Spontaneous motion and deformation of a droplet

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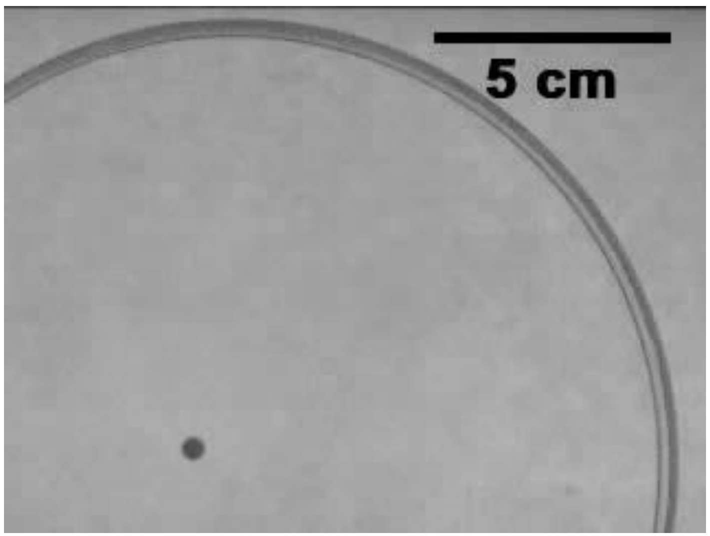
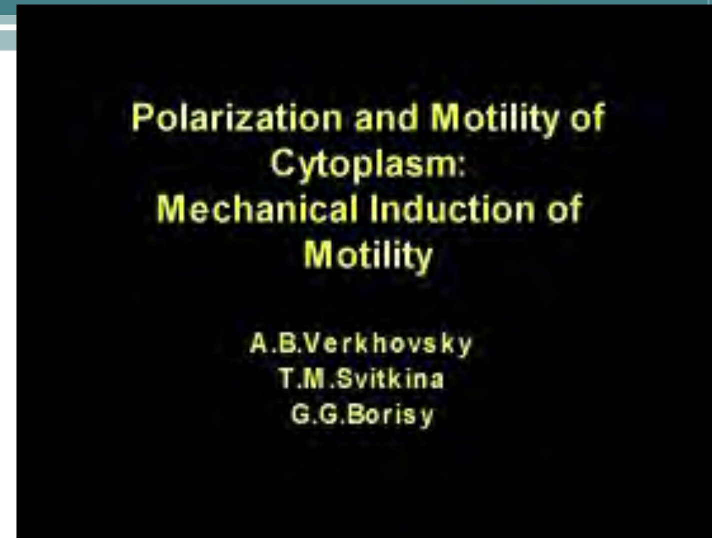
Motivation
- Motility and Shape -

pentanol drop



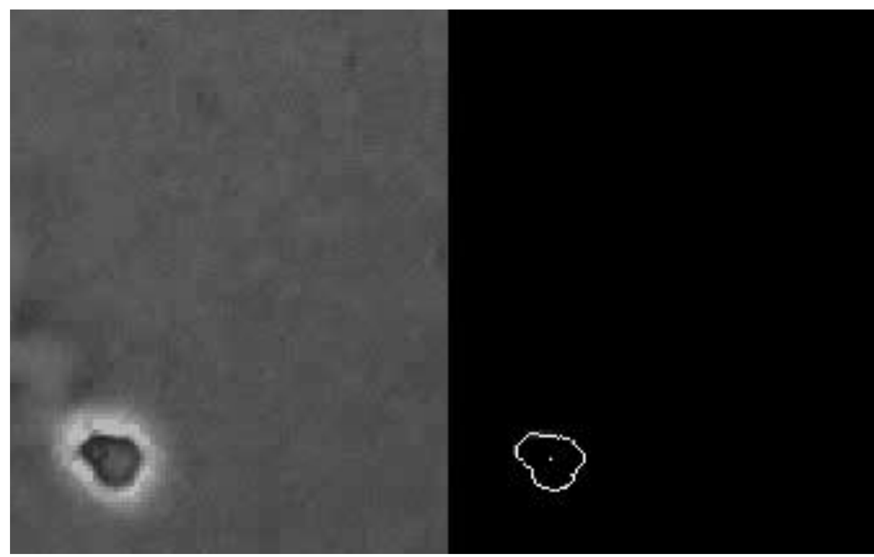
K. Nagai (2005)

keratocyte



A. Verkhovsky (1999)

Dictyostelium

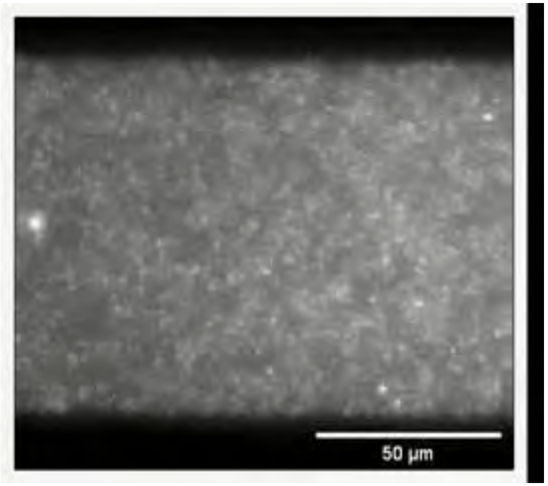


E. Cox, et al. (2008)
Y. Maeda and M. Sano (2008)

1. Force free
2. Symmetry breaking

Motion “without” external force

- Phoresis
  - Electrophoresis
  - Diffusiophoresis
  - Soret Effect (thermophoresis)



D. Braun et al. (2002)

- Marangoni Effect
  - liquid-liquid, gas-liquid interface

No mechanical force acting on the drop

$$f_z = \int \sigma \cdot \mathbf{n} dS = 0$$

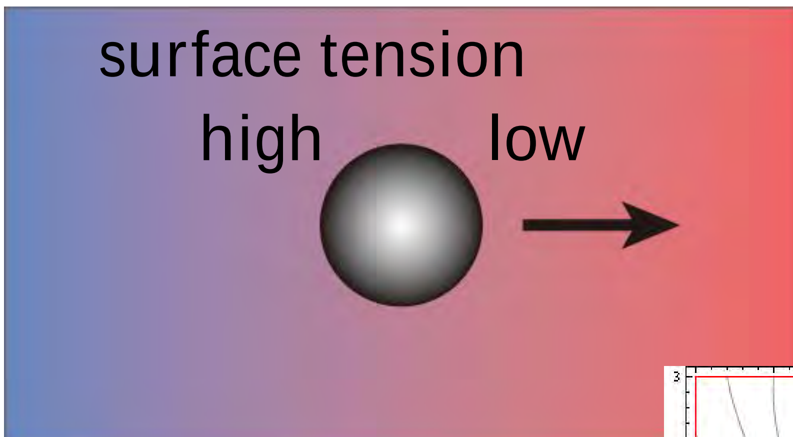
J. L. Anderson,(1989)

Motion of a droplet in a linear temperature gradient

- Drops move toward a hotter place

drop velocity

$$u = -\frac{2}{3(3\eta_i + 2\eta_o)}\gamma_T \nabla T$$

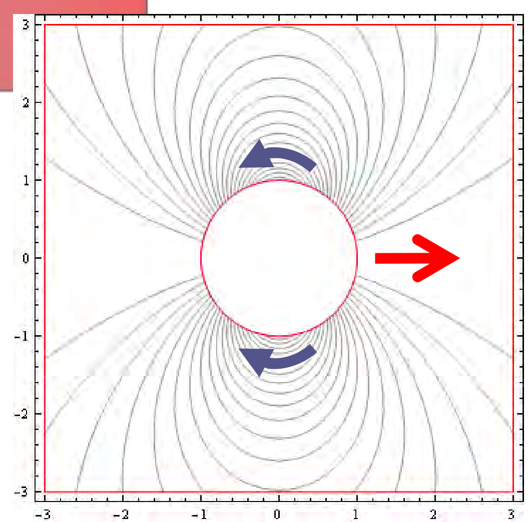


N. Young et al. (1959)

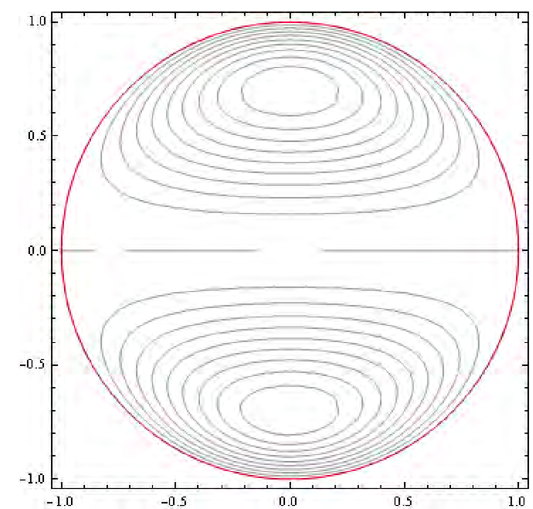
$$f_z = \int \sigma \cdot \mathbf{n} dS = 0$$

The drop is swimming in a fluid.

outer flow

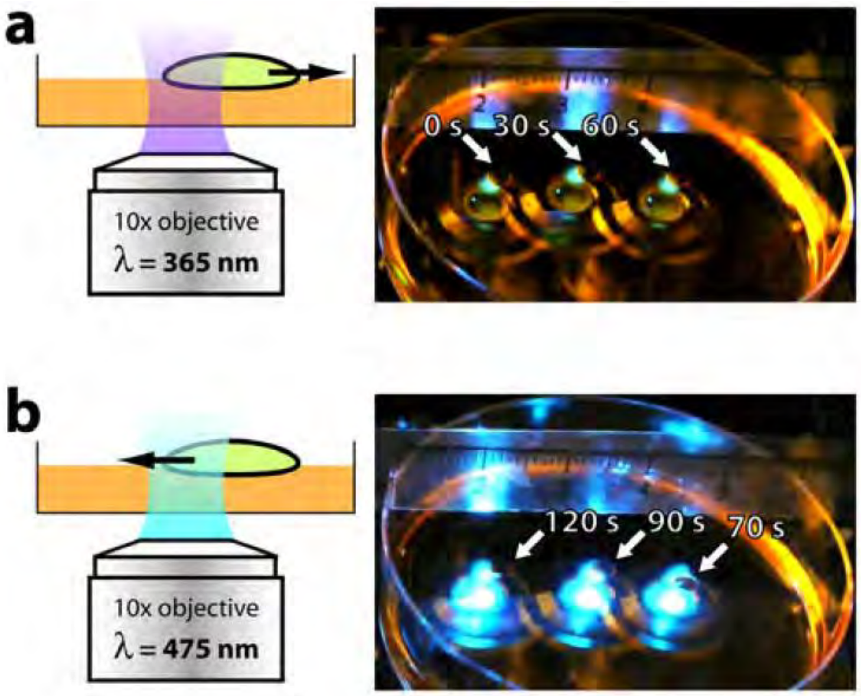


inner flow



Light-guided drop

Photo-sensitive surfactant

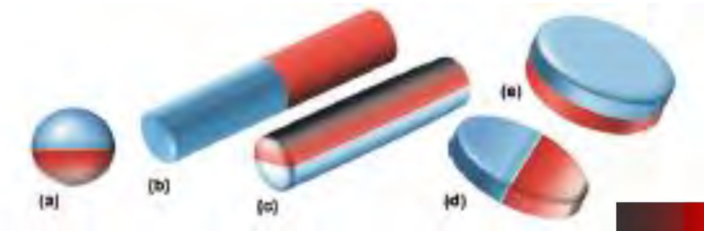


A. Diguet, et al. *Ang. Chim.* (2009)

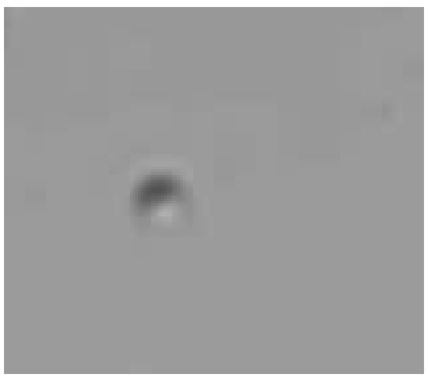
A drop partially illuminated by laser may have inhomogeneous distribution of surface tension due to photo-sensitive surfactants, which leads to motion.

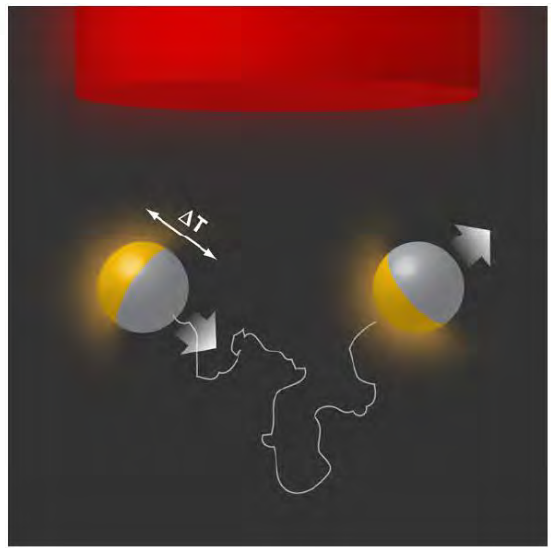
Active motion of Janus particles

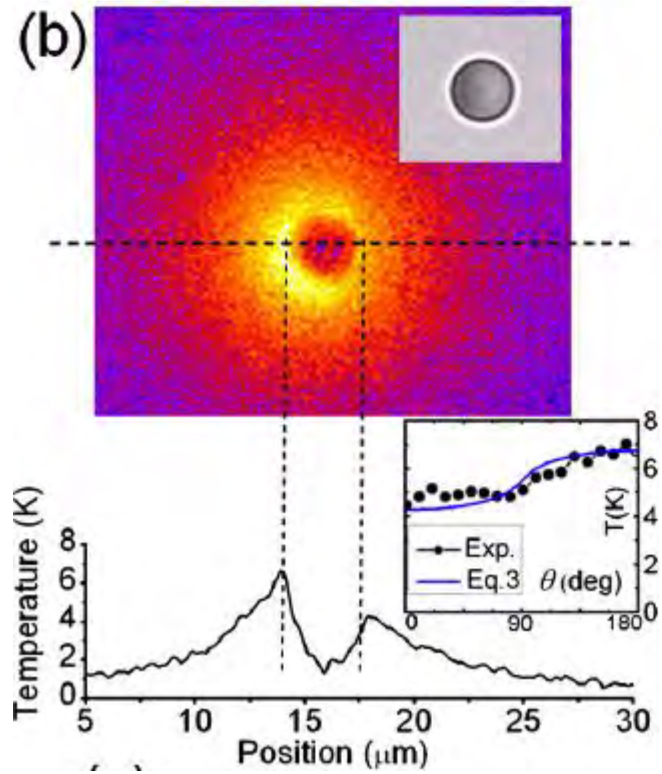
a particle with two faces



A. Walther,
Soft Matter (2008)







H.-R. Jiang, H. Wada, NY, and M. Sano. *PRL* (2009)

H.-R. Jiang, NY, and M. Sano. *PRL* (2010)

W. F. Paxton, JACS (2004)

J. R. Howse, PRL (2004)

$$u = -DS_T \frac{\Delta T}{3R}$$

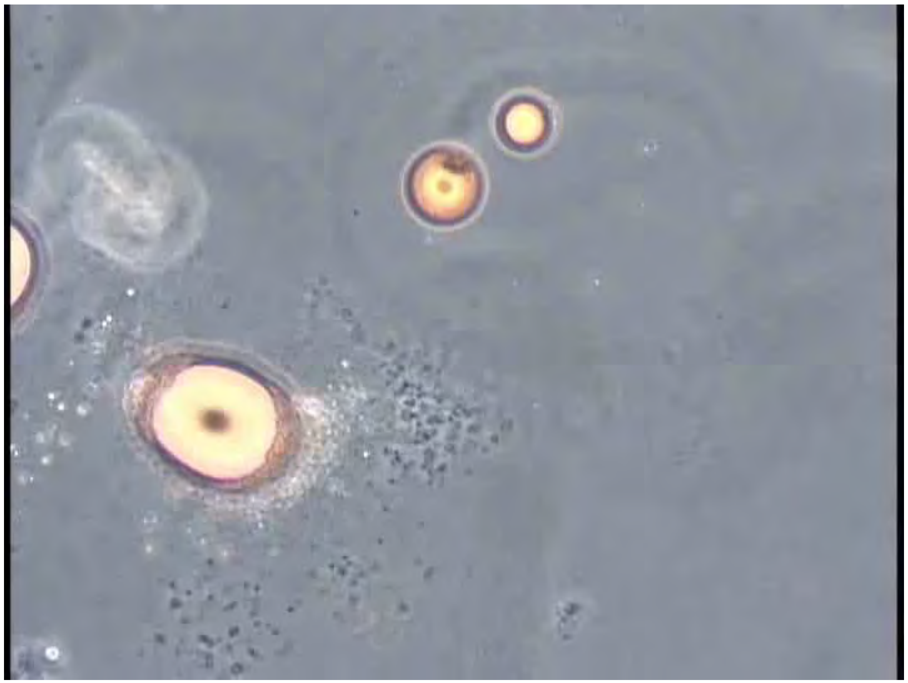
~ laser intensity

Spontaneous motion

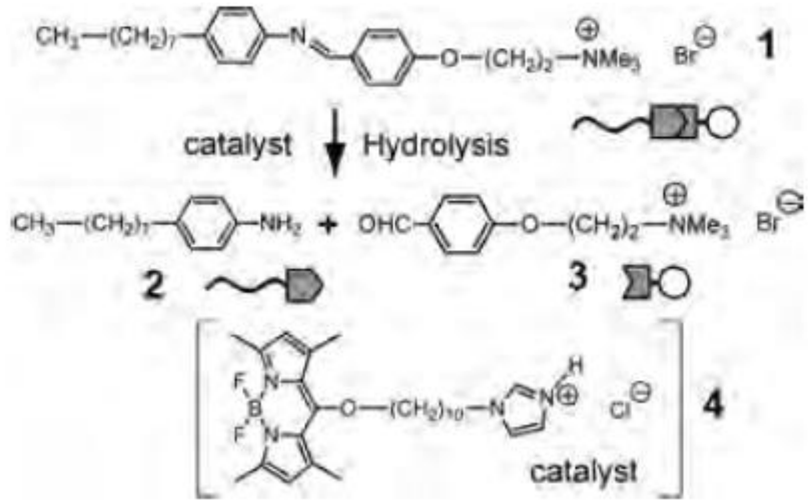
- Motion under an external field (gradient)
- Motion of an asymmetric particle under a uniform (isotropic) field
- Spontaneous symmetry breaking
  - Motion itself breaks symmetry (two isotropic fields have different centers)
  - Internal nonlinear pattern formation breaks symmetry

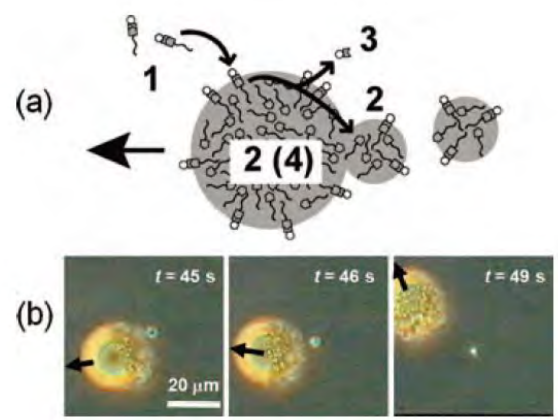
Spontaneous Symmetry Breaking

oil drop consuming surfactant



T. Toyota, *JACS* (2009)



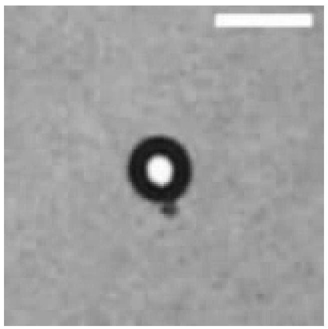


also S. Thutupalli *et al.*, *NJP* (2011)

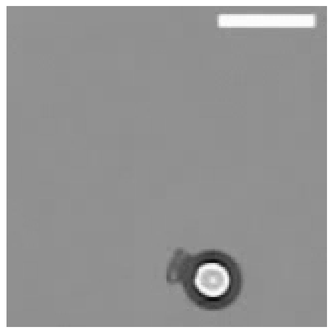
Spontaneous rotational motion

oil drop + solid soap

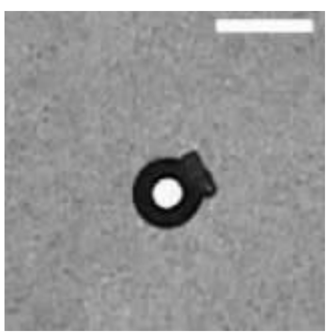
10 mm



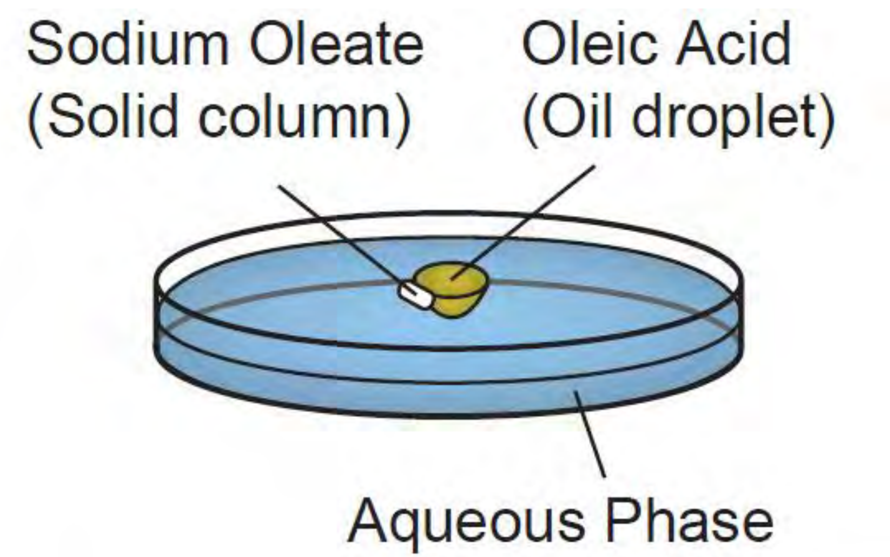
spinning



translation



rotation



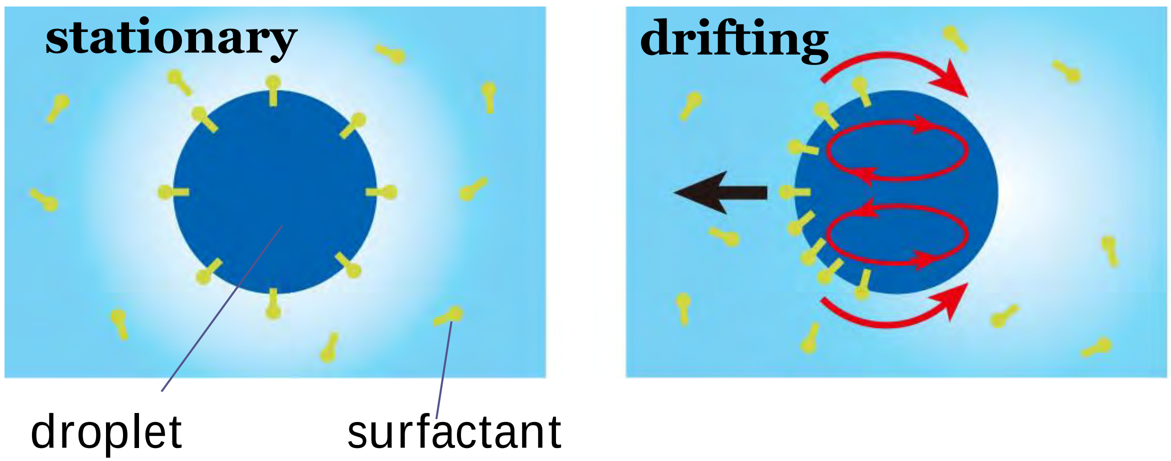
Experiments

F. Takabatake, *et al.*, *JCP* (2011)

Theory

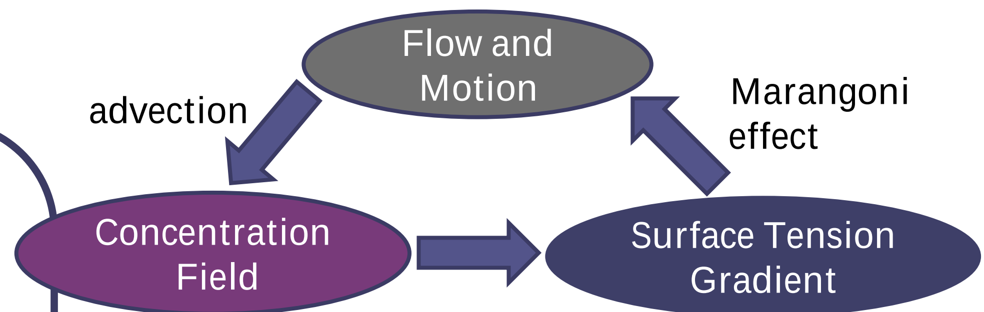
K.H. Nagai, F. Takabatake, Y. Sumino,
H. Kitahata, M. Ichikawa, NY (2013)

“Reactive” droplet



self-propulsion of
uniform systems

- reactive surfactant
- heating inside a drop
- production inside a drop



- Bulk Concentration Field

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c = D\nabla^2 c - \kappa (c - c_\infty)$$

- Hydrodynamics

$$-\nabla p + \eta \nabla^2 \mathbf{v} + \mathbf{f} = 0$$

- Surface Concentration Field

$$\frac{\partial \Gamma}{\partial t} + v_s \nabla_s \Gamma = D_s \nabla_s^2 c - k_s \Gamma \pm \left. \frac{\partial c}{\partial r} \right|_{r=R}$$

Amplitude Equation of Droplet Velocity

$$\frac{du}{dt} = (-1 + \tau) u - g u^3$$

NY, K.H. Nagai, Y. Sumino, H. Kitahata , PRE (2012)

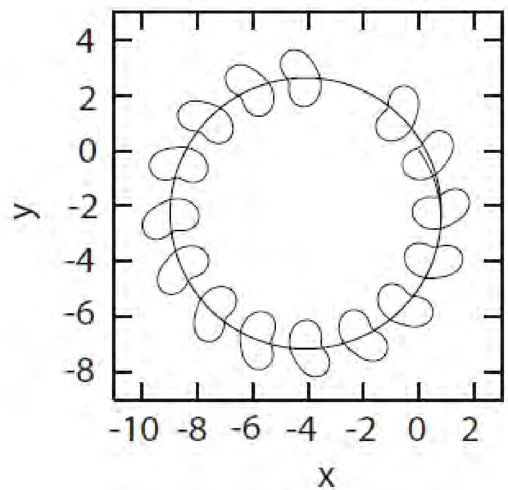
Phenomenological model for motion and deformation

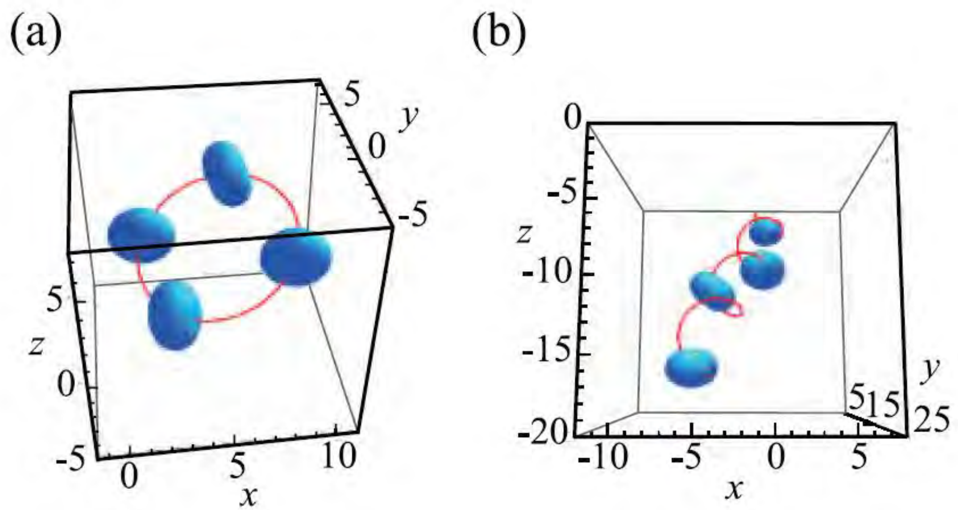
Motion

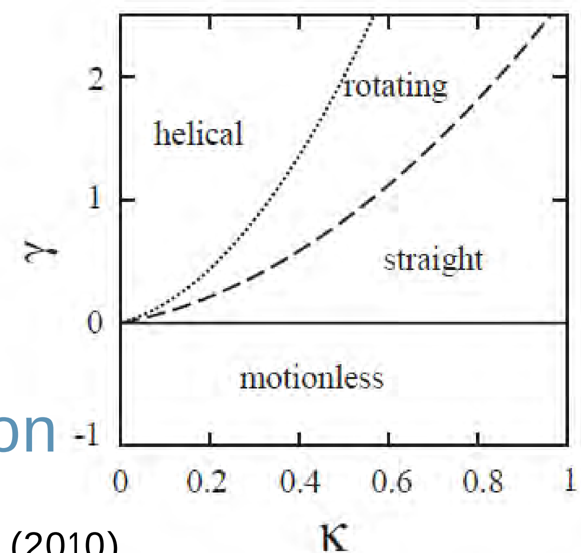
$$m\frac{\partial u_i}{\partial t} = (-1+\tau)u_i - gu^2u_i + bu_iS_{ij}$$

Deformation

$$\tau_2\frac{\partial S_{ij}}{\partial t} = -\kappa S_{ij} - \lambda\left(u_iu_j - \frac{1}{3}\delta_{ij}u^2\right)$$







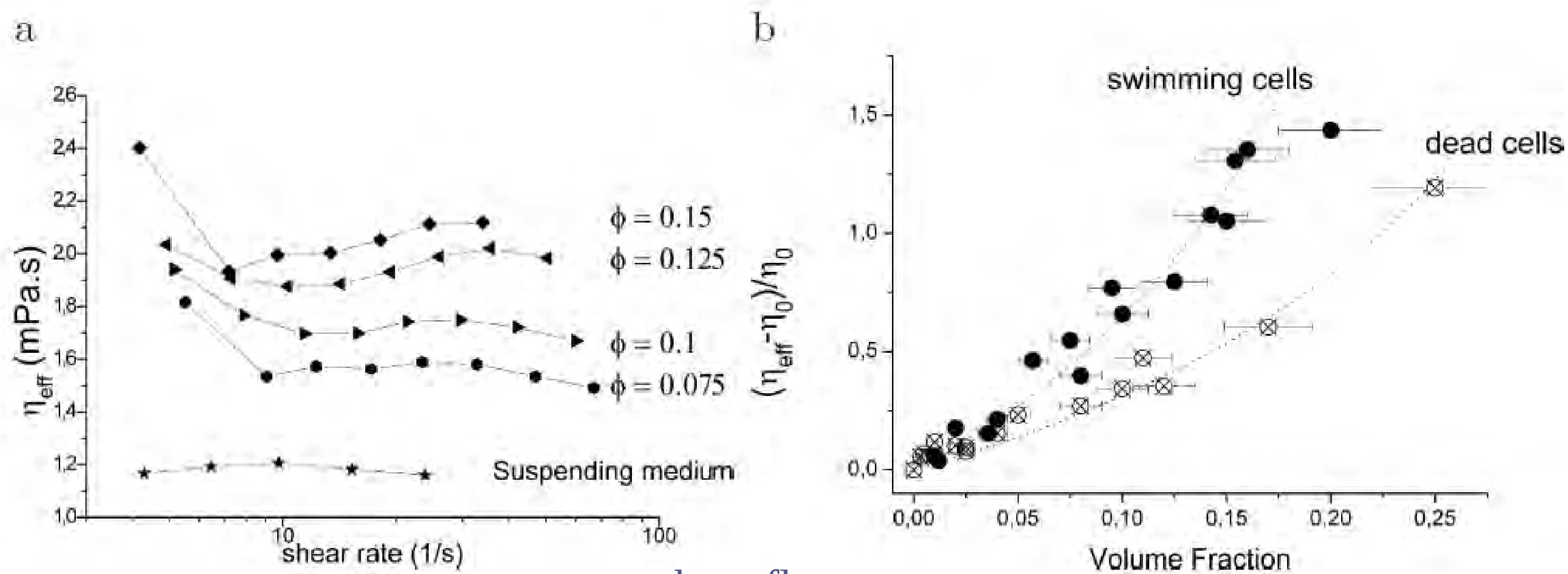
also derived from reaction-diffusion equation

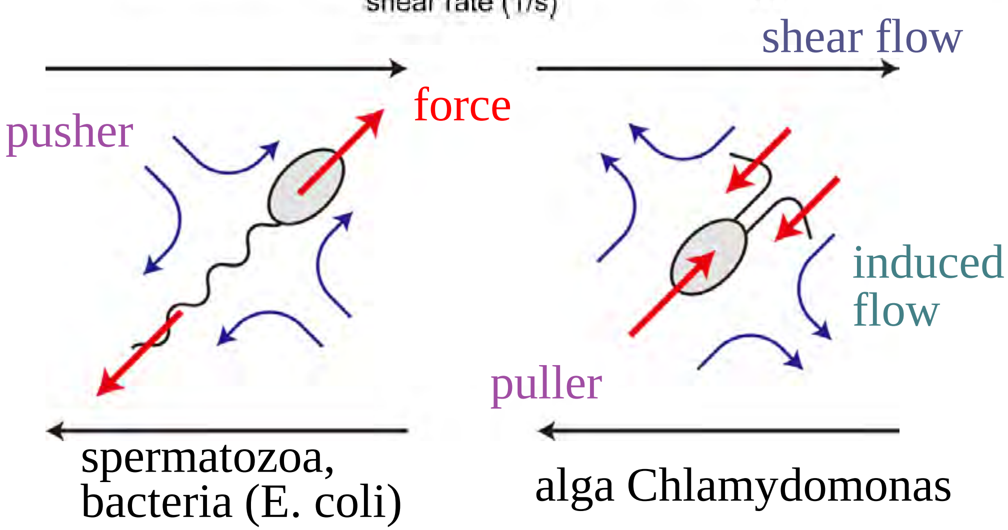
Mechanics ??

T. Ohkuma, and T. Ohta (2009)
T. Hiraiwa, K. Shitara, and T. Ohta (2010)
K. Shitara, T. Hiraiwa, and T. Ohta (2011)

Pusher or Puller?

Viscosity of *Chlamydomonas* suspension





P. Peyla, et al., (2010)
Effective viscosity of microswimmer suspensions

S. Ramaswamy, et al., (2004)
Rheology of Active-Particle Suspensions

Phase-field model

Dynamics of phase , $\phi(\mathbf{r})$

$$\frac{\partial \phi}{\partial t} + \nabla \cdot (\mathbf{v}\phi) = \nabla^2 \frac{\delta F}{\delta \phi}$$

$$F[\phi(\mathbf{r}), c(\mathbf{r})] = \int d^3 r \left[f_{GL}(\phi) + f_0(c) + \frac{1}{2} B(c) |\nabla \phi|^2 \right]$$

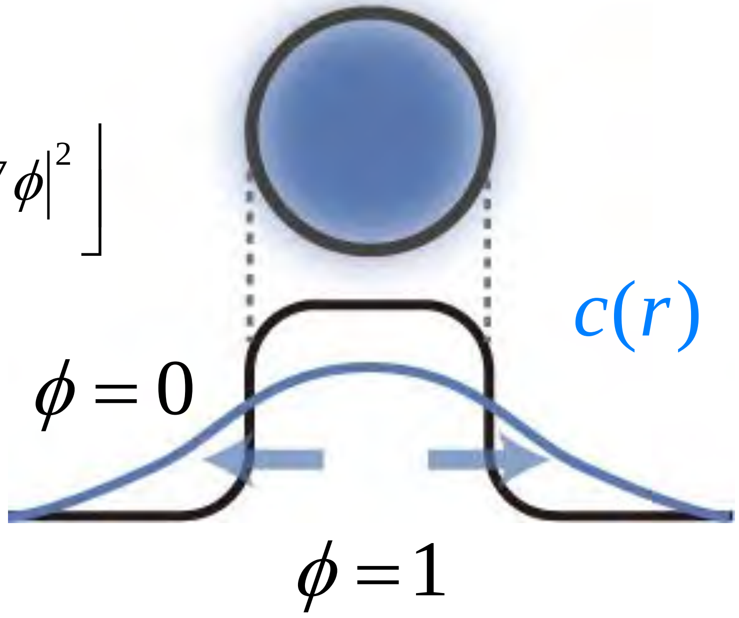
$$f_{GL}(\phi) = \frac{1}{4}\phi^2(1-\phi)^2$$

$$f_0(c) = c \ln c$$

$$B(c) = B_0 + B_1 c$$

surface tension

$$B[c(\mathbf{R}_s)] \simeq \gamma(\theta, \varphi)$$



S. Yabunaka, T. Ohta, and NY , J. Chem. Phys (2012)

Concentration Field

Dynamics of chemical

$$\frac{\partial c}{\partial t} + \nabla \cdot (\mathbf{v} c) = D\nabla^2 c - kc + A\Theta(R - |\mathbf{r} - \mathbf{r}_G|)$$

motion

production of chemicals

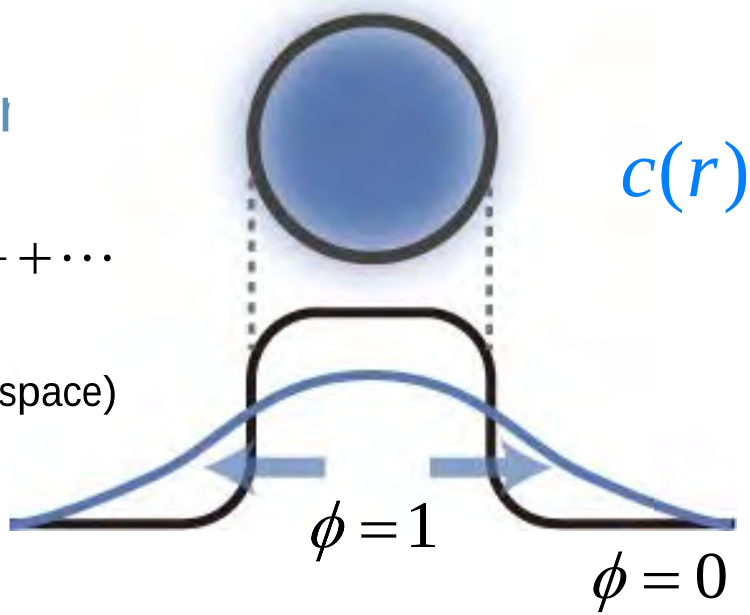
Velocity field, $\mathbf{v}(\mathbf{r})$ $\eta \nabla^2 \mathbf{v} - \nabla p - \mathbf{f} = 0$

Perturbation around drift bifurcatio

$$c_{\mathbf{q}} = \frac{G_q}{D} H_{\mathbf{q}} - \frac{G_q^2}{D^2}\frac{\partial H_{\mathbf{q}}}{\partial t} + \frac{G_q^3}{D^3}\frac{\partial^2 H_{\mathbf{q}}}{\partial^2 t} - \frac{G_q^4}{D^4}\frac{\partial^3 H_{\mathbf{q}}}{\partial^3 t} + \cdots$$

$H_{\mathbf{q}}$: source (in Fourier space)

G_q: Green's functin



Expansion in terms of

$$\epsilon \sim u = \dot{r}_G$$

S. Yabunaka, T. Ohta, and NY , J. Chem. Phys (2012)

Hydrodynamics

low-Reynolds Stokes flow

$$\eta\nabla^2\mathbf{v} - \nabla p - \nabla\cdot\Pi = 0$$

sharp-interface limit

$$\mathbf{f} = \mathbf{f}_s\delta(r - R)$$

→ boundary-value problem

$$\eta\nabla^2\mathbf{v} - \nabla p = 0$$

$$\mathbf{v}_{\text{in}}(R) = \mathbf{v}_{\text{out}}(R)$$

$$\mathbf{n}\cdot\boldsymbol{\sigma}_{\text{in}}(R) - \mathbf{n}\cdot\boldsymbol{\sigma}_{\text{out}}(R) = \partial_s\left[\gamma(\theta)\mathbf{t}\right]$$

conventional Marangoni effect

$$c(\mathbf{r}) = c_0 + c_1 z \longrightarrow u = -\frac{2\gamma_C c_1 R}{15\eta}$$

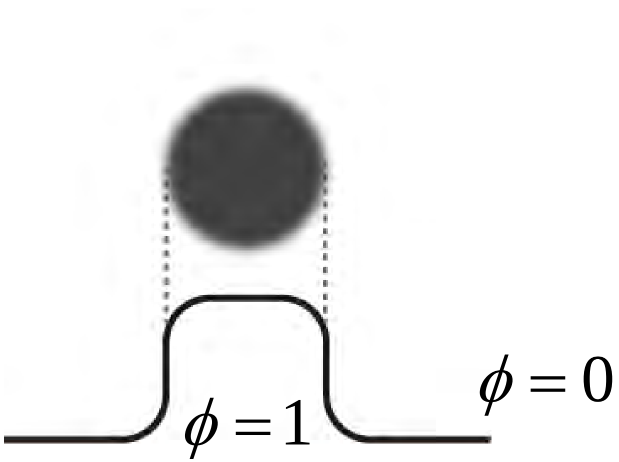
Korteweg stress

$$\Pi_{ij} = -B(c)\nabla_i\phi\nabla_j\phi$$

$$B(c) = B_0 + B_1 c$$

$$B\left[c(\mathbf{R}_s)\right] \sim \gamma(\theta,\varphi)$$

surface tension



Interfacial force

Force acting on an interface

$$\mathbf{f} = \mathbf{f}_s \delta(r - R)$$

$$\mathbf{f}_s = -\kappa\gamma(a)\mathbf{n} + \nabla_s \gamma(a)\mathbf{t}$$

curvature

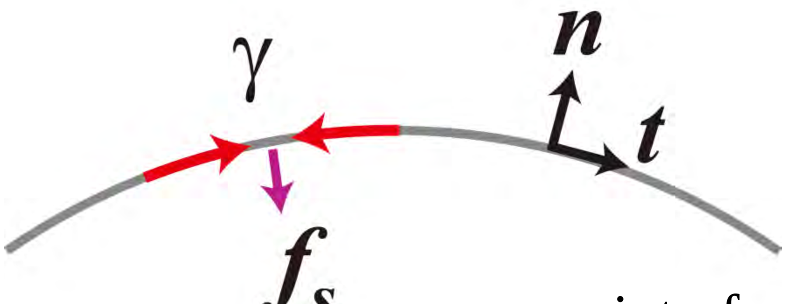
Laplace pressure

small R

large R

inhomogeneous surface tension

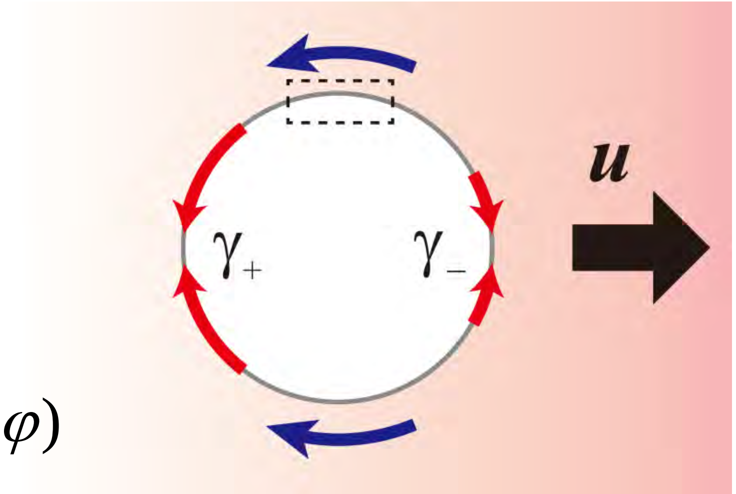
$$\gamma(a) = \sum_{l=0}^{\infty} \sum_{m=-l}^{l} \gamma_{lm} Y_l^m(a)$$



$a = (\theta, \varphi)$

interface

Marangoni force



Force moments

Mutipole expansion of flow field

$$v_i = \mathrm{T}_{ij} F_j - \mathrm{T}_{ij,k} D_{jk} + \frac{1}{2} \mathrm{T}_{ij,kl} G_{jkl} + \cdots$$

monopole (Stokeslet)

$$F_i = \int f_i dV = 0 \quad \longrightarrow \quad \mathbf{v} \sim \frac{1}{r} \cos\theta$$

dipole (stress let)

$$D_{ij} = \int f_i x_j dV = \gamma_2 \begin{pmatrix} -1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 2 \end{pmatrix} \quad \longrightarrow \quad \mathbf{v} \sim \frac{\gamma_2}{r^2} P_2(\cos\theta)$$

deformation

$\gamma_2 < 0$: puller

$\gamma_2 > 0$: pusher

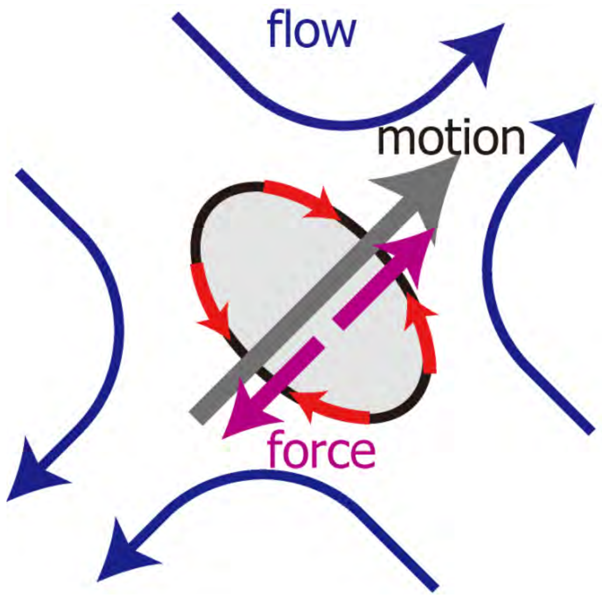
quadrupole

$$G_{ijk} = \int f_i x_j x_k dV \quad \longrightarrow \quad \mathbf{v} \sim \frac{\gamma_1}{r^3} \cos\theta + \mathcal{O}(P_3)$$

motion (F. Jülicher and J. Prost, 2009)

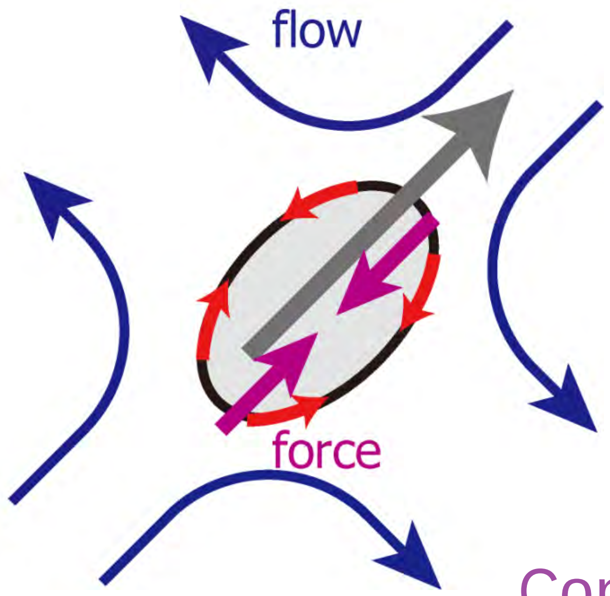
Shape and Force moment

pusher



$$\gamma_2 > 0$$

puller



$$\gamma_2 < 0$$

Concentration

$$\gamma = \gamma_c c$$

Surface tension

Motion

Velocity field

Oseen Tensor

$$\mathbf{v}(\mathbf{r}) = \gamma_c \int dS \left[\mathrm{T}(\mathbf{r},\mathbf{r}')\cdot\mathbf{n}(a')c(a')\kappa(a') + \mathrm{T}(\mathbf{r},\mathbf{r}')\cdot\left(\mathrm{I}-\mathbf{n}(a')\mathbf{n}(a')\right)\nabla c(a') \right]$$

Laplace pressure

tangential force

Velocity of a droplet

$$\mathbf{u} = \frac{1}{V}\int v_n \mathbf{R} dS$$

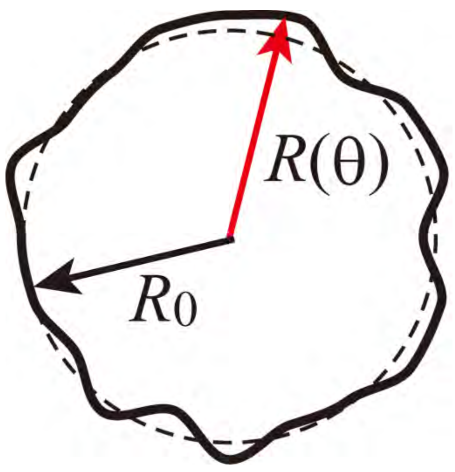
Kinematic condition

$$v_n = \mathbf{u}\cdot\mathbf{n} + \frac{d\delta R(a,t)}{dt}$$

Shape of the droplet

$$R(\theta,\varphi) = R_0 + \delta R(\theta,\varphi)$$

$$\delta R(\theta,\varphi) = \sum_{l=2}^{\infty}\sum_{m=-l}^{l} w_{l,m} Y_l^m(\theta,\varphi)$$



Deformation

Order parameter tensor

$$R_{ijkl\cdots} = \int \delta R \left[n_i^{(0)} n_j^{(0)} n_k^{(0)} n_l^{(0)} \cdots \right]_{\substack{\text{symm}\\ \text{traceless}}} dS$$

Kinematic condition

$$\frac{d\delta R(a,t)}{dt} = \mathbf{u}\cdot\mathbf{n} - v_n$$

$$\times\left(n_i n_j - \frac{1}{3}\delta_{ij} \right) \longrightarrow \frac{dS_{ij}}{dt} = \ldots\ldots$$ second mode

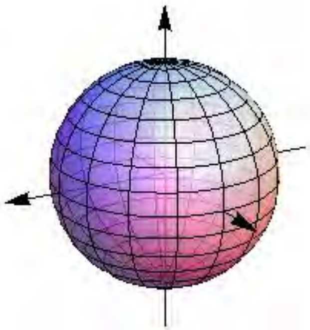
$$\times\left(n_i n_j n_k - \frac{1}{5}\left(\delta_{ij} n_k + \delta_{jk} n_i + \delta_{ik} n_j \right) \right) \longrightarrow \frac{dT_{ijk}}{dt} = \ldots\ldots$$ third mode

Deformation

Second mode

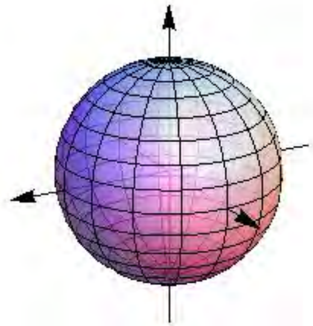
$S_{xx} - S_{yy}$

$w_{2,2} + w_{2,-2}$



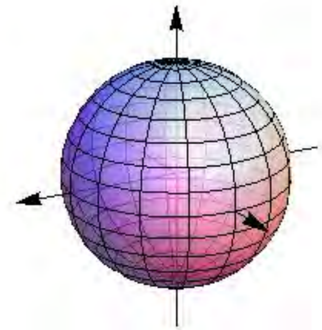
$\frac{1}{2}\left(S_{xy} + S_{yx}\right)$

$i\left(w_{2,2} - w_{2,-2}\right)$



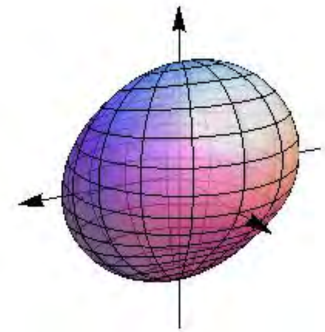
$\frac{1}{2}\left(S_{xz} + S_{zx}\right)$

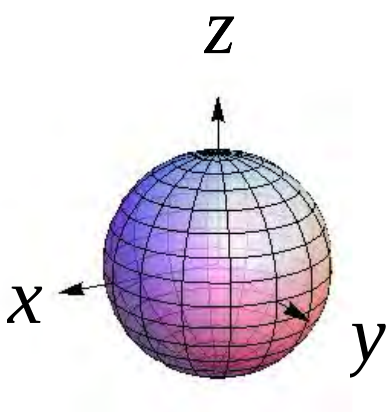
$w_{2,1} - w_{2,-1}$



$\frac{1}{2}\left(S_{yz} + S_{zy}\right)$

$i\left(w_{2,1} + w_{2,-1}\right)$





S_{zz}

$w_{2,0}$

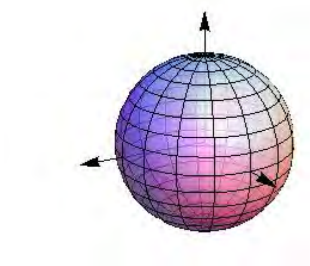
5 independent modes

Deformation

Third mode

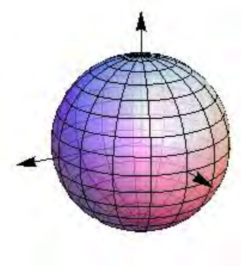
$$T_{zzz} - \frac{1}{3}\left(T_{xxz} + T_{xzx} + T_{zxx}\right) - \frac{1}{3}\left(T_{yyz} + T_{yzy} + T_{zyy}\right)$$

$$w_{3,0}$$



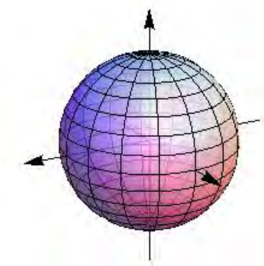
$$T_{xxx} - T_{xyy} - T_{yxy} - T_{yyx}$$

$$w_{3,3} - w_{3,-3}$$



$$T_{yyy} - T_{yxy} - T_{yyx} - T_{xyy}$$

$$i\left(w_{3,3} + w_{3,-3}\right)$$



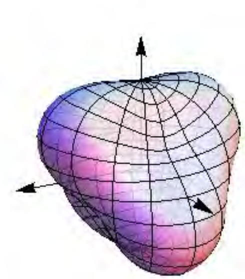
7 independent modes

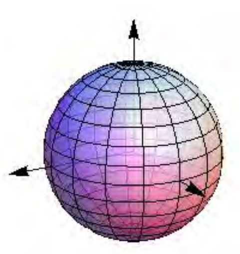
$$T_{xyz} + T_{xzy} + T_{yxz} + T_{yzx} + T_{zxy} + T_{zyx}$$

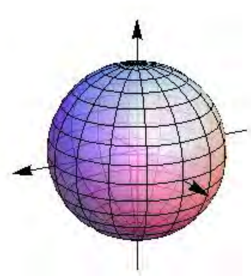
$$i\left(w_{3,2} - w_{3,-2}\right)$$

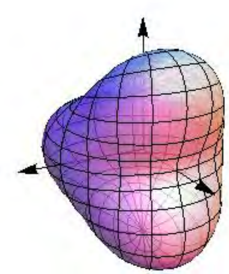
$$\frac{1}{3}\left(T_{yzz} + T_{zyz} + T_{zzy}\right) - \frac{1}{4}T_{yyy} - \frac{1}{12}\left(T_{yxx} + T_{xyx} + T_{xxy}\right)$$

$$i\left(w_{3,1} + w_{3,-1}\right)$$









$$T_{zxx} - T_{zyy} + T_{xzx} - T_{yzy} + T_{xxz} - T_{yyz}$$

$$w_{3,2} + w_{3,-2}$$

$$\frac{1}{3}\left(T_{xzz} + T_{zxz} + T_{zzx}\right) - \frac{1}{4}T_{xxx} - \frac{1}{12}\left(T_{xyy} + T_{yxy} + T_{yyx}\right)$$

$$w_{3,1} - w_{3,-1}$$

Motion and Deformation

First mode

$$m\frac{\partial u_i}{\partial t} = (-1+\tau)u_i - gu^2u_i + bu_iS_{ij} + \mathcal{O}(\epsilon^4)$$

Second mode

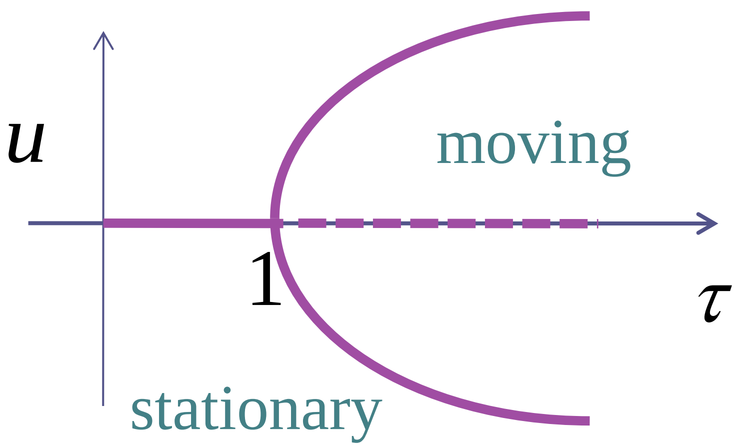
$$\tau_2\frac{\partial S_{ij}}{\partial t} = -\kappa S_{ij} + \lambda\left(u_iu_j - \frac{1}{3}\delta_{ij}u^2\right) + \mathcal{O}(\epsilon^4)$$

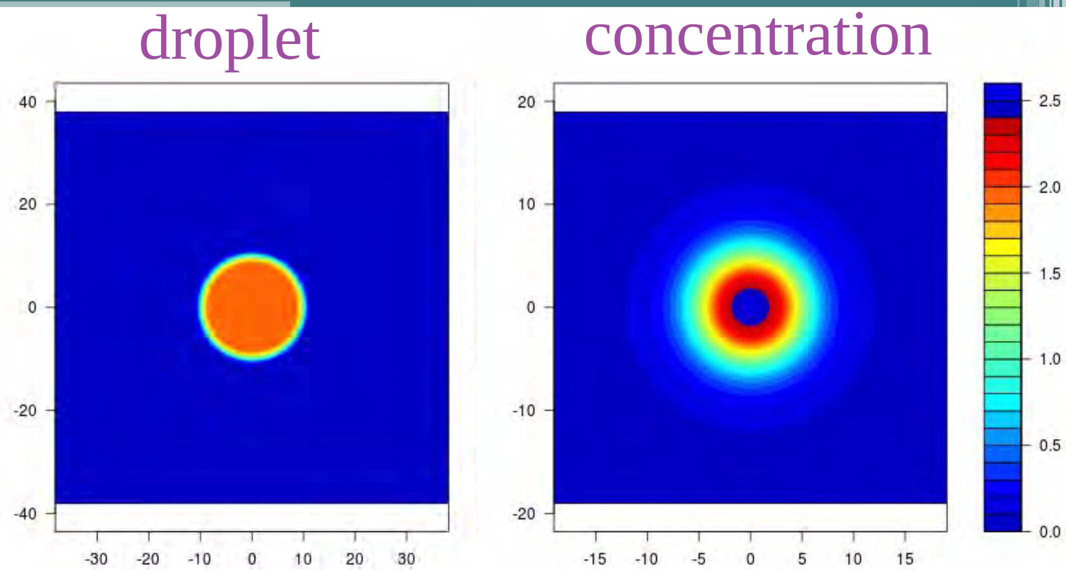
Third mode

$$\tau_3\frac{\partial T_{ijk}}{\partial t} = -\kappa_3T_{ijk} + \lambda_3\left[u_iu_ju_k - \frac{1}{5}u^2\left(u_i\delta_{jk} + u_j\delta_{ik} + u_k\delta_{ij}\right)\right]$$
$$+b_3\left[\left(S_{jk}u_i + S_{ik}u_j + S_{ij}u_k\right) - \frac{2}{5}u_\alpha\left(S_{j\alpha}\delta_{ik} + S_{k\alpha}\delta_{ij} + S_{i\alpha}\delta_{jk}\right)\right] + \mathcal{O}(\epsilon^4)$$

Drift bifurcation

pitchfork bifurcation





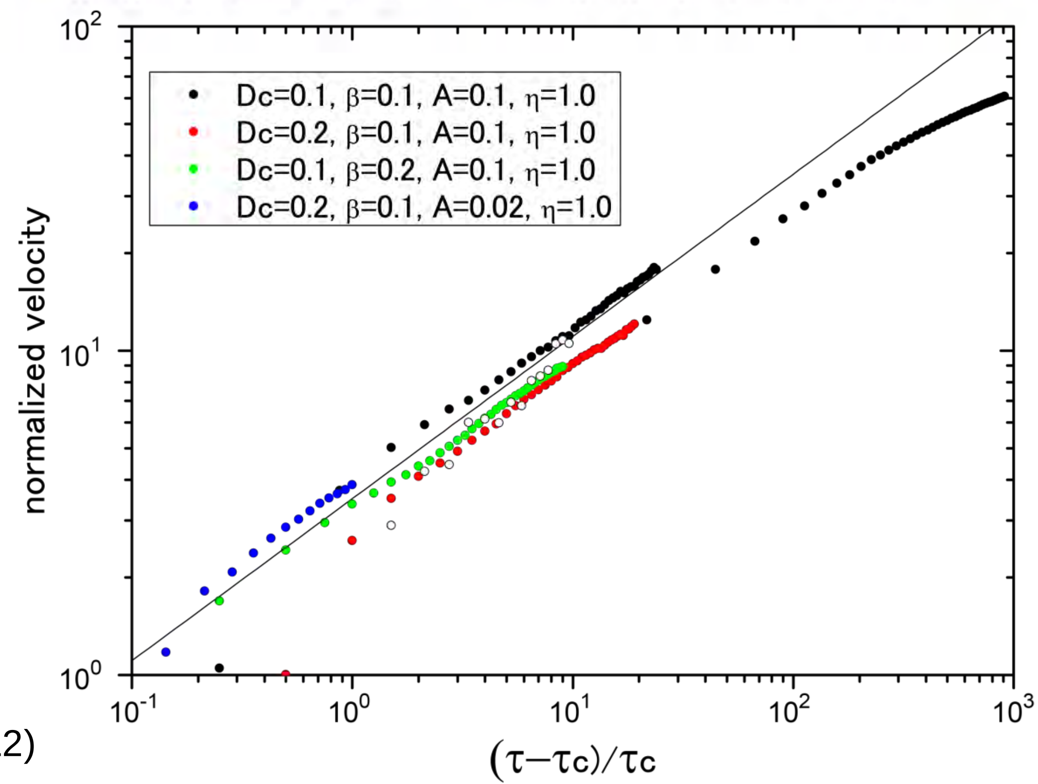
nondimensional activity

$$\frac{\tau}{\tau^*} = F_\tau(\beta R) \qquad \tau^* = \frac{\gamma_c A}{\eta D^2 \beta^3}$$

$$\beta \sim \sqrt{\frac{k}{D}}$$

stationary velocity

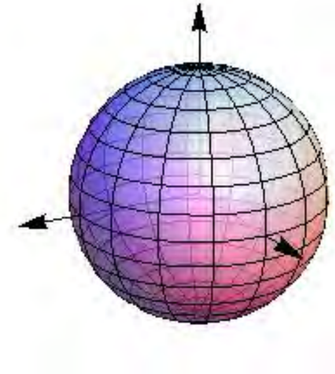
$$u \sim \sqrt{kD}\, F_u(\beta R)$$



S. Yabunaka, T. Ohta, and NY , J. Chem. Phys (2012)

Deformation

$S_{zz} > 0$



First mode

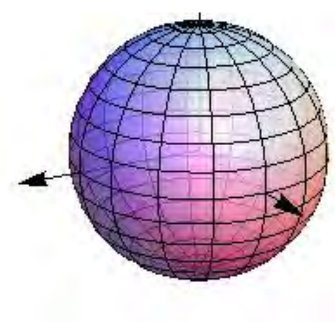
$$m\frac{\partial u_i}{\partial t} = (-1+\tau)u_i - gu^2u_i + bu_iS_{ij}$$

motion

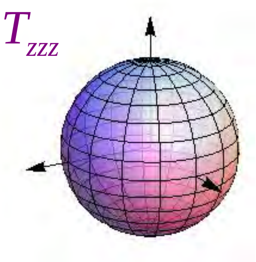
Second mode

$$\tau_2 \frac{\partial S_{ij}}{\partial t} = -\kappa S_{ij} + \lambda\left(u_iu_j - \frac{1}{3}\delta_{ij}u^2\right)$$

$S_{zz} < 0$



T_{zzz}



le

$$+\lambda_3\left[u_iu_ju_k - \frac{1}{5}(u_i\delta_{jk} + u_j\delta\right.$$

$$\left. {}_ku_i + S_{ik}u_j + S_{ij}u_k) - \frac{2}{5}u_\alpha\left(S_{j\alpha}\delta_{ik} + S_{k\alpha}\delta_{ij} + S_{i\alpha}\delta_{jk}\right)\right]$$

The drop is pusher

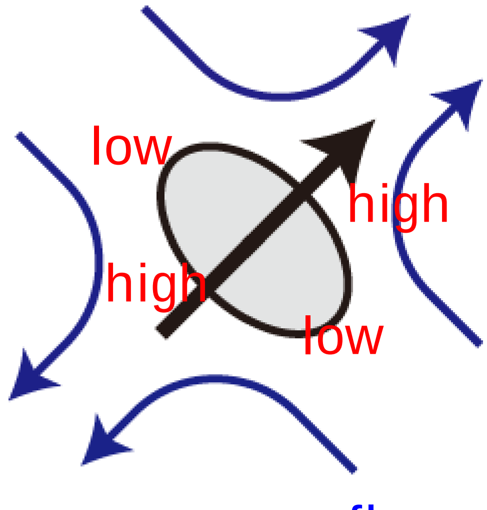
Second mode

$$\tau_2 \frac{\partial S_{ij}}{\partial t} = -\kappa S_{ij} + \lambda \left(u_i u_j - \frac{1}{3}\delta_{ij} u^2 \right)$$

Stationary shape

$$S_{ij} = \frac{\lambda}{\kappa}\left(u_i u_j - \frac{1}{3}\delta_{ij} u^2 \right) \longrightarrow \gamma_2 > 0$$

surface tension



low conc.

low conc.

NY , Phys. Rev. E (2014)

Lyapunov function

$$E = (1-\tau)u^2 + gu^4 + S_{ij}\left(u_i u_j - \frac{1}{3}u^2\delta_{ij}\right)$$

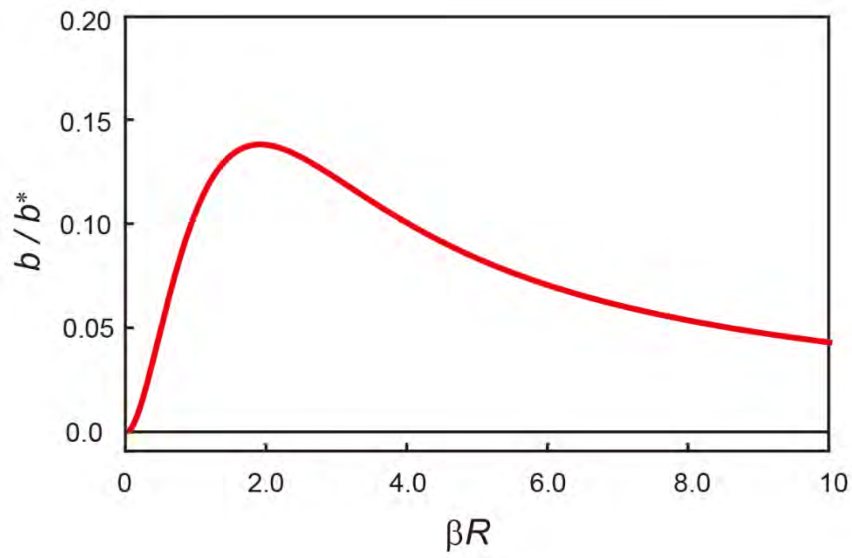
First mode

$$m\frac{\partial u_i}{\partial t} = -\frac{\partial E}{\partial u_i}$$

Second mode

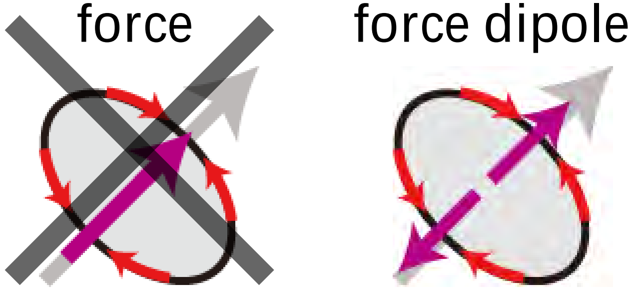
$$\tau_2 \frac{\partial S_{ij}}{\partial t} = -\frac{\partial E}{\partial S_{ij}}$$

Lyapunov function exists when $b\lambda > 0$



There is no Lyapunov function

Active Stress



Kortweg Stress

$$\Pi_{ij} = -B(c)\nabla_i \phi \nabla_j \phi$$

Active Stress

$$\sigma_a = \frac{1}{R_0^3}\int_\Omega \Pi_a \simeq -\frac{\gamma_c}{R_0}\int c(\mathbf{r}) n_i n_j dS$$

$$\longrightarrow \quad \sigma_a \sim \lambda\left(u_i u_j - \frac{1}{3}\delta_{ij} u^2\right)$$

$$D \sim 10^{-3}\,\mathrm{mm}^2/\mathrm{s}$$

$$R_0 \sim \beta^{-1} \sim 100\,\mu\mathrm{m}$$

$$\gamma_c A \sim 1\,\mathrm{mN/m{\cdot}s}$$

$$\sigma_a \sim 100\,\mathrm{Pa}$$

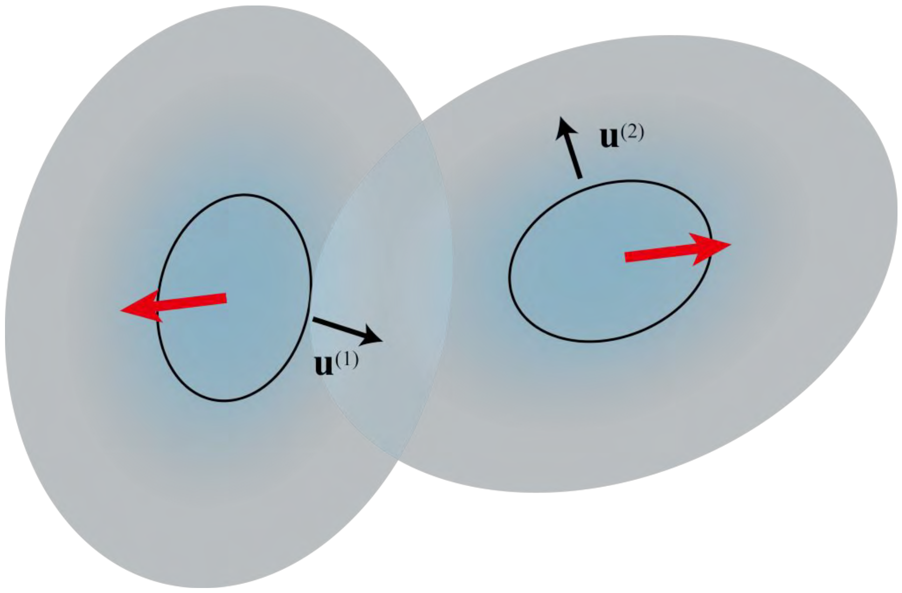
NY , Phys. Rev. E (2014)

Interaction

Shape of the droplet

$$R_i(\theta,\varphi) = R_0 + \delta R_i(\theta,\varphi)$$

$$\delta R_i(\theta,\varphi) = \sum_{l=2}^{\infty} \sum_{m=-l}^{l} w_{l,m}^{(i)} Y_l^m$$



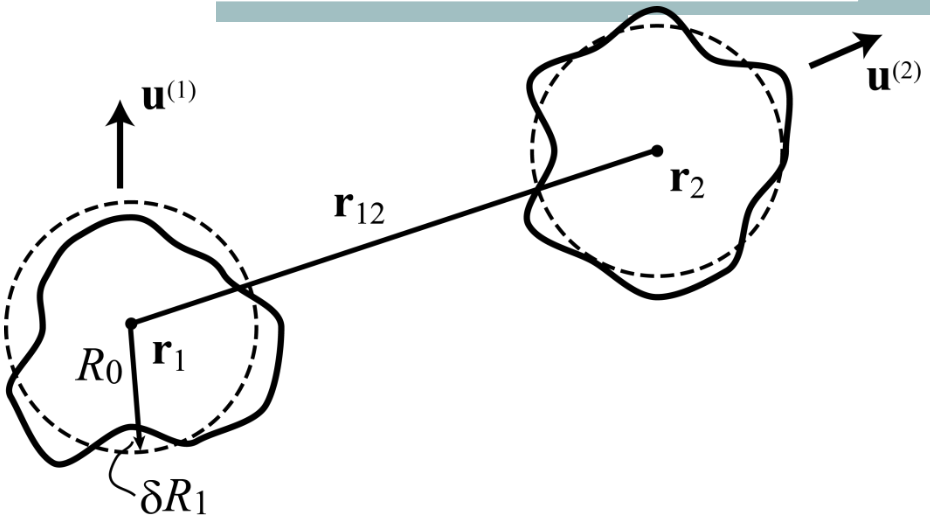
Dynamics of chemical

$$\frac{\partial c}{\partial t} = D\nabla^2 c - \kappa c + \sum_i A_i \Theta(R_i - |\mathbf{r} - \mathbf{r}_{G,i}|)$$

velocity of the droplet

$$m\frac{\partial u_i^{(1)}}{\partial t} = (-1+\tau)u_i^{(1)} - g\left|u^{(1)}\right|^2 u_i^{(1)} + b u_j^{(1)} S_{ij}^{(1)} + u_i^{\mathrm{int}}$$

Interaction



velocity of the droplet

$$m\frac{\partial u_i^{(1)}}{\partial t} = (-1+\tau)u_i^{(1)} - g\left|u^{(1)}\right|^2 u_i^{(1)} + bu_j^{(1)}S_{ij}^{(1)} + u_i^{\text{int}}$$

$$u_i^{\text{int}} = \nabla_{r_{i,1}}U(r_{12}) + F_1(r_{12})N_j S_{ij}^{(2)} + F_3(r_{12})N_{ijk}S_{jk}^{(2)} + \cdots$$

interaction between spherical droplets

interaction between deformed droplets

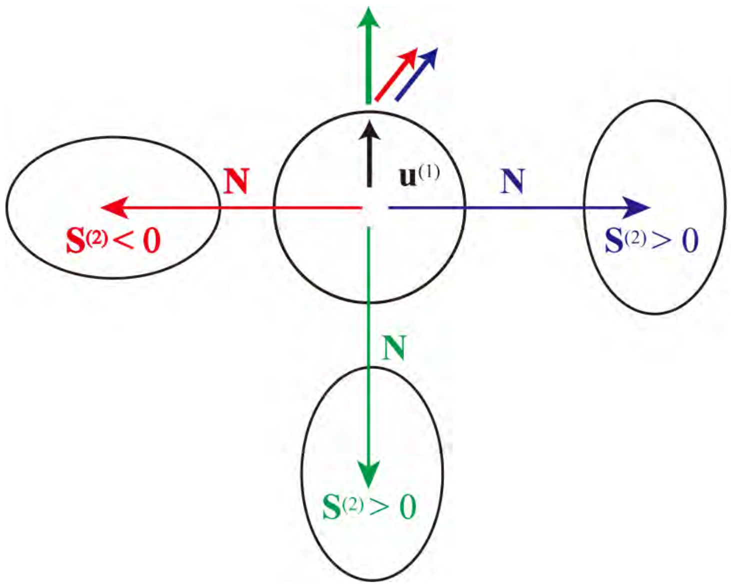
$$U(r_{12}) = \frac{A^{(2)}\gamma_c^{(1)}}{\eta D\beta^3} g(\beta R_0)k_0(\beta r_{12}) \sim \frac{e^{-\beta r_{12}}}{r_{12}}$$

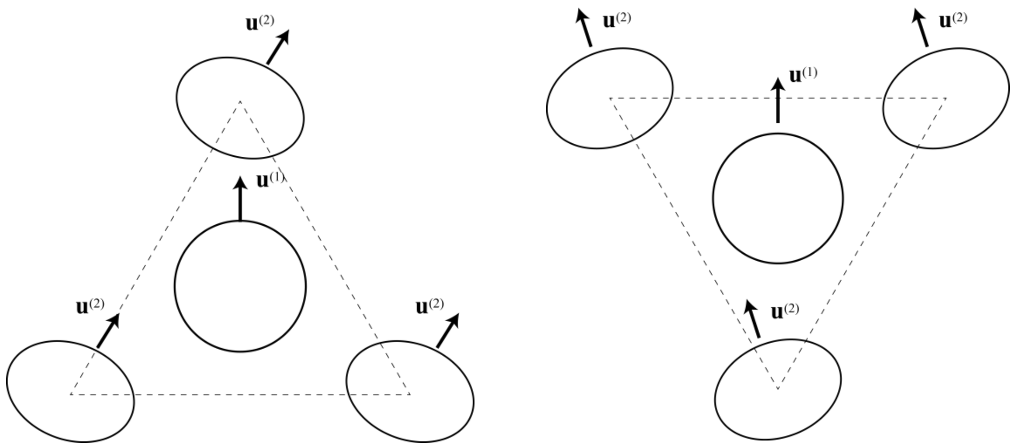
$$\begin{cases} A^{(1)}A^{(2)} > 0 & \text{repulsive} \\ A^{(1)}A^{(2)} < 0 & \text{attractive} \end{cases}$$

Interaction

$$u_i^{\text{int}} \sim F_1(r_{12}) N_j S_{ij}^{(2)}$$

$$u_i^{\text{int}} \sim F_3(r_{12}) N_{ijk} S_{jk}^{(2)}$$

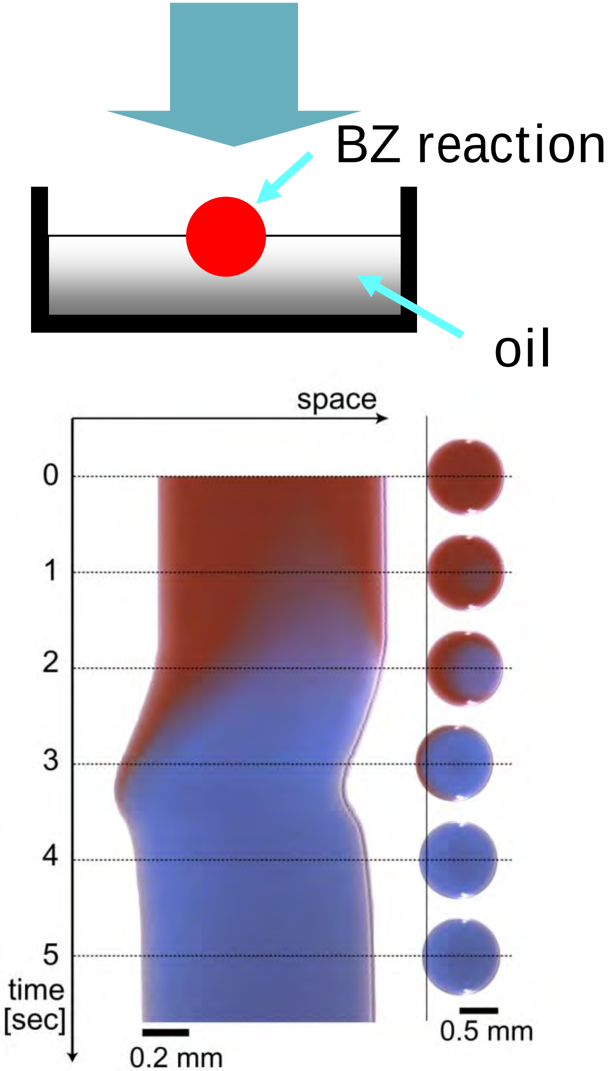




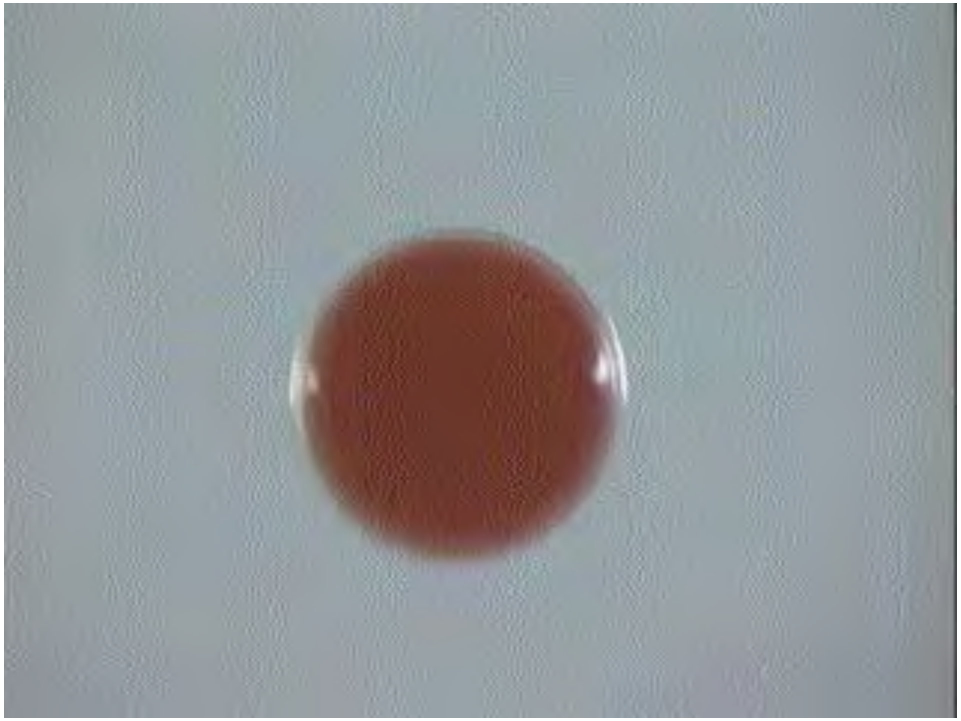
Tensor describing relative position

$$N_{ijk} = \hat{r}_{12,i}\hat{r}_{12,j}\hat{r}_{12,k} - \frac{1}{d+2}(\hat{r}_{12,i}\delta_{jk} + \hat{r}_{12,j}\delta_{ik} + \hat{r}_{12,k}\delta_{ij})$$

Self-propulsion of a BZ drop



BZ reaction medium on oil



Top view

1 mm

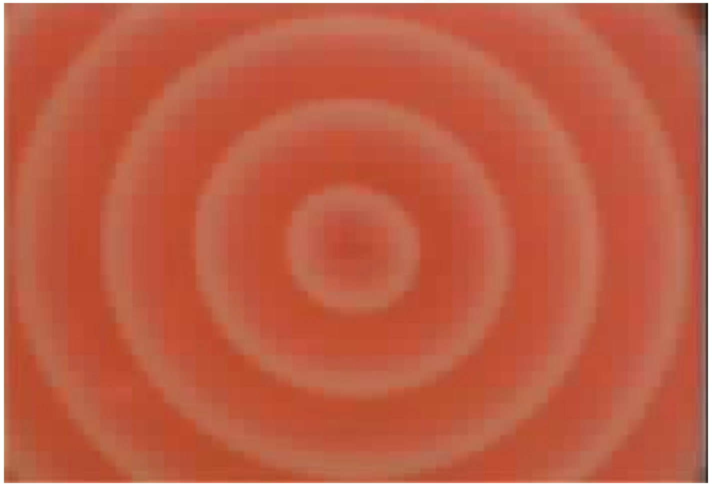
H. Kitahata et al. (2002)

H. Kitahata, NY, K. H. Nagai and S. Yutaka, *PRE* **84** 015101 (2011)

Pattern in Chemical Reactions

Target Pattern

Spiral Pattern



(x4 in time)

3 mm

(x4 in time)

3 mm

Chemical reactions and flow field

- Chemical Reaction (Oregonator Model)

Activator: $$\frac{\partial U}{\partial t} + \mathbf{v} \cdot \nabla U = \frac{1}{\varepsilon}\left[U(1-U) - fV\frac{U-q}{U+q} \right] + D_U \nabla^2 U$$

Inhibitor: $$\frac{\partial V}{\partial t} + \mathbf{v} \cdot \nabla V = U - V + D_V \nabla^2 V$$

J. P. Keener and J. J, Tyson (1986)

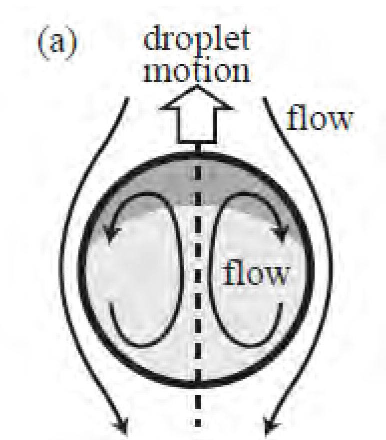
- Hydrodynamics

$$-\nabla p + \eta \nabla^2 \mathbf{v} = 0$$

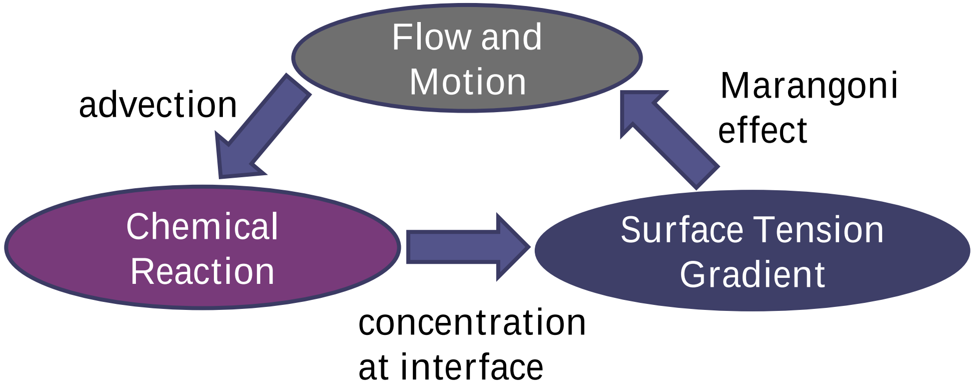
$$\nabla \cdot \mathbf{v} = 0$$

Surface tension

$$\gamma \propto V$$



$$\boldsymbol{n} \cdot \boldsymbol{\sigma}^{(o)} - \boldsymbol{n} \cdot \boldsymbol{\sigma}^{(i)} = \partial_s (\gamma \boldsymbol{t})$$

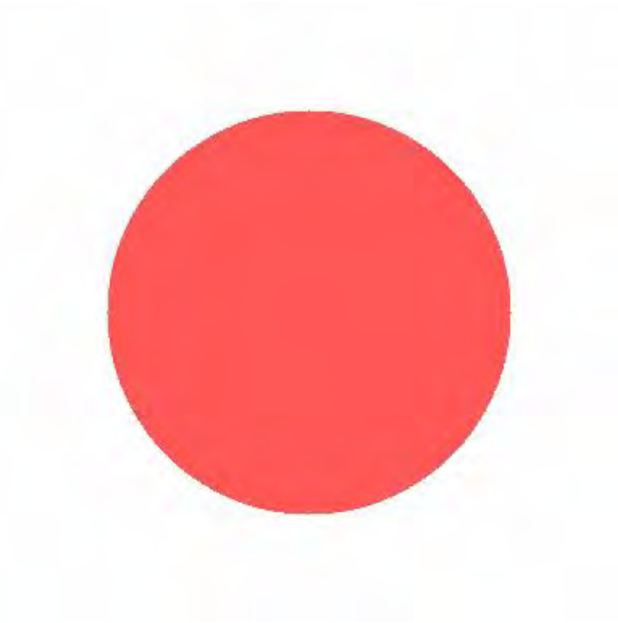


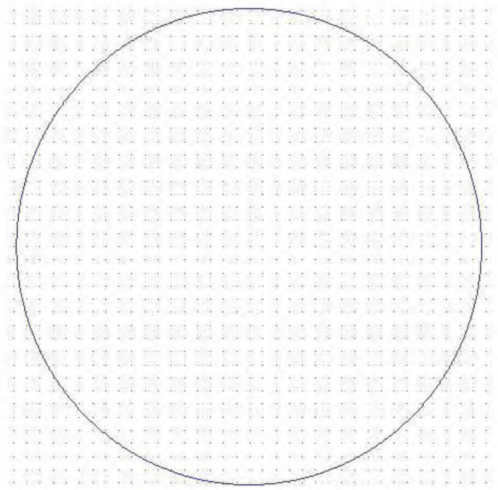
H. Kitahata, NY, K. H. Nagai and S. Yutaka, *PRE* **84** 015101 (2011)

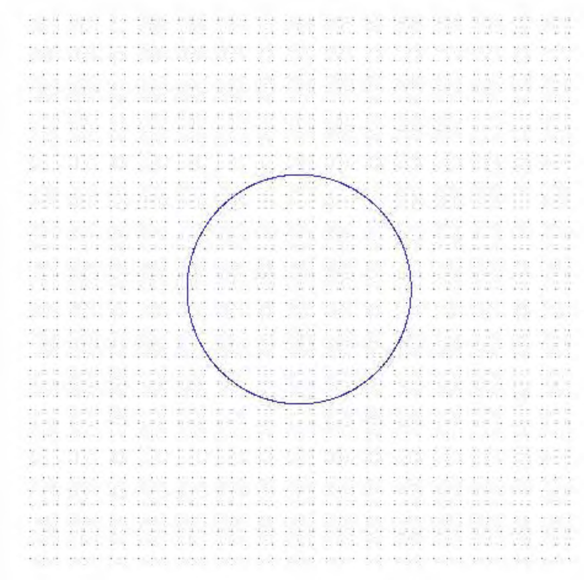
Target pattern and Motion

concentration

velocity field





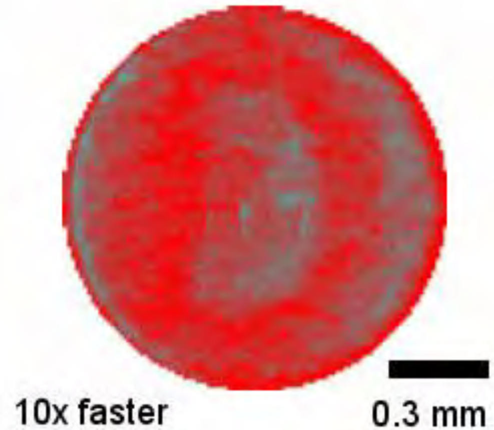


H. Kitahata, NY, K. H. Nagai and S. Yutaka, *PRE* **84** 015101 (2011)

Pattern and motion

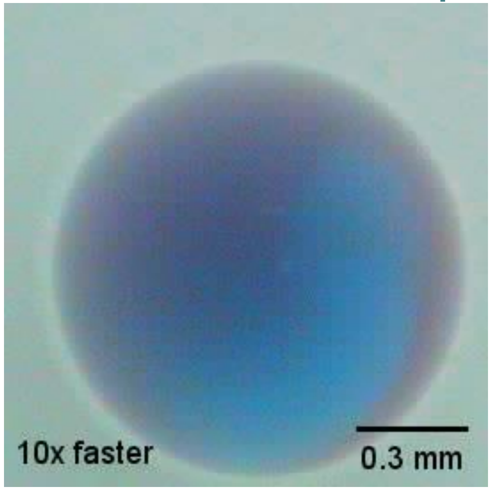
theory

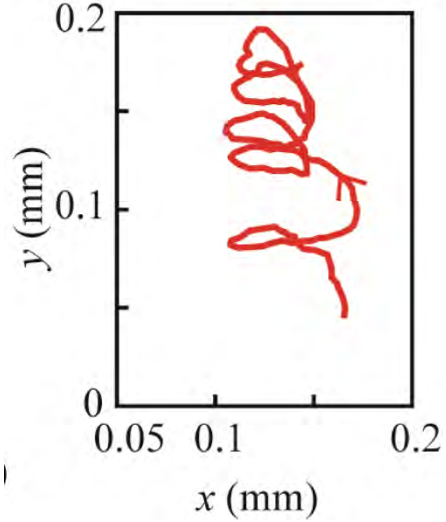
experiment



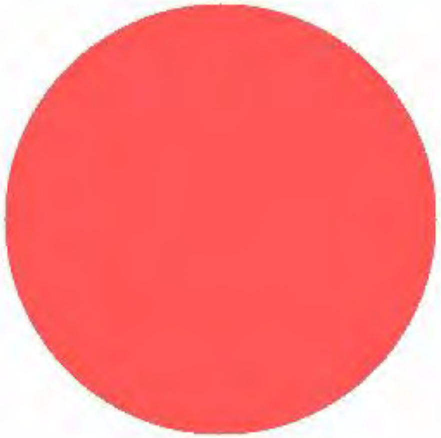
spiral pattern

experiment





theory



scrolling pattern

H. Kitahata, NY, K. H. Nagai and S. Yutaka, *Chem. Lett.* (2012)

Summary and Remarks

- Spontaneous motion and deformation
  - The amplitude equations for spontaneous symmetry breaking of self-propulsion is obtained.
  - Force moment and flow are characterized as a pusher.
- Motion and pattern formation
  - Pattern induced by chemical reaction makes a droplet motion.
- Future works
  - Collective phenomena, and hydrodynamic theory
  - Fluctuation (Langevin, stochastic PDE)
  - Complex fluid inside a droplet

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