k(\mathbf{r}) = \int f(\mathbf{r}, \theta) e^{ik\theta} d\theta$ and identify $f_0 \rightarrow \rho(\mathbf{r}, t)$ and $(\text{Re} f_1, \text{Im} f_1) \rightarrow \mathbf{w}$. [3] Straightforward algebra leads to the following equation for the Fourier coefficients:

$$\dot{f}_k = -\frac{v_0}{2} [(\partial_x - i\partial_y) f_{k+1} + (\partial_x + i\partial_y) f_{k-1}] - k^2 D_r f_k + \frac{iGk}{2\pi} \sum_m f_{k-m} F_{-m} f_m + \frac{\beta_R |\nabla c| k}{2} (f_{k+1} e^{i\delta} - f_{k-1} e^{-i\delta}) + ik\Omega f_k \quad (5)$$

For $k = 0$ we quickly find $\dot{\rho} = -v_0 \nabla \cdot \mathbf{w}$. To achieve a closed equation for f_1 we neglect f_k with $k \geq 3$ and assume that f_2 is fast, i.e. we set $\dot{f}_2 \rightarrow 0$ (compare [2, 4]). This yields a closed equation of motion for f_1 and hence for $\mathbf{w}$. This latter equation could in principle be solved numerically, but is rather involved and hardly allows us to analyse

the interplay between chemotaxis and active rotations. To highlight this interplay within a ‘minimal model’ we apply a second layer of approximations and directly neglect all contributions which are both nonlinear and involve gradient terms [5]. This procedure leads to

$$\dot{\rho} = -v_0 \nabla \cdot \mathbf{w} \quad (6)$$

$$\dot{\mathbf{w}} = \left(\frac{G\rho}{2} - D_r \right) \mathbf{w} + \Omega \mathbf{w}_\perp - \frac{v_0}{2} \nabla \rho + \frac{\beta_R \rho}{2} \nabla c + \frac{v_0^2 D_r}{b} \nabla^2 \mathbf{w} + \left(\frac{v_0^2 \Omega}{2b} \right) \nabla^2 \mathbf{w}_\perp - \left(\frac{2G^2 D_r}{b} \right) |\mathbf{w}|^2 \mathbf{w} - \frac{G^2 \Omega}{b} |\mathbf{w}|^2 \mathbf{w}_\perp \quad (7)$$

$$\dot{c} = k_0 \rho - k_d c + D_c \nabla^2 c + \epsilon (c - c_0)^3 \quad (8)$$

where $\mathbf{w}_\perp \equiv (-w_y, w_x)$ and $b = 16D_r^2 + 4\Omega^2$. Two solutions of these equations are: (i) the uniform unpolarized state $(\rho, \mathbf{w}, c) = (\rho_0, \mathbf{0}, \rho_0 k_0/k_d)$, and (ii) a polarized and coherently rotating state with uniform density $(\rho, w, \phi, c) = (\rho_0, w^*, \Omega^* t, k_0 \rho_0/k_d)$ with $w^* = \sqrt{[G\rho_0 - 2D_r](\Omega^2/(G^2 D_r) + 4D_r/G^2)}$, $\Omega^* = \Omega(3/2 - G\rho_0/(4D_r))$. Here, ϕ is defined via $\mathbf{w} = w\mathbf{p}$ with $\mathbf{p} = (\cos \phi, \sin \phi)^T$ representing the average (collective) self-propulsion direction. If $G\rho_0 < 2D_r$ the unpolarized state is stable. It becomes unstable in favour of the coherently rotating state when alignment interactions are strong enough to suppress dephasing by rotational noise ($G\rho > 2D_r$). At the onset of polarization this state rotates with a frequency Ω but slows down as more and more particles align.

In the main text, we focus on the regime of sufficiently strong alignment interactions and hence consider this coherently rotating state as the relevant uniform state whose dynamics we explore in presence of chemotaxis. Also, if self-propulsion is not too strong, the term $(-v_0/2)\nabla \rho$ mainly reduces the chemotactic coupling in our simulations where we typically have $c \sim \rho$ and $\beta_R \rho_0 \gg v_0$. Hence we also omit this term for simplicity as well as ∇^2 terms in Eq. (7), which are not important at long wavelength or for almost uniform $\mathbf{w}$. We now define the polarization $\mathbf{P}$ which measures the degree of local alignment (per particle), as usual, via $\mathbf{w} = \rho \mathbf{P} = \rho |\mathbf{P}| \mathbf{p}$. Since we are mainly interested in the competition of the chemical alignment and active rotations, we do not describe spatial modulations of the ‘average’ polarization but assume it as constant. Although, only choices of $|\mathbf{P}| \ll 1$ are strictly consistent with the above approximation $f_{k>3} \rightarrow 0$ [2], we choose $|\mathbf{P}| = 1$ for simplicity, since our results do not depend much on the exact value of $|\mathbf{P}|$ as our simulations reveal.

Assuming also $\omega := \Omega - \frac{G\Omega}{b} w^2 \approx \Omega - \frac{G\Omega}{b} \rho_0^2$ in Eq. (7) and defining $\beta := \frac{\beta_R}{2}$, after projecting Eq. (7) onto $\hat{\mathbf{p}}_\perp = (-\sin \phi, \cos \phi)$ our minimal description of chemotactic rotors reads

$$\dot{\rho} = -v_0 \nabla \cdot (\rho \mathbf{p}) + D_\rho \nabla^2 \rho + K \nabla^2 \rho^3 \quad (9)$$

$$\dot{\phi} = \omega + \beta \mathbf{p} \times \nabla c \quad (10)$$

$$\dot{c} = k_0 \rho - k_d c + D_c \nabla^2 c + \epsilon (c - c_0)^3 \quad (11)$$

In Eq. (9), we added a phenomenological term describing isotropic short ranged repulsions among colloids ($K \nabla^2 \rho^3$), whose main effect is to prevent strong gradients on too small scales in our simulations. While we chose here a cubic term for convenience (retaining symmetry under $\rho \rightarrow -\rho$), replacing this by a quadratic term, $\nabla^2 \rho^2$ leads, according to our simulations, to an almost identical phenomenology when modifying the coefficient K appropriately. Given the complexity of the system under consideration, our description in Eqs. (9-11) is far from complete, but it highlights the competition between chemotactic alignment and active rotations, allowing us to focus on the physical mechanism underlying structure formation in signalling rotors. Our approach can be straightforwardly extended to derive a more precise but rather complex coarse grained description of (chemotactic) active rotors.

2. Generalization to non-identical rotation frequencies

Here, we generalize the above approach to rotors with non-identical frequencies Ω_i , i.e. we replace $\Omega \rightarrow \Omega_i$ in Eq. (2) and ask to which extent this alters Eqs. (9-11). Accordingly, this paragraph can be seen as an alternative to the paragraph ‘non-identical frequencies’ in the main text.

The basic idea is to replace $f_i \Omega_i$ by its mean plus a typical fluctuation

$$\Omega_i f_i \longrightarrow \bar{\Omega} f + \sqrt{\Delta_\Omega f} \eta \quad (12)$$

where η describes Gaussian random numbers with zero mean and unit variance $\langle \eta(\mathbf{r}, \theta, t) \eta(\mathbf{r}', \theta', t) \rangle = \delta(\mathbf{r} - \mathbf{r}') \delta(\theta - \theta')$ and $\bar{\Omega}$; Δ_Ω are defined by

$$\langle \Omega_i \Omega_j \rangle = \bar{\Omega}^2 + \Delta_\Omega \delta_{ij} \quad (13)$$

This replacement of course changes, in general, the dynamics of f_i but not the statistical properties of a large rotor ensemble, since the distributions $\{\Omega_i f_i\}$ and $\{\bar{\Omega} f + \sqrt{\Delta_\Omega f} \eta\}$ have identical statistical properties. (This statement

holds true at each point in time, independent of time correlations of η).

Physically, we should understand Eq. (12) as a local replacement and interpret the above averages as mesoscopic ones over all particles within the interaction range around a given point $\mathbf{x}$ rather than a global average over the whole rotor ensemble. Accordingly, we allow $\bar{\Omega}$ and η to fluctuate in space and time. We now define a field $\omega(\mathbf{x}, t)$ representing the deviation from the global (time-independent) average rotation frequency $\bar{\Omega}_0 = \sum_{i=1}^N \Omega_i/N$.

In the following we do not worry about the specific form of $\omega(\mathbf{x}, t)$ but assume it to be random, with a correlation time on the order of the ‘mixing time’, i.e. the time after which a given set of rotors in one ‘interaction domain’ is replaced by another one. In non-synchronized states, we expect that the mixing time is on the order of the time a particle needs to traverse a distance given by the range of the alignment interactions. As this timescale is, for the assumed short ranged alignment interactions, short compared to all other relevant timescales in the system ($1/k_d, 1/k_0, 1/\omega$) we allow it to tend to zero for simplicity. Hence we assume ω , and analogously also η , to represent spatiotemporal white noise. Conversely, in the synchronized regime, rotors can move together for some time and hence we expect substantial time-correlations. As these correlations should become relevant only after the onset of synchronization we do not need to care about them since our aim is to understand where synchronization sets in.

We now expand $\sqrt{f}$ for modest deviations from isotropy as follows

$$\sqrt{f} = \sqrt{\sum_{k=-\infty}^{\infty} f_k \exp[-ik\theta]} \approx \sqrt{f_0}/2 + \frac{1}{2\sqrt{f_0}} \sum_{k=-\infty}^{\infty} f_k e^{-ik\theta}. \quad (14)$$

Using this approximation, the only modifications of our mean field results Eqs. (6-11) due to non-identical rotor frequencies correspond to replacing

$$\bar{\Omega} \rightarrow \bar{\Omega}_0 + \omega(\mathbf{x}, t) \quad (15)$$

$$D_r \rightarrow D_r + \sqrt{\frac{\Delta\Omega}{4\rho_0}} \quad (16)$$

in these equations. Consequently, for non-identical rotors the unpolarized state becomes unstable for $G\rho_0 > 2D_r + \sqrt{\frac{\Delta\Omega}{4\rho_0}}$. This is very similar to the result of the Kuramoto model (main text) which assumes a Lorentzian distribution for the frequencies, and justifies the assumption of locally coherent rotations underlying model (9-11).

B. Nondimensionalisation and relation to experimental parameters

The parameter space of Eqs. (9-11) can be reduced to four dimensions (plus an effective density which is fixed by the initial state) by introducing the dimensionless quantities $\tilde{x} = \sqrt{k_d/D_\rho}x$, $\tilde{t} = k_d t$. Defining $\tilde{\rho} = \rho k_0 \beta / (k_d v_0)$, $\tilde{c} = c\beta/v_0$ and $\tilde{\mathbf{p}} = v_0 \mathbf{p} / \sqrt{D_\rho k_d}$ we obtain (now omitting tildes)

$$\dot{\rho} = -\nabla \cdot (\rho \mathbf{p}) + \nabla^2 \rho + \kappa \nabla^2 \rho^3 \quad (17)$$

$$\dot{\phi} = \Omega + \mathbf{p} \times \nabla c \quad (18)$$

$$\dot{c} = \rho - c + \mathcal{D}_c \nabla^2 c + \varepsilon (c_0 - c)^3 \quad (19)$$

Here, $\Omega = \omega/k_d$ and $\mathcal{D}_c = D_c/D_\rho$ determine the linear behaviour together with the effective density ρ_0 which is conserved in the course of the dynamics, while $\kappa = K k_d^2 v_0^2 / (D_\rho k_0^2 \beta^2)$ and $\varepsilon = \epsilon v_0^2 / (k_d \beta^2)$ control non-linear saturation effects (besides the κ term producing some contribution to the colloidal diffusion term). As a key control parameter we identify $\Delta := \Omega/\rho_0 = v_0 \omega / (\beta k_0 \rho_0)$ which measures (for given self-propulsion speed) the relative importance of active rotations and chemotaxis.

Promising candidates to physically realize our predictions are auto-chemotactic strains of E.coli which rotate naturally close to a wall or interface. Here, one could measure the initial time lag followed by a delayed onset of clustering as an indication of the nonlinear locking instability. In addition, one could also measure the suppression of rotations which arises exclusively at the interface between dense and dilute regions for comparatively low effective rotation frequencies (see below) and the sudden arrest of coarsening when for larger rotation frequencies.

The parameter values used in this work (Fig. 2) correspond to typical experimental values; the most relevant dimensionless control parameters Ω , $\tilde{\rho}$ and $\mathcal{D}_c$ translate as follows. (i) The choice $D_c/D_\rho = 1$ approximately matches with measurements of $D_\rho \sim 2 \times 10^{-6} - 1.5 \times 10^{-5} \text{cm}^2/\text{s}$ and $D_c \sim 10^{-5} \text{cm}^2/\text{s}$ [6–9]. (ii) Our parameter choice in Fig. 2 corresponds to $\rho_0 \beta k_0 / k_d v_0 = 50$ which can be matched for typical ‘chemotactic sensitivities’ of $\chi \sim (1.5 -$

$75) \times 10^{-5} \text{cm}^2/\text{s}$ [6, 7] with $\chi \sim \beta v_0/(2D_r)$ when assuming $D_r \sim 0.1 - 1/\text{s}$ and rates $k_d \sim 0.09/\text{s}$ and $k_0 \sim 0.6/\text{s}$ typical for the chemoattractant of Dicty (cAMP [10, 11], rates for E.coli are unknown [9]) when using systems with $> 5 \times 10^3$ bacteria per mm^2 or area fractions > 0.1 . (iii) The parameter $\Omega = \omega/k_d \sim 2$ for typical swimming radii of $50\mu\text{m}$ [12] and swimming speeds of $10\mu\text{m}/\text{s}$ which is close to the value at which we observed the transition from a large cluster to a stripe pattern; see Figs. 2i and l. Faster swimming leads to larger values of Ω and could allow to observe the decrease of the wavelength of our travelling wave pattern. Smaller values of Ω corresponding to parameters in Figs. 2d-f would require slower swimming velocities or faster decay (consumption) of the E.coli chemoattractant aspartate than for cAMP. Alternatively, by working with magnetotactic bacteria which have a permanent dipole moment or active bimetallic colloids [13], slow rotations could be easily generated by exposing these particles to an external rotating magnetic field.

C. Reduced models and linear stability analysis

We now derive a reduced model for the rotor distribution $\rho(\mathbf{r}, t)$ by assuming that $\mathbf{p}$ is a fast variable, which is a good approximation for the parameter regime considered in the main text. This allows us to adiabatically eliminate the orientation equation. First, we demonstrate that in absence of active rotations and alignment interactions $\Omega = G = 0$, Eqs. (6-8) reduce to the well-known Keller-Segel model [14, 15] describing chemotaxis of signalling microorganisms, and are therefore consistent with previous studies of active colloids based on this model [16–18]. In particular, the Keller-Segel model for chemoattractive particles ($\beta > 0$) allows for cluster growth typically proceeding to phase separation. Here, as we shall see, local alignment interactions ($G > 0$) will support this instability in the sense that they allow for cluster growth already at very low particle densities. Second, we will derive a corresponding reduced model in presence of active rotations, starting with Eqs. (1-3) in the main text. This model shows that active rotations suppress linear instability in its complete parameter space, even if strong alignment interactions are present.

1. Keller-Segel model

Starting from Eq. (7) and adiabatically eliminating $\dot{\mathbf{w}} \rightarrow \mathbf{0}$ as well as neglecting gradient terms of order ∇^2 , with $\Omega = G = 0$ we find $\mathbf{w} = -v_0 \nabla \rho / (2D_r) + \beta \rho \nabla c / 2D_r$. Plugging this into Eq. (6) leads together with Eq. (8) to the following model which resembles the Keller-Segel model of chemotaxis (compare [14–18])

$$\dot{\rho} = -\frac{v_0 \beta}{2D_r} \nabla \cdot (\rho \nabla c) + D \nabla^2 \rho \quad (20)$$

$$\dot{c} = k_0 \rho - k_d c + D_c \nabla^2 c + \epsilon (c - c_0)^3 \quad (21)$$

Here $D = v_0^2 / (2D_r)$ is the effective ‘active’ diffusion constant.

Linearizing around the uniform solution $(\rho, c) = (\rho_0, k_0 \rho_0 / k_d)$ and using $\rho' = \rho - \rho_0, c' = c - c_0$ leads to:

$$\begin{aligned} \dot{\rho}' &= -B \nabla^2 c' + D \nabla^2 \rho' \\ \dot{c}' &= k_0 \rho' - k_d c' + D_c \nabla^2 c' \end{aligned} \quad (22)$$

where $B = \frac{v_0 \beta \rho_0}{2D_r}$. We now test the stability of these equations with respect to plane wave perturbations. Fourier transforming (22) and evaluating the respective linear stability problem we find a long wavelength instability for $Bk_0 > Dk_d$ with instability band $q^2 < (Bk_0 - Dk_d)/(DD_c)$. This instability is based on the positive feedback loop explained in our introduction (see main text) and hinges on aligning particles up the chemical gradient. Accordingly, it is no surprise that local alignment interactions only help destabilize the uniform state and allow for cluster growth already at very low effective densities. For a later comparison with the case of active rotations it is instructive to further reduce model (22). Assuming that c' is fast compared to the conserved colloidal field after adiabatic elimination $c' \rightarrow 0$ and neglecting $D_c \nabla^2 c'$ Eq. (22) simplifies to

$$\dot{\rho}' = [D - (k_0/k_d)B] \nabla^2 \rho' \quad (23)$$

This is a closed model for the particle density close to the uniform state where chemotaxis directly competes with the effective diffusion. This equation reproduces the correct instability criterion, but leads to a short wavelength divergence, normally prevented by chemical diffusion.

2. Adiabatic solution of the Adler equation

To derive a reduced model for active rotors ($\Omega > 0$) we need to solve Eq. (18) explicitly. Rewriting Eq. (18) leads to the Adler equation $\dot{\phi} = \Omega + |\nabla c| \sin(\phi + \delta)$, here with $\delta = \text{atan2}(\partial_y c, -\partial_x c)$. We now solve this equation in the adiabatic limit of fast response of ϕ to changes in the chemical field. Since in presence of active rotations the collective propulsion direction $\mathbf{p}$ as determined by ϕ does not simply follow the chemical field but is determined by a competition between chemotaxis and active rotations, we may not simply set $\dot{\phi} \rightarrow 0$, but need to integrate Eq. (4), MT for a (quasi-)stationary chemical field to describe fast orientational response appropriately, i.e. we integrate the Adler equation for temporarily fixed c , restoring its time-dependence only after integration. In this adiabatic approximation we find (compare also [19])

$$\phi_{\text{ad}} = -\text{atan2}(\partial_y c, -\partial_x c) + 2 \arctan \left[\frac{-|\nabla c| + \sqrt{\Omega^2 - |\nabla c|^2} \tan \left(\sqrt{\Omega^2 - |\nabla c|^2} t / 2 + \theta_0 \right)}{\Omega} \right] \quad (24)$$

Here $\theta_0(\mathbf{x})$ is an integration constant that is fixed by the initial orientations of the rotors. For $|\nabla c| < \Omega$ (24) describes an anharmonic and anisotropic periodic oscillation. As the chemical gradient increases, the rotation frequency decreases as $\sqrt{\Omega^2 - |\nabla c|^2}$, i.e. $\mathbf{p}$ rotates slower where chemical gradients are strong.

At $|\nabla c| = \Omega$ the colloids cease to rotate (on average), i.e. we have a transition from rotations to locking. Here, we have an equilibrium between chemotaxis and rotations, where rotations steer the director field away from the ‘optimal’ swimming direction up the chemical gradient (which is approached for $|\nabla c| \rightarrow \infty$), and towards a direction perpendicular to it. From Eq. (24) it is straightforward to derive the distance from the ‘optimal’ angle as

$$\Delta := \phi_{\text{ad}}(\Omega) - \phi_{\text{ad}}(\Omega \rightarrow 0) = 2 \arctan \left[\frac{\gamma}{1 + \sqrt{1 - \gamma^2}} \right] \quad (25)$$

where we used the notation $\gamma := \Omega/|\nabla c|$. For $\Omega < 1/2$ we can approximate $\Delta \approx \gamma$ and conversely, for Ω close to $|\nabla c|$, we have $\Delta \approx \pi/2 - \sqrt{2(\Omega/|\nabla c| - 1)}$. In other words, the advective particle flux up density (chemical) gradients ($\propto \cos \Delta$) decreases slowly with increasing Ω when Ω is small, but decreases faster and faster for large Ω .

3. Linear stability of active rotors

We now use Eq. (24) to formulate a reduced model for the density of rotors whose orientation responds quickly to changes in the chemical field. For a quasi-instantaneous orientational dynamics Eqs. (17,19) lead to

$$\begin{aligned} \dot{\rho} &= \nabla^2 \rho + \kappa \nabla^2 \rho^3 - \nabla \cdot (\rho \mathbf{p}_{\text{ad}}(t)) \\ \dot{c} &= \rho - c + \mathcal{D}_c \nabla^2 c \end{aligned} \quad (26)$$

where $\mathbf{p}_{\text{ad}} = (\cos(\phi_{\text{ad}}), \sin(\phi_{\text{ad}}))^T$ and ϕ_{ad} is given by Eq. (24). To understand the linear stability of the uniform solution $(\rho, c) = (\rho_0, \rho_0)$ of these equations with $\theta(\mathbf{x}) = \text{const}$, we choose a coordinate system with x-axis parallel to $\mathbf{p}(t=0)$, i.e. $\theta = \arctan \left[(|\nabla c| + \Omega \tan(\delta/2)) / \sqrt{\Omega^2 - |\nabla c|^2} \right]$ and expand $\mathbf{p}_{\text{ad}}$ in $\partial_x c, \partial_y c$:

$$\mathbf{p}_{\text{ad}} = \begin{pmatrix} \cos \Omega t \\ \sin \Omega t \end{pmatrix} + \frac{1 - \cos \Omega t}{\Omega} \begin{pmatrix} \sin \Omega t \\ -\cos \Omega t \end{pmatrix} \partial_x c + \frac{\sin \Omega t}{\Omega} \begin{pmatrix} -\sin \Omega t \\ \cos \Omega t \end{pmatrix} \partial_y c \quad (27)$$

Hence, we obtain:

$$\begin{aligned} \dot{\rho}' &= (1 + 3\rho_0^2 \kappa) \nabla^2 \rho' - \begin{pmatrix} \cos \Omega t \\ \sin \Omega t \end{pmatrix} \nabla \rho' + \frac{\sin^2 \Omega t + \cos \Omega t - \cos^2 \Omega t}{\Omega} \partial_x \partial_y c - \frac{\sin \Omega t}{\Omega} [1 - \cos \Omega t] \partial_x^2 c - \frac{\cos \Omega t \sin \Omega t}{\Omega} \partial_y^2 c \\ \dot{c}' &= \rho' - c' + \mathcal{D}_c \nabla^2 c' \end{aligned} \quad (28)$$

Remarkably, the active rotations enter the linearized equations as explicitly time-dependent ‘driving’ terms. To understand how this leads to linear stability of the uniform state, we first consider the instructive case of fast chemical dynamics $\dot{c} \rightarrow 0$, before generalizing our approach in the next paragraph. Neglecting also chemical diffusivity ($\mathcal{D}_c \rightarrow 0$) which only helps stabilizing the uniform state, we have $c' = \rho'$. Now using a plane wave Ansatz $\rho' = r(t) \exp[-i\mathbf{q} \cdot \mathbf{x}]$ in Eqs. (28), Floquet theory predicts $r(t) = r(0) \exp[\mu t] g(t)$ [20], where $g(t)$ is some $2\pi/\Omega$ -periodic function. Here, μ is called the Floquet exponent and serves as a linear stability parameter; if it is positive $r(t)$ oscillates with growing

amplitude, but for $\mu < 0$ small density fluctuations around the uniform state decay. According to the Liouville formula in Floquet theory [20], here μ is given by the time-average of the right hand side of Eqs. (28). Thus, we have $\mu = -(1 + 3\rho_0^2\kappa)\mathbf{q}^2$ meaning that active rotations *suppress* the chemotactic instability, which occurs in their absence (Eq. 23). More specifically, the time-dependent driving terms in Eq. (28) effectively suppress the impact of self-advection on the dynamics: in the absence of rotations this leads to a spinodal instability (see Eq. 23). This confirms the intuitive argument given in the introduction of the main text: close to the uniform state, chemical gradients are too weak to align the swimming directions of our rotors and so rotations dominate chemotaxis. As we discuss in the main text, this picture breaks down away from the uniform state and can give rise to a nonlinear instability allowing for structure formation with chemically interacting rotors despite the fact that the uniform state is now linearly stable.

4. Generalized linear stability analysis: Floquet-Magnus expansion

Finally, we generalize the linear stability analysis of the previous paragraph to cases where we may not assume that c is a fast variable and have to account for corresponding delay effects. We first plug the Ansatz $\rho' = r(t)\exp[-i\mathbf{q} \cdot \mathbf{x}]$, $c' = C(t)\exp[-i\mathbf{q} \cdot \mathbf{x}]$ into Eqs. (28), which leads to

$$\begin{pmatrix} \dot{r} \\ \dot{C} \end{pmatrix} = A \begin{pmatrix} r \\ C \end{pmatrix} \quad (29)$$

where the $2\pi/\omega$ -periodic matrix $A = (a)_{ij}$ has the following components

$$a_{11} = -\mathbf{q}^2(1 + 3\rho_0^2\kappa) - i[q_x \cos \Omega t + q_y \sin \Omega t] \quad (30)$$

$$a_{12} = \frac{\rho_0}{\Omega} [q_x q_y (\cos^2 \Omega t - \sin^2 \Omega t - \cos \Omega t) + (q_y^2 - q_x^2) \cos \Omega t \sin \Omega t + q_x^2 \sin \Omega t] \quad (31)$$

$$a_{21} = 1 \quad (32)$$

$$a_{22} = -\mathcal{D}_c \mathbf{q}^2 \quad (33)$$

Denoting the fundamental matrix solution to Eq. (29) as X , we have $\dot{X}(t) = A(t)X(t)$, where $X(t) = P(t)\exp[tB]$ with $P(t+2\pi/\omega) = P(t)$, according to the Floquet theorem [20]. The eigenvalues of B are the Floquet exponents $\mu_{1,2}$ which determine the linear stability of the present problem. From here, we follow [21] to perform a Floquet-Magnus expansion, i.e. we expand P, B

$$P = \exp[\Lambda]; \quad \Lambda = \sum_{k=1}^{\infty} \Lambda_k; \quad \Lambda_k(t+T) = \Lambda_k(t) \quad (34)$$

$$B = \sum_{k=1}^{\infty} B_k \quad (35)$$

and determine B up to order $k = 2$. To first order, we find

$$\mu_1 = -(1 + 3\rho_0^2\kappa)\mathbf{q}^2; \quad \mu_2 = -1 - \mathcal{D}_c \mathbf{q}^2 \quad (36)$$

which suggests linear stability for all values of Ω . The general result at second order ($k = 2$) is more involved and its physical meaning is not immediate; thus we perform an asymptotic expansion up to second order in $\nu = 1/\Omega$ which yields

$$\mu_{1,2} = -\frac{1}{2} [1 + (1 + 3\rho_0^2\kappa + \mathcal{D}_c)\mathbf{q}^2 + |1 + (\mathcal{D}_c - 1 - 3\rho_0^2\kappa)\mathbf{q}^2|] \pm i \frac{2q_x \mathbf{q}^2 \rho_0}{\Omega^2 |1 + (\mathcal{D}_c - 1 - 3\rho_0^2\kappa)\mathbf{q}^2|} \quad (37)$$

The real parts of $\mu_{1,2}$ (Lyapunov exponents) represent the average growth rate of small perturbations of the uniform state. Thus, even if c is not a fast variable, at least for large Ω active rotations suppress linear stability. Note that convergence of the Floquet-Magnus expansion can be shown here for large Ω when choosing the 2-norm of A to satisfy the convergence criterion in [21].

To generalize our approach further, for small Ω we calculated the Floquet exponents $\mu_{1,2}$ numerically for different portions of the parameter space. Here, we found that $\mu_{1,2}$ have negative real parts (linear stability of the uniform state) in most regions of the parameter regime, even for relatively slow driving. However, interestingly, they can become positive if the effective ρ_0 is large enough and $\kappa, \mathcal{D}_c, \Omega$ are sufficiently small. This demonstrates that besides the nonlinear instability mechanism we discussed in the main text, also delay effects in the response of c to changes in ρ could provide a route to structure formation in slowly rotating chemically interacting particles. Although this linear route to structure formation applies only to a restricted subset of the parameter space, it could be an interesting topic for further investigations.

D. Videos

Video 1 shows the dynamics of signalling rotors at short and intermediate times for $\Omega = 0$ (this corresponds to Fig. 2a-c, main text); $\Omega = 1.2$ (Fig. 2g-i); $\Omega = 2.2$ (Fig. 2j-l) and $\Omega = 7$. Initial states are identical in all four cases. The two latter cases ($\Omega = 2.2$ and $\Omega = 7$) lead to travelling wave patterns on timescales beyond the ones shown in the video. Other parameters are chosen as in Fig. 2. Videos 2,3,4 show the rotor dynamics (ρ and $\mathbf{p}$) on comparatively long timescales for $\Omega = 0.25$ (This case corresponds to Fig. 2d-f, but for larger system size), $\Omega = 1.2$ (g-i) and $\Omega = 2.2$ (j-l), respectively. Video 5 shows c and $\mathbf{p}$ for parameters as in Fig. 2d-f and comparatively short timescales.

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