Work in progress:
Flow and phase separation of active spinners

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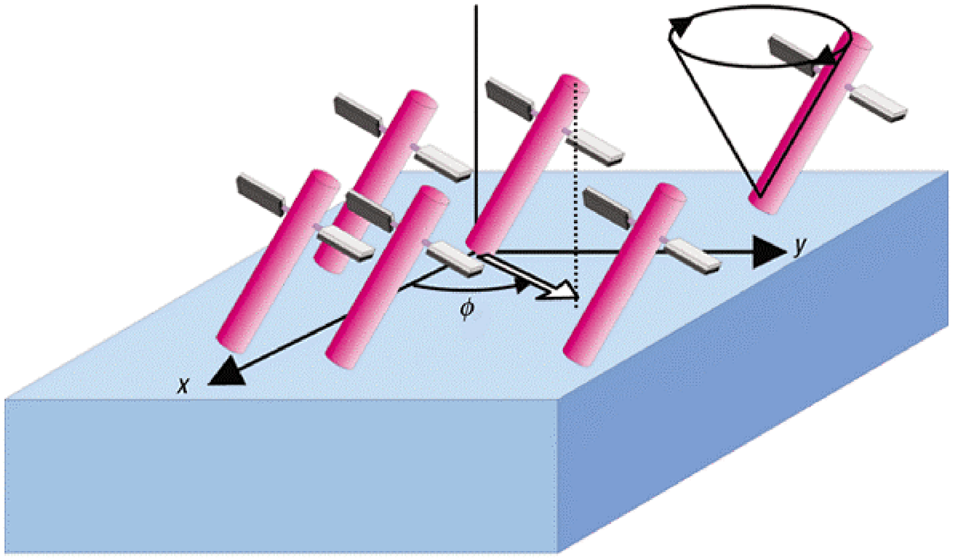
I. **Flow:** Inspired by experiments of Yuka Tabe (Work with former graduate student Lena Lopatina, now at Los Alamos National Laboratory)

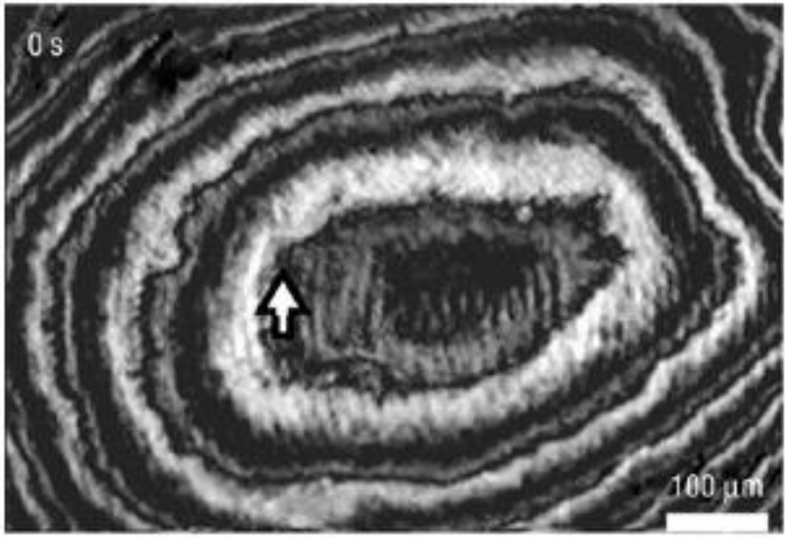
II. **Phase separation:** Inspired by simulations of Sharon Glotzer

Experiment: Driven flow in chiral liquid-crystal films

Experiments by Yuka Tabe (Waseda Univ.) and Hiroshi Yokoyama (LCI)

- Experiment #1: Langmuir monolayers of chiral molecules on glycerol
  - Change humidity of air ⇒ Water transport across monolayer
  - Motion of water ⇒ Precession of chiral liquid-crystal molecules
  - Boundary conditions on domain walls, competition between driving and elastic interactions ⇒ Complex director modulations
  - *Observe only precession, not flow ⇒ Presumably because of viscosity of underlying fluid*





Experiment: Driven flow in chiral liquid-crystal films

- Experiment #2: Freely suspended films of chiral liquid crystals in smectic-C phase
  - Different humidity on two sides $\Rightarrow$ Water transport across film
  - Motion of water $\Rightarrow$ Precession of chiral liquid-crystal molecules
  - No underlying viscous fluid $\Rightarrow$ *Observe both director rotation and large-scale flow* (based on motion of dust particles)

- Experiment #3: Freely suspended films of chiral liquid crystals in smectic-A phase
  - Motion of water $\Rightarrow$ Rotation of chiral liquid-crystal molecules about their long axes
  - Cannot observe director rotation, because there is no director
  - *Observe large-scale flow* (based on motion of dust particles), less than in smectic-C case

Simulation: Phase separation of active spinners

Emergent collective phenomena in a mixture of hard shapes through active rotation

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We investigate collective phenomena with rotationally driven spinners of concave shape. Each spinner experiences a constant internal torque in either a clockwise or counterclockwise direction. Although the spinners are modeled as hard, otherwise non-interacting rigid bodies, we find that their active motion induces an effective interaction that favors rotation in the same direction. With increasing density and activity, phase separation occurs via spinodal decomposition, as well as self-organization into rotating crystals. We observe the emergence of cooperative, super-diffusive motion along interfaces, which can transport inactive test particles. Our results demonstrate novel phase behavior of actively rotated particles that is not possible with linear propulsion or in non-driven, equilibrium systems of identical hard particles.

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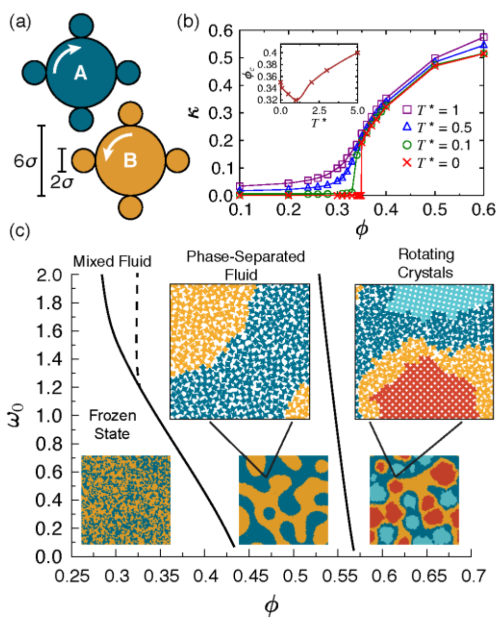
Introduction.—Active matter is a rapidly growing branch of non-equilibrium soft matter physics with relevance to fields such as biology, energy, and complex systems [1]. In active matter, dissipative, steady-state structures far-from-equilibrium can emerge in systems of particles by converting energy to particle motility [1, 2]. Recent works have reported novel collective behavior not possible with passive matter, such as giant number fluctuations [3], clustering [4], swarming [5, 6], fluid-solid phase separation of repulsive disks [7–9], and collective rotors [10]. Effective interactions emerging between hard, self-propelled particles were shown to cause phase separation [9, 11–13] and coexistence [9] in simulations. Experimentally, some of these phenomena were demonstrated by driving the system via vibration [14–16], chemical reaction [17, 18], and light activated propulsion [19]. To date, most studies have focused on self-propulsion where the constant input of energy to each particle goes directly into translational motion and hence active forces couple to particle velocities. Converting the input of energy into *rotational* motion, however, does not directly influence translational motility, and couples only to the particles' angular momentum. We denote such a coupling of active driving forces to angular velocity as *active rotation.*

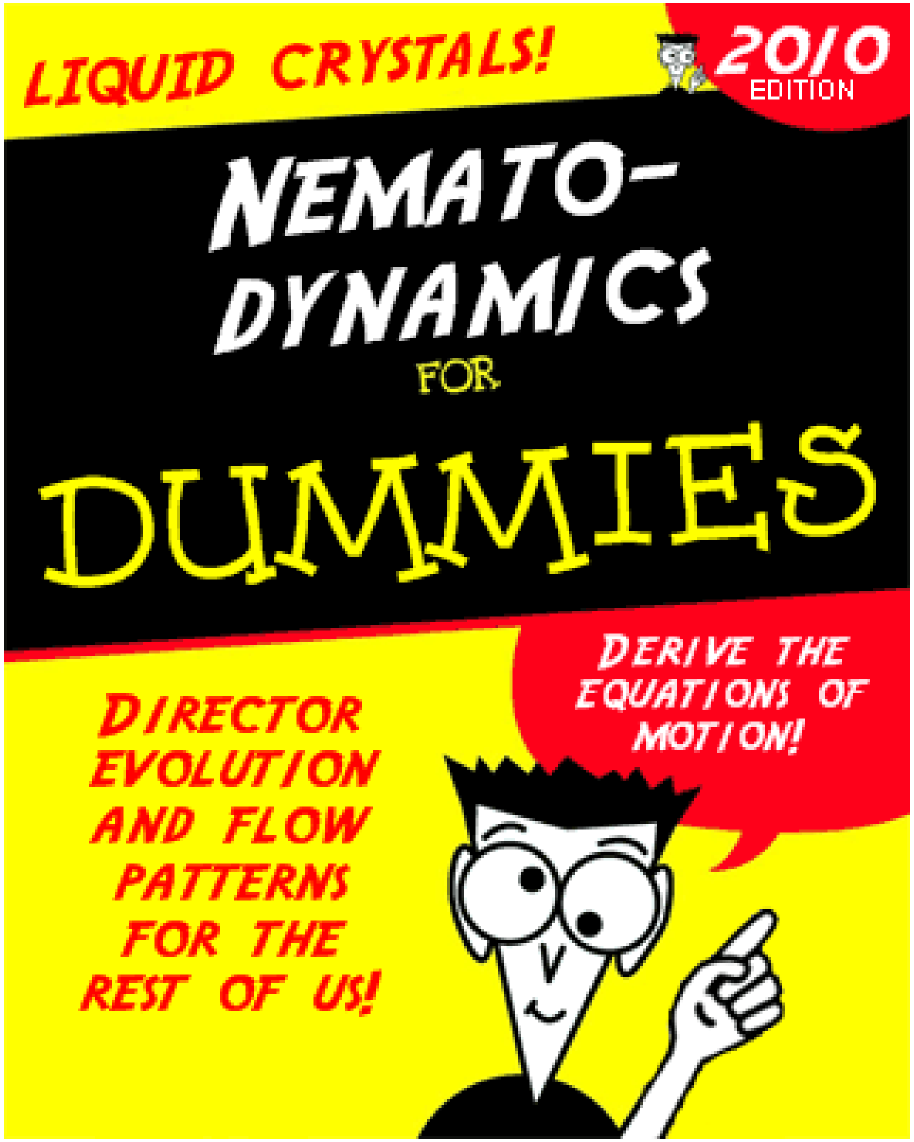


Active rotation may be achieved by various methods, *e.g.* external magnetic fields [20, 21] and optical tweezers [22–24]. Biological organisms can spin naturally, such

Theoretical issues

- Specific connection between experiment and simulation
  - Driven rotation in experiment ⇔ active rotation in simulation
  - Future experiment could investigate racemic mixture of right- and left-handed molecules, look for chiral phase separation induced by water transport
- General principles
  - How does driving/activity induce flow?
  - How does driving/activity induce phase transition?

Flow:
Use my favorite textbook



Lagrangian dynamics

- Lagrangian $L = T - V$

kinetic → T; potential → V

- Equation of motion

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{q}_j}\right) - \frac{\partial L}{\partial q_j} = 0$$

generalized coordinates → q_j

- With dissipation

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{q}_j}\right) - \frac{\partial L}{\partial q_j} + \frac{\partial D}{\partial \dot{q}_j} = 0$$

D = Rayleigh dissipation function
= half the rate of energy dissipation

- With constraints

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{q}_j}\right) - \frac{\partial L}{\partial q_j} = \sum_{\ell}\left(\lambda_\ell \frac{\partial c_\ell}{\partial q_j} + \mu_\ell \frac{\partial c_\ell}{\partial \dot{q}_j}\right)$$

- With dissipation *and* constraints

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{q}_j}\right) - \frac{\partial L}{\partial q_j} + \frac{\partial L}{\partial \dot{q}_j} = \sum_{\ell}\left(\lambda_\ell \frac{\partial c_\ell}{\partial q_j} + \mu_\ell \frac{\partial c_\ell}{\partial \dot{q}_j}\right)$$

Strategy

- Construct general expressions for *T*, *V*, and *D* allowed by symmetry
- Derive Lagrangian equations of motion

Case 1: Ordinary fluid

- Generalized coordinates

- Generalized velocities

$$\mathbf{v}(\mathbf{r},t)$$

- Kinetic energy density

$$T = \tfrac{1}{2}\rho|\mathbf{v}|^2$$

- Potential energy density

$$V = 0$$

- Constraints

$$\nabla\cdot\mathbf{v} = 0$$ ← incompressibility

- Modes that dissipate energy

$$A_{ij} = \tfrac{1}{2}\left(\partial_i v_j + \partial_j v_i\right)$$ ← shear rate

- Dissipation function density

$$D = \eta A_{ij} A_{ij}$$

- Lagrangian equation of motion

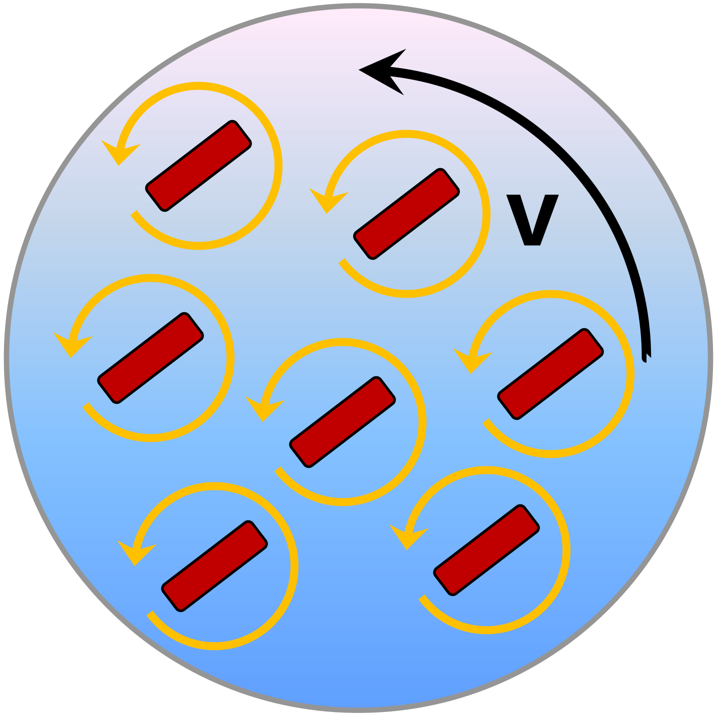
$$\rho\frac{d\mathbf{v}(\mathbf{r},t)}{dt} - \eta\nabla^2\mathbf{v} = -\nabla p(\mathbf{r},t)$$

↑ Lagrange multiplier is pressure *p*

Navier-Stokes equation

Case 2:
Nematic liquid crystal

Key issue: Coupling between orientation and flow



Case 2: Nematic liquid crystal

- Generalized coordinates

$$\hat{\mathbf{n}}(\mathbf{r},t)$$

- Generalized velocities

$$\mathbf{v}(\mathbf{r},t), \quad \dot{\mathbf{n}}(\mathbf{r},t)$$

- Kinetic energy density

$$T = \tfrac{1}{2}\rho|\mathbf{v}|^2 + \tfrac{1}{2}I|\dot{\mathbf{n}}|^2$$

- Potential energy density

$$V = 0$$

- Constraints

$$\nabla \cdot \mathbf{v} = 0$$

$$|\hat{\mathbf{n}}|^2 = 1$$

- Modes that dissipate energy

$$A_{ij} = \tfrac{1}{2}\left(\partial_i v_j + \partial_j v_i\right)$$

$$\mathbf{N} = \dot{\mathbf{n}} - \boldsymbol{\omega} \times \mathbf{n}$$ ← director rotation wrt background flow

where background rotational flow is

$$\boldsymbol{\omega} = \tfrac{1}{2}\nabla \times \mathbf{v}$$

- Dissipation function density

$$D = \quad \left(A_{ij}A_{ij}\right) + \quad \left(n_i A_{ij} n_j\right)^2$$
$$+ \quad \left(n_i A_{ij} A_{jk} n_k\right)$$
$$+ \quad \left(N_i N_i\right) + \quad \left(N_i A_{ij} n_j\right)$$

Case 2: Nematic liquid crystal

- Generalized coordinates

$$\hat{\mathbf{n}}(\mathbf{r},t)$$

- Generalized velocities

$$\mathbf{v}(\mathbf{r},t), \quad \dot{\mathbf{n}}(\mathbf{r},t)$$

- Kinetic energy density

$$T = \tfrac{1}{2}\rho|\mathbf{v}|^2 + \tfrac{1}{2}I|\dot{\mathbf{n}}|^2$$

- Potential energy density

$$V = 0$$

- Constraints

$$\nabla \cdot \mathbf{v} = 0$$

$$|\hat{\mathbf{n}}|^2 = 1$$

Ericksen-Leslie theory

- Modes that dissipate energy

$$A_{ij} = \tfrac{1}{2}\left(\partial_i v_j + \partial_j v_i\right)$$

$$\mathbf{N} = \dot{\mathbf{n}} - \boldsymbol{\omega} \times \mathbf{n}$$ ← director rotation wrt background flow

where background rotational flow is

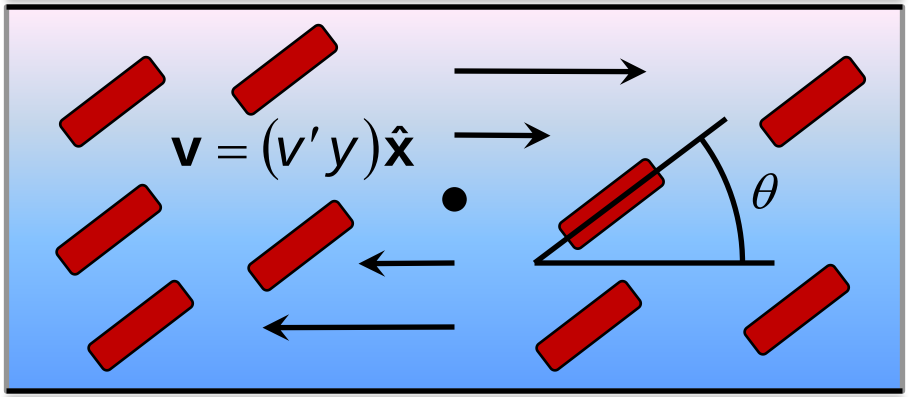
$$\boldsymbol{\omega} = \tfrac{1}{2}\nabla \times \mathbf{v}$$

- Dissipation function density

$$D = \tfrac{1}{2}\alpha_4\left(A_{ij}A_{ij}\right) + \tfrac{1}{2}\alpha_1\left(n_i A_{ij} n_j\right)^2 + \tfrac{1}{2}\left(\alpha_5 + \alpha_6\right)\left(n_i A_{ij} A_{jk} n_k\right) + \tfrac{1}{2}\gamma_1\left(N_i N_i\right) + \gamma_2\left(N_i A_{ij} n_j\right)$$

where α's = Miesowicz viscosities
γ_1 = rotational viscosity
γ_2 = torsion coefficient

Example: Shear alignment



- Impose a shear flow profile
  $$\mathbf{v} = (v'y)\hat{\mathbf{x}}$$
- Calculate the bulk nematic alignment, parameterized by
  $$\hat{\mathbf{n}} = (\cos\theta, \sin\theta, 0)$$
- Generalized coordinate θ
- Generalized velocity $\dot{\theta}$
- Kinetic energy density
  $$T = \text{const} + \tfrac{1}{2} I \dot{\theta}^2$$
- Potential energy density $V = 0$ (can add elasticity and anchoring later if we wish)
- Dissipation function density
  $$D = \text{const} + \tfrac{1}{2}\gamma_1\left(\dot{\theta} + \tfrac{1}{2}v'\right)^2 + \tfrac{1}{2}\gamma_2 v'\left(\dot{\theta} + \tfrac{1}{2}v'\right)\cos 2\theta$$
- Constraints automatically satisfied
- Equation of motion
  $$I\ddot{\theta} + \gamma_1\left(\dot{\theta} + \tfrac{1}{2}v'\right) + \tfrac{1}{2}\gamma_2 v' \cos 2\theta = 0$$
- Steady-state solution
  $$\theta = \frac{1}{2}\cos^{-1}\left(-\frac{\gamma_1}{\gamma_2}\right)$$

Case 1 ¾: Fluid with spinning particles but no director

- Generalized coordinates

- Generalized velocities

$$\mathbf{v}(\mathbf{r},t), \quad \mathbf{\Omega}(\mathbf{r},t)$$ ← spin

- Kinetic energy density

$$T = \tfrac{1}{2}\rho|\mathbf{v}|^2 + \tfrac{1}{2}I|\mathbf{\Omega}|^2$$

- Potential energy density

$$V = 0$$

- Constraints

$$\nabla \cdot \mathbf{v} = 0$$

- Modes that dissipate energy

$$A_{ij} = \tfrac{1}{2}\left(\partial_i v_j + \partial_j v_i\right)$$

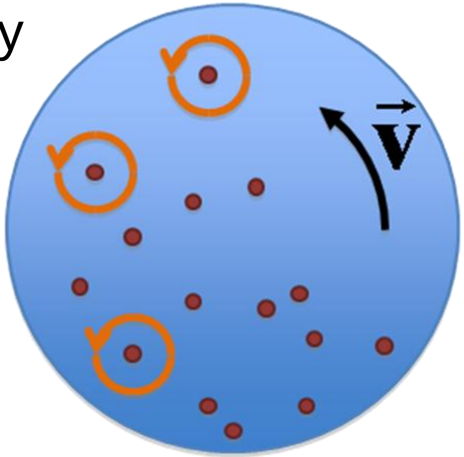
$$\mathbf{\Delta\Omega} = \mathbf{\Omega} - \boldsymbol{\omega}$$ ← spin wrt background flow

where background rotational flow is

$$\boldsymbol{\omega} = \tfrac{1}{2}\nabla \times \mathbf{v}$$

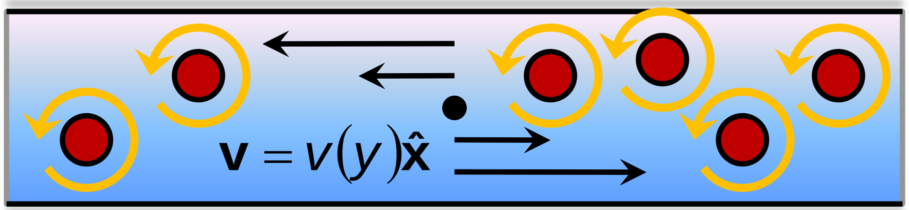
- Rayleigh dissipation function

$$D = \eta A_{ij}A_{ij} + \tfrac{1}{2}\gamma_1|\mathbf{\Delta\Omega}|^2$$



Similar to Ye-Lubensky theory for chiral granular materials

Model for chiral SmA film: Simple rectangular geometry



top view

- Generalized velocities

$$v(y,t), \quad \Omega(y,t)$$

- Kinetic energy density

$$T = \tfrac{1}{2}\rho v^2 + \tfrac{1}{2} I \Omega^2$$

- Dissipation function density

$$D = \frac{1}{2}\eta\left(\frac{\partial v}{\partial y}\right)^2 + \frac{1}{2}\gamma_1\left(\Omega + \frac{1}{2}\frac{\partial v}{\partial y}\right)^2$$

- Additional dissipation function due to surface stress σ

$$D_{\text{surf}} = \tfrac{1}{2}\sigma\left(v(y_{\min}) - v(y_{\max})\right)$$

- Constraints automatically satisfied
- Equations of motion

torque from water transport

$$I\frac{d\Omega}{dt} + \gamma_1\left(\Omega + \frac{1}{2}\frac{\partial v}{\partial y}\right) = \tau_{\text{water}}$$

$$\rho\frac{dv}{dt} - \eta\frac{\partial^2 v}{\partial y^2} - \frac{1}{2}\gamma_1\frac{\partial}{\partial y}\left(\Omega + \frac{1}{2}\frac{\partial v}{\partial y}\right) = 0$$

- Boundary conditions

$$\eta\frac{\partial v}{\partial y} + \frac{1}{2}\gamma_1\left(\Omega + \frac{1}{2}\frac{\partial v}{\partial y}\right) - \frac{1}{2}\sigma = 0$$

Question of theoretical formalism

Where does this term come from?
Possibilities are:

- Dissipation function: Term of

$$D = \ldots - \tau_{\text{water}}\Omega = \ldots - \tau_{\text{water}}\dot{\theta}$$

- Free energy: Term of

$$F = \ldots - \tau_{\text{water}}\theta$$

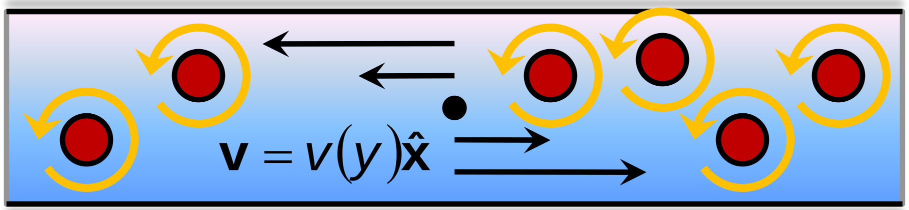
- Somewhere else?

torque from water transport

$$I\frac{d\Omega}{dt} + \gamma_1\left(\Omega + \frac{1}{2}\frac{\partial v}{\partial y}\right) = \tau_{\text{water}}$$

$$\rho\frac{dv}{dt} - \eta\frac{\partial^2 v}{\partial y^2} - \frac{1}{2}\gamma_1\frac{\partial}{\partial y}\left(\Omega + \frac{1}{2}\frac{\partial v}{\partial y}\right) = 0$$

Model for chiral SmA film: Simple rectangular geometry



top view

- Steady state solution

$$v(y) = v_0 - \left(\frac{\tau_{\text{water}} - \sigma}{2\eta}\right) y$$

$$\Omega(y) = \frac{\tau_{\text{water}}}{\gamma_1} + \frac{\tau_{\text{water}} - \sigma}{4\eta}$$

- If there's no shear stress σ resisting the shear flow, then

$$v(y) = v_0 - \left(\frac{\tau_{\text{water}}}{2\eta}\right) y$$

$$\Omega(y) = \tau_{\text{water}}\left(\frac{1}{\gamma_1} + \frac{1}{4\eta}\right)$$

- If the boundaries prevent shear flow, then they must be providing a shear stress of $\sigma = \tau_{\text{water}}$

Fluid must be providing shear stress on boundary of $\sigma = -\tau_{\text{water}}$

Phase separation

Question:

- How to combine active rotation with continuum hydrodynamic description of the phase transition?

Variables:

- Chiral composition $\psi(\mathbf{r},t) = \rho_{\text{right}}(\mathbf{r},t) - \rho_{\text{left}}(\mathbf{r},t)$
- Spin $\Omega(\mathbf{r},t)$
- Flow velocity $\mathbf{v}(\mathbf{r},t)$

Analogy with hypothetical equilibrium problem

If it were equilibrium statistical mechanics…

- Use free energy

entropy of mixing

$$F = k_B T\left[\left(\frac{1+\psi}{2}\right)\log\left(\frac{1+\psi}{2}\right)+\left(\frac{1-\psi}{2}\right)\log\left(\frac{1-\psi}{2}\right)\right]+\frac{1}{2}\kappa\left|\nabla\psi\right|^2+\frac{1}{2}a\Omega^2-b\psi\Omega$$

$$\approx \frac{1}{2}k_B T\psi^2+\frac{1}{12}k_B T\psi^4+\frac{1}{2}\kappa\left|\nabla\psi\right|^2+\frac{1}{2}a\Omega^2-b\psi\Omega$$

- Minimize over $\Omega \Rightarrow$ effective free energy in terms of ψ only

$$F_{\text{eff}} = \frac{1}{2}\left(k_B T-\frac{b^2}{a}\right)\psi^2+\frac{1}{2}\kappa\left|\nabla\psi\right|^2+\frac{1}{12}k_B T\psi^4$$

- Chiral symmetry-breaking transition at critical temperature $k_B T = \frac{b^2}{a}$

How to formulate the corresponding active problem?

Case A

- Zero-dimensional, no conservation law, no flow velocity
- By analogy with previous problems, equations of motion must be

Derive from rotational viscosity term in dissipation function

Active spinning term: Derive from dissipation function, free energy, or ?

$$\frac{d\Omega}{dt} = -\ldots\Omega + \ldots\psi$$

$$\frac{d\psi}{dt} = -\ldots\psi + \ldots\Omega$$

Derive from entropy of mixing term in free energy

Active spinning term: Derive from ?

Case B

- One-dimensional with conservation law for chiral particles, still no flow velocity
- Conservation law gives $\dfrac{\partial \psi}{\partial t} = -\dfrac{\partial J}{\partial x}$

  where current is $J = -\dfrac{\partial}{\partial x}\left[\ldots \psi - \ldots \dfrac{\partial^2 \psi}{\partial x^2} - \ldots \Omega \right]$

  Derive from entropy of mixing

  Derive from gradient term in free energy

  Active spinning term: Derive from ?

- Equations of motion become

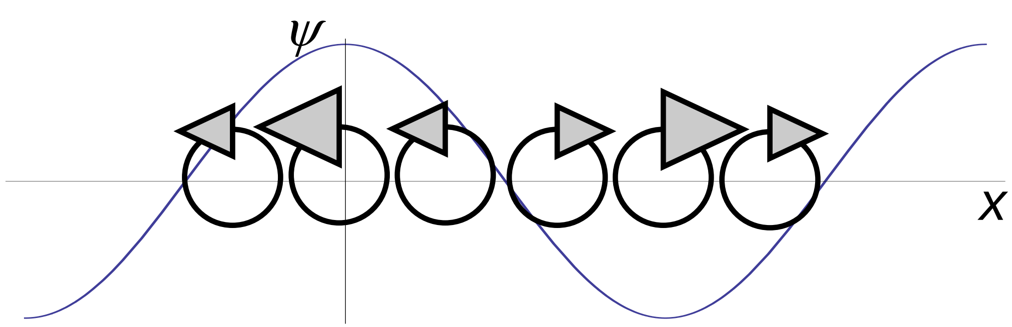
$$\frac{d\Omega}{dt} = -\ldots \Omega + \ldots \psi$$

$$\frac{d\psi}{dt} = \frac{\partial^2}{\partial x^2}\left[+\ldots \psi - \ldots \frac{\partial^2 \psi}{\partial x^2} - \ldots \Omega \right]$$

Case B solution

- In steady state for rotation: $\frac{d\Omega}{dt} = 0 \Rightarrow \Omega = \ldots\psi$
- Substitute into equation of motion for chiral composition $\psi(x,t)$
- When active coupling is big enough: $\frac{d\psi(x,t)}{dt} = \frac{\partial^2}{\partial x^2}\left[-\ldots\psi - \ldots\frac{\partial^2\psi}{\partial x^2}\right]$
- Fourier transform $x \to q$: $\frac{d\psi(q,t)}{dt} = \left(+\ldots q^2 - \ldots q^4\right)\psi(q,t)$
- Spinodal decomposition with favored wavevector

$$\psi(x,t) \sim \Omega(x,t) \sim e^{+\ldots t}\cos(qx)$$

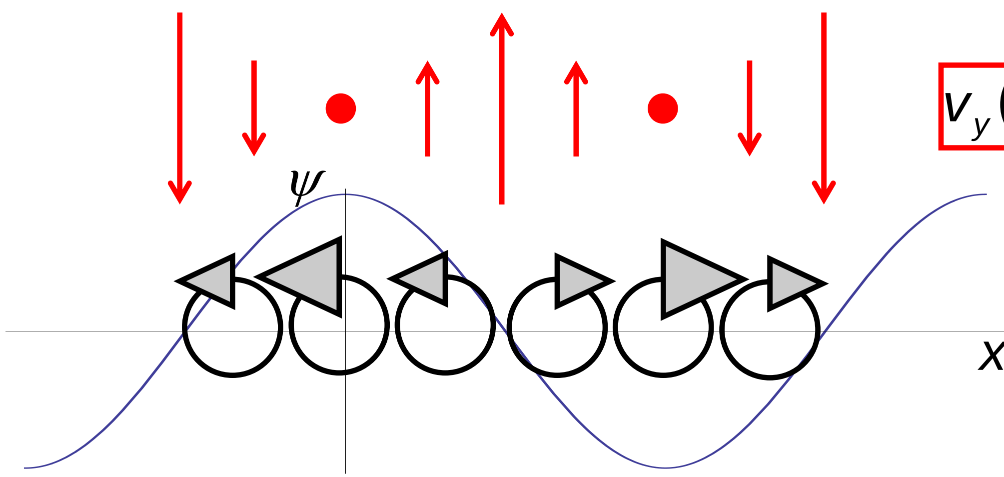


Case C

- Two-dimensional with conservation law for chiral particles and with flow velocity $v_y(x,t)$
- Equations of motion
- Similar solution as in case B, but with flow:

$$\begin{cases} \rho \dfrac{\partial v}{\partial t} - \eta \dfrac{\partial^2 v}{\partial y^2} + \dfrac{1}{2}\gamma_1 \dfrac{\partial}{\partial x}\left(\Omega - \dfrac{1}{2}\dfrac{\partial v_y}{\partial x}\right) = 0 \\ I \dfrac{\partial \Omega}{\partial t} + \gamma_1 \left(\Omega - \dfrac{1}{2}\dfrac{\partial v_y}{\partial x}\right) = \ldots \psi \\ \dfrac{\partial \psi}{\partial t} = \dfrac{\partial^2}{\partial x^2}\left[\ldots \psi - \ldots \dfrac{\partial^2 \psi}{\partial x^2} - \ldots \left(\Omega - \dfrac{1}{2}\dfrac{\partial v_y}{\partial x}\right)\right] \end{cases}$$

$$v_y(x,t) \sim e^{+\ldots t} \sin(qx)$$



Open questions

1. How to justify the way I introduced driving?
   How to relate the two driving coefficients?

2. What if the system had a first-order transition?
   How to model nucleation and growth of active system?