Self-diffusiophoresis in the strongly advecting regime

Gareth Alexander

Timon Idema

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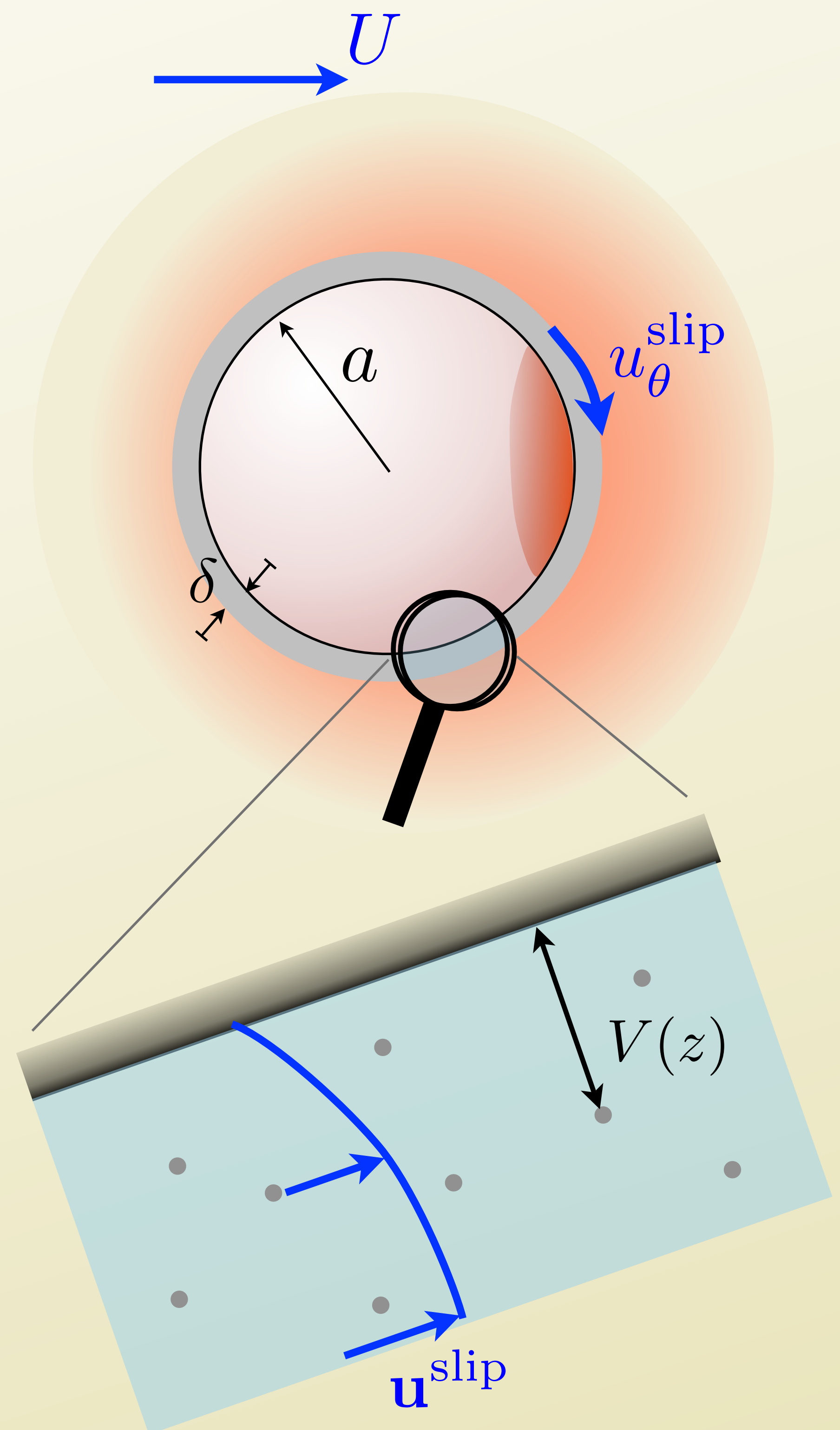

Deparment of Physics & Astronomy
University of Pennsylvania

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Mathematics and Physics of Soft and Biological Matter
ICTP Trieste, May 9th to 13th 2011

ACTIN BASED PROPULSION

Listeria monocytogenes

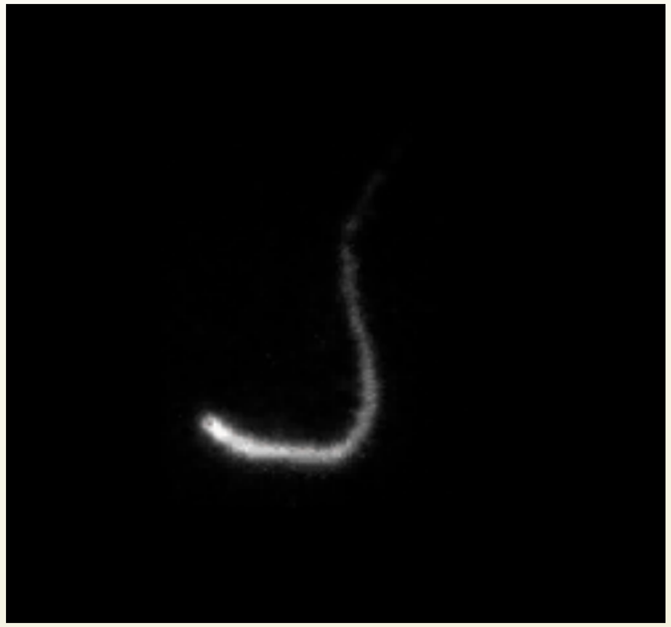
Courtesy of Julie Theriot
http://cmgm.stanford.edu/theriot/movies.htm

IN–VITRO REALISATIONS

Courtesy of Julie Theriot
http://cmgm.stanford.edu/theriot/movies.htm

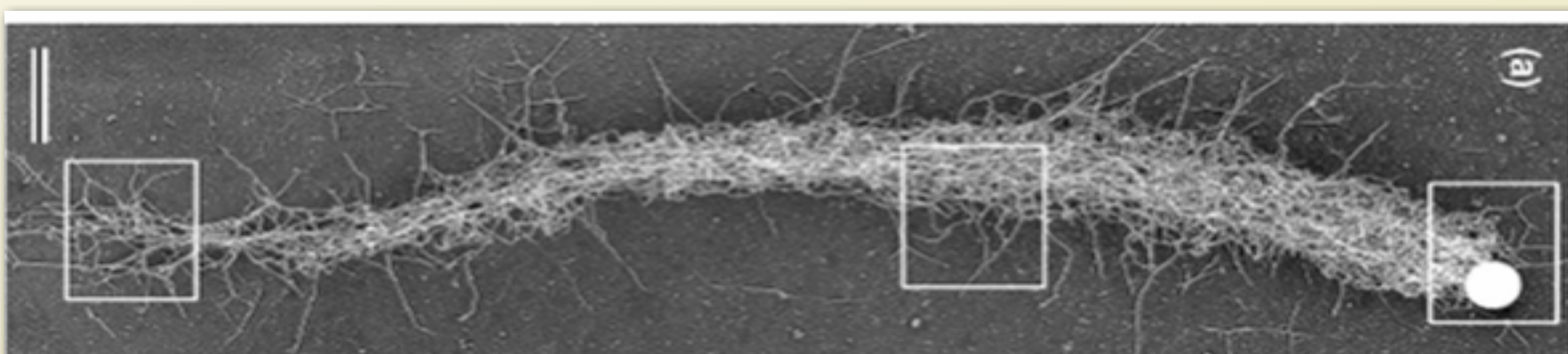
$\times 60$

sphere



"All" you need is

- Actin and buffer w/ATP
- Arp2/3 makes new growing ends
- Capping protein kills them off
- ADF/cofilin severs filaments
- Profilin converts ADP-G-actin to ATP-G-actin
- Bead coated with ActA, VCA, activates Arp2/3



CAMERON ET AL *Curr. Biol.* **11**, 130 (2001)

How does self-assembly into a branched structure lead to motility?

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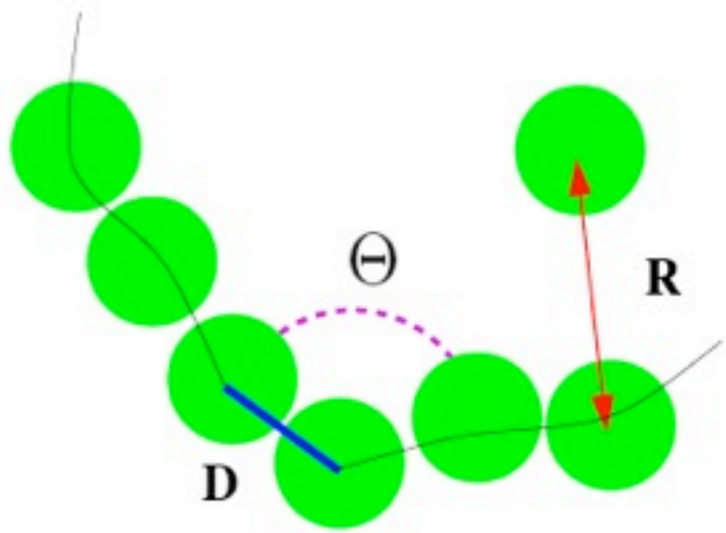
Brownian Dynamics Simulations



$$\eta \frac{\mathrm{d}X}{\mathrm{d}t} = -\nabla\Phi_{\mathrm{H}} - \nabla\Phi_{\mathrm{S}} - \nabla\Phi_{\mathrm{B}} + F_R$$

F_R = Random Force

$$\langle F_R \rangle = 0$$

$$\langle F_R(t) F_R(t') \rangle = 6 k_{\mathrm{B}} T \eta \delta(t - t')$$

Φ_{H} = Hardcore interaction

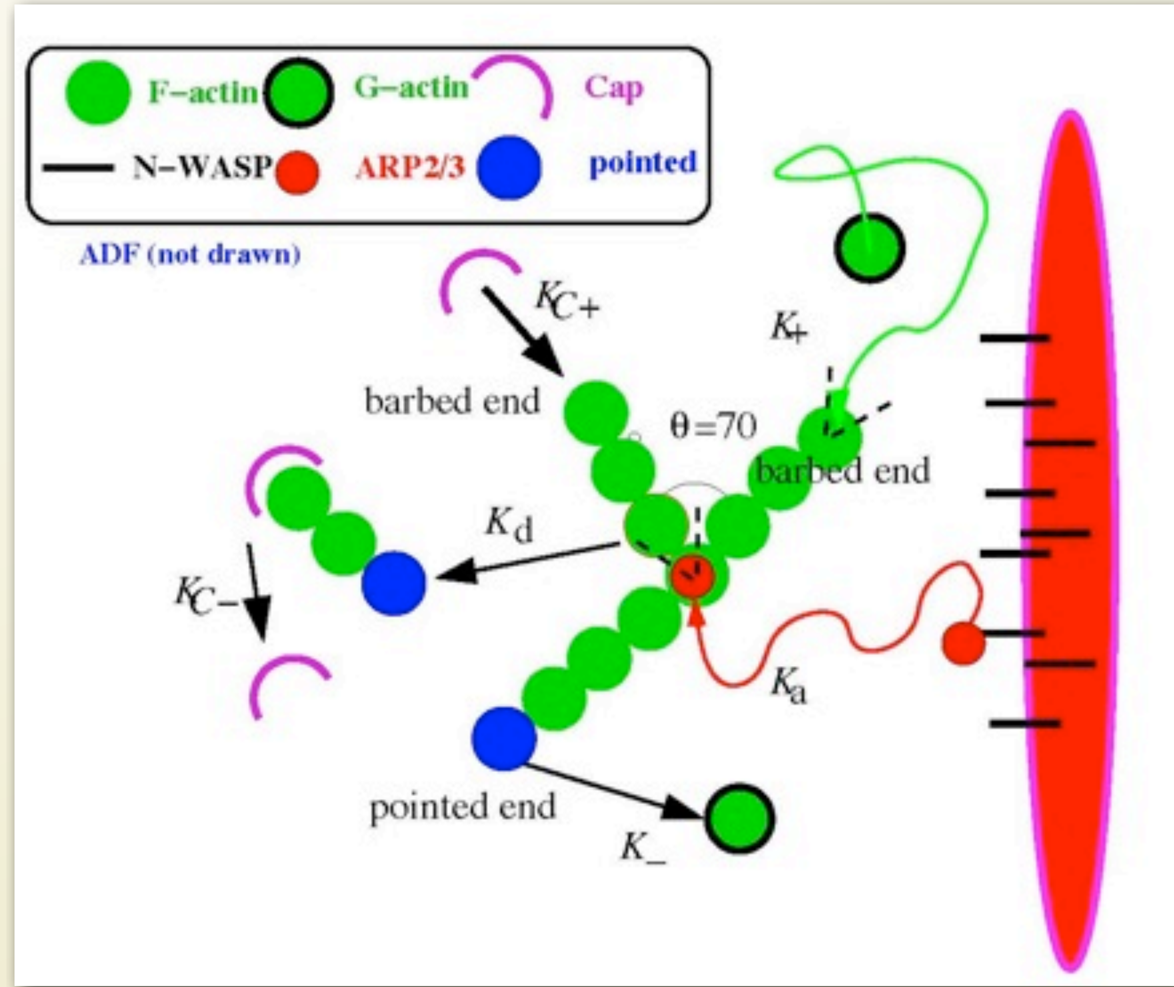
$$\Phi_{\mathrm{H}} = \tfrac{1}{2} K_h (R - R_0)^2, \quad R < R_0$$

Φ_{S} = Bond Stretch interaction

$$\Phi_{\mathrm{S}} = \tfrac{1}{2} K_s (D - D_0)^2$$

Φ_{B} = Bending interaction

$$\Phi_{\mathrm{B}} = \tfrac{1}{2} K_b (\cos(\Theta) - \cos(\Theta_0))^2$$



- Polymerisation at + end (K_+)
- Depolymerisation at - end (K_-)
- Branching (K_a)
- Debranching (K_d)
- Capping

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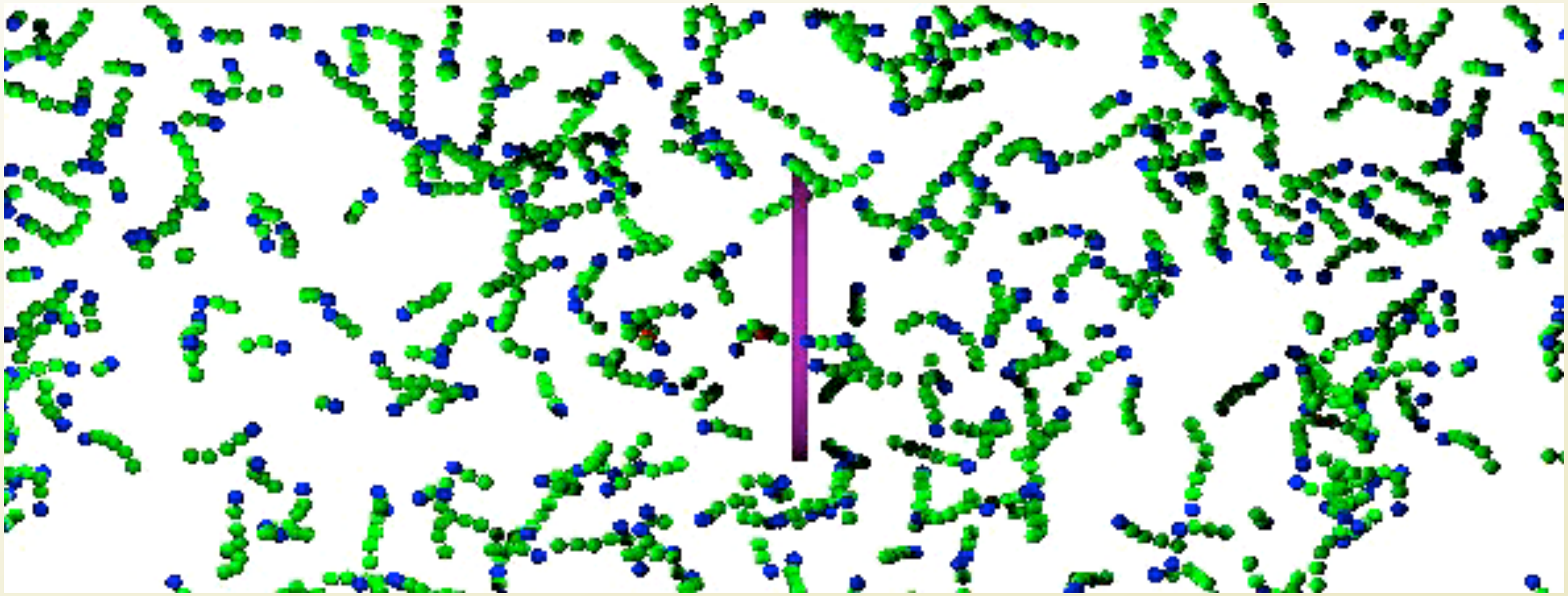
Lee & Liu *Biophys. J.* **95**, 4529–4539 (2008)
Lee & Liu *Biophys. J.* **97**, 1295–1304 (2009)

BROWNIAN DYNAMICS SIMULATIONS



- 2D projection of 3D simulation
- Motion allowed in $\pm z$ direction only

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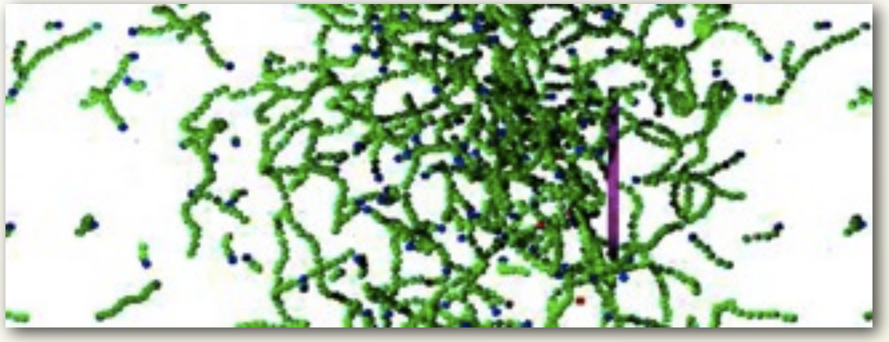
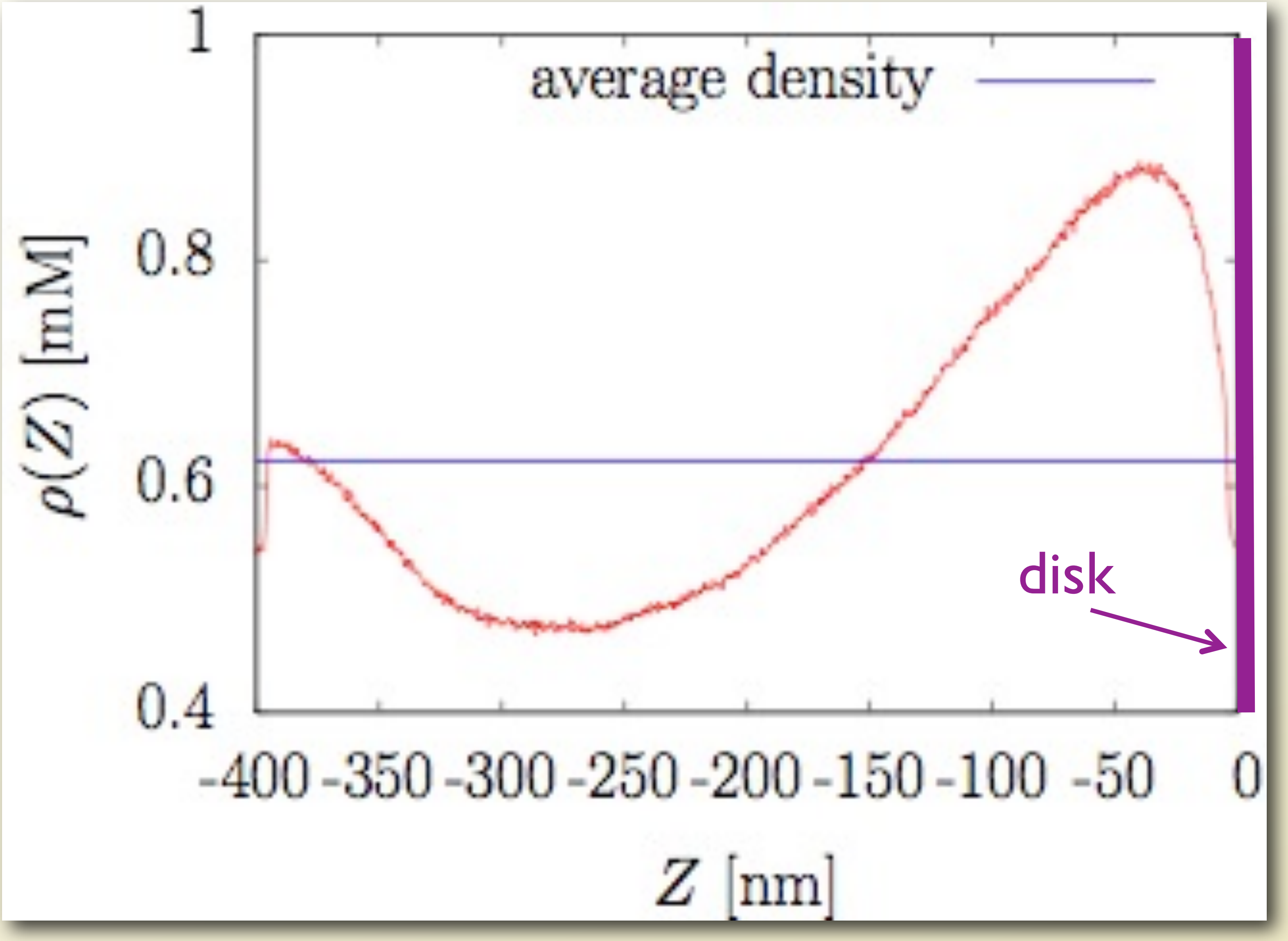
LEE & LIU *Biophys. J.* **95**, 4529–4539 (2008)
LEE & LIU *Biophys. J.* **97**, 1295–1304 (2009)

ACTIN CONCENTRATION GRADIENT



Courtesy of J. Theriot



- Disk activates Arp2/3, which recruits F-actin
- Concentration of F-actin is high behind the disk compared to average
- If the disk repels actin then it will move forwards to avoid F-actin
- In real systems the concentration gradient is even bigger; mechanism should still apply

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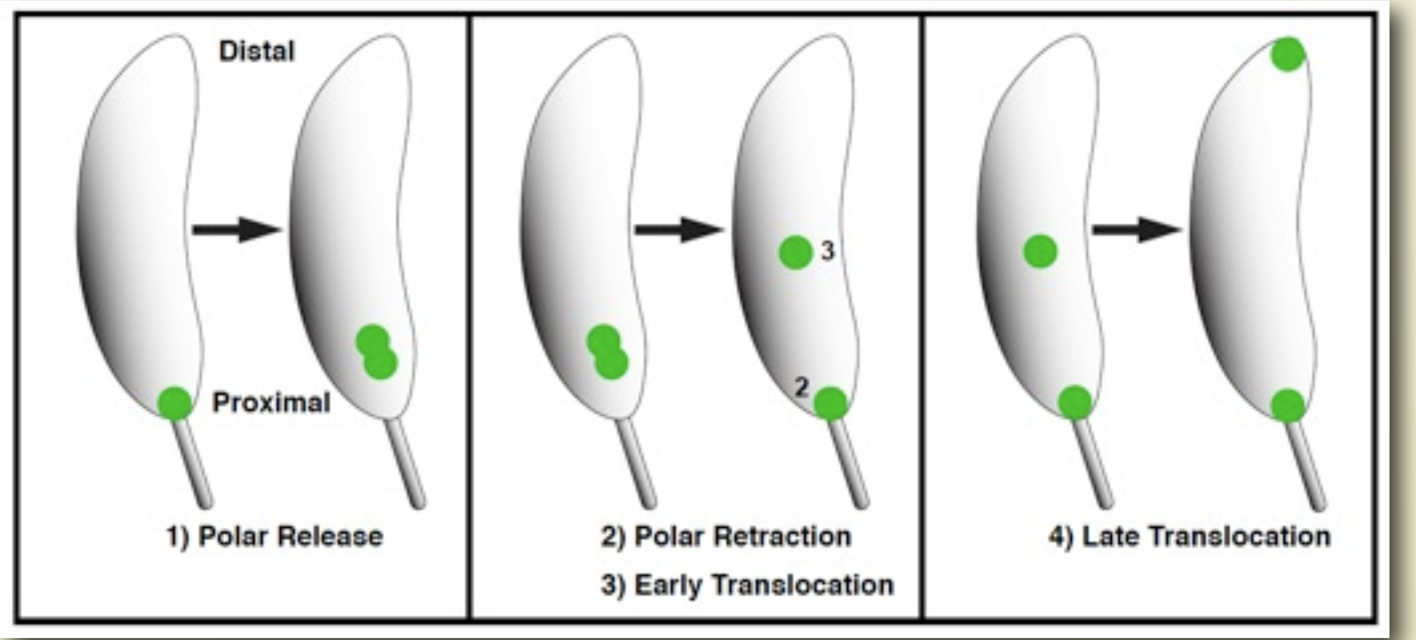
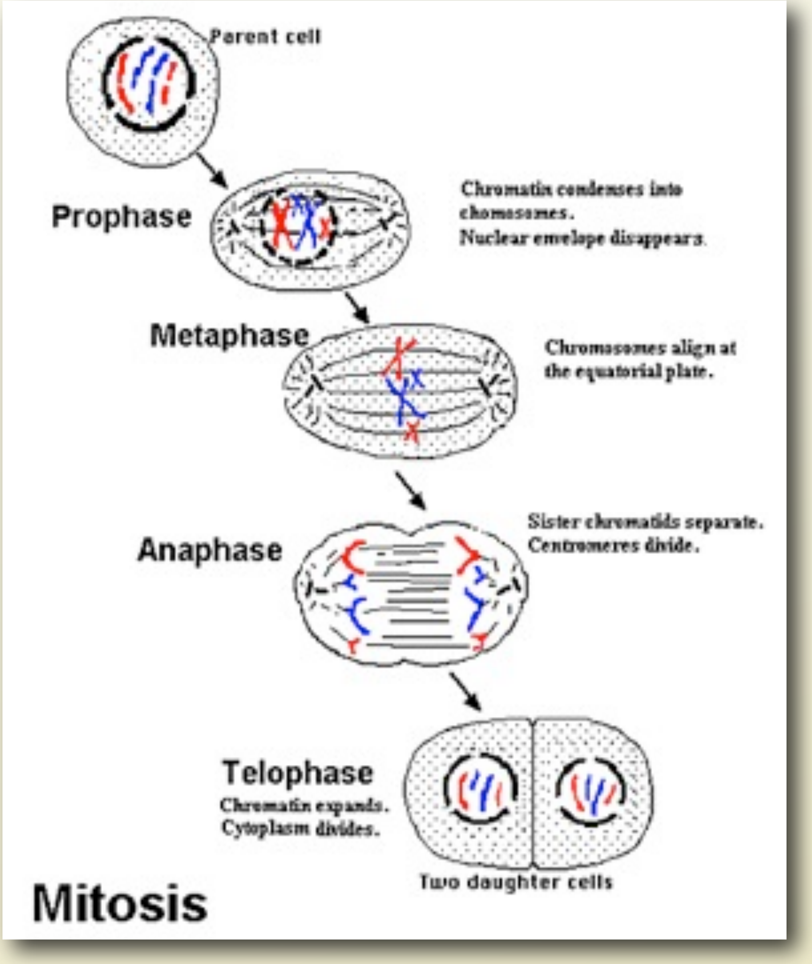
Lee & Liu *Biophys. J.* **95**, 4529–4539 (2008)
Lee & Liu *Biophys. J.* **97**, 1295–1304 (2009)

ASYMMETRIC CELL DIVISION

- Most cells divide symmetrically

Caulobacter crescentus and *Vibrio cholerae* divide asymmetrically





- What is the mechanism for chromosomal motility?

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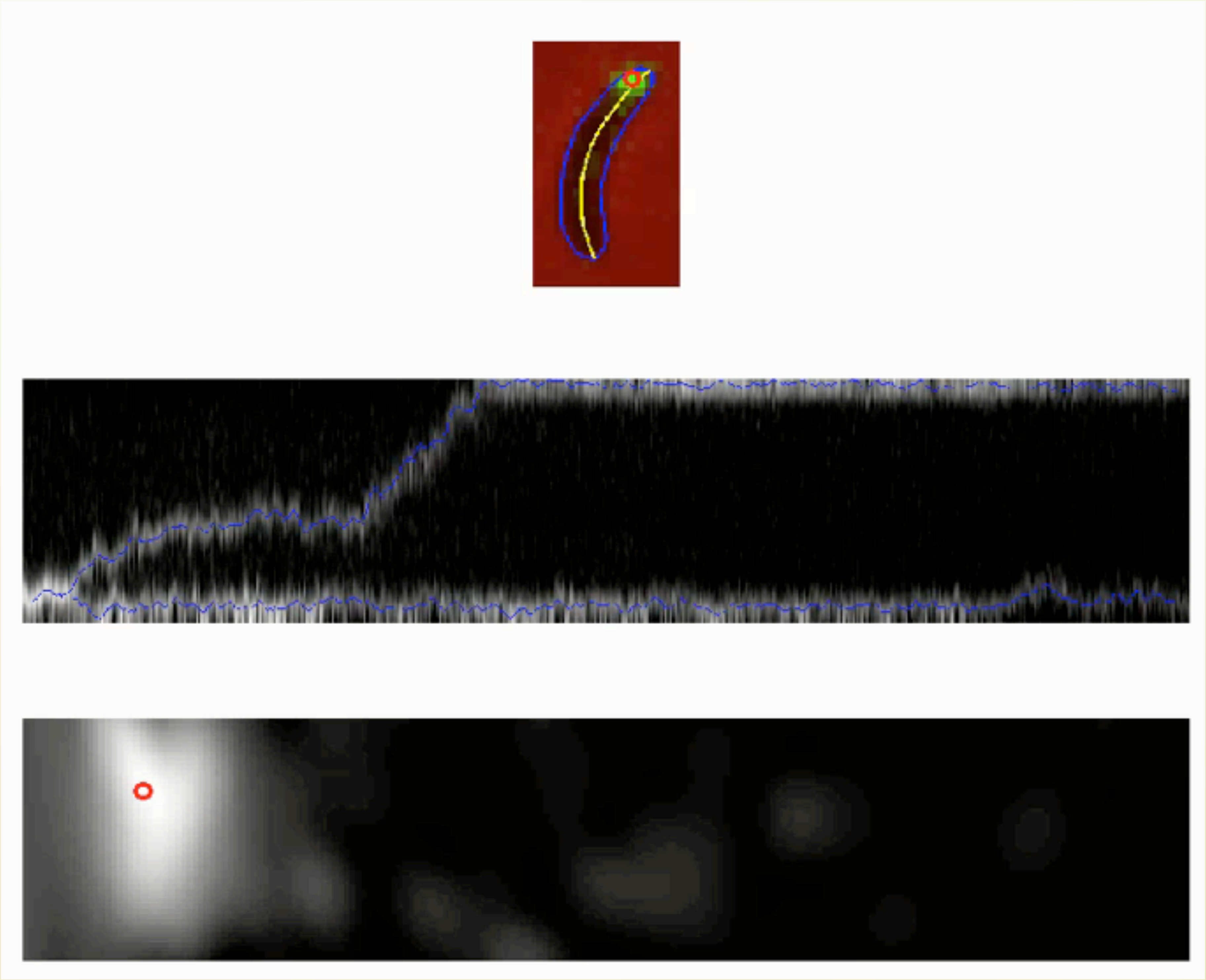
DISASSEMBLY DRIVEN MOTILITY

- How does the chromosome move across the cell during chromosomal segregation in certain asymmetric bacteria?

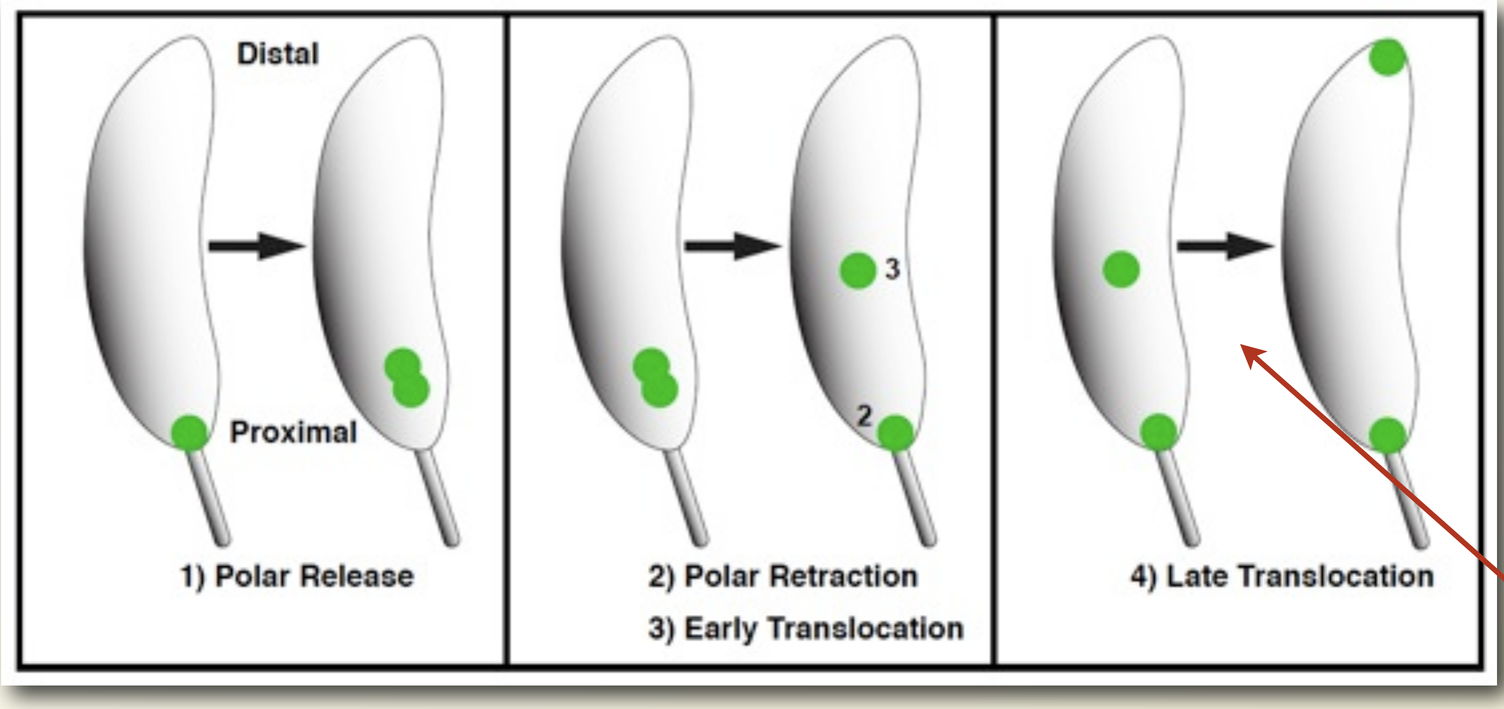


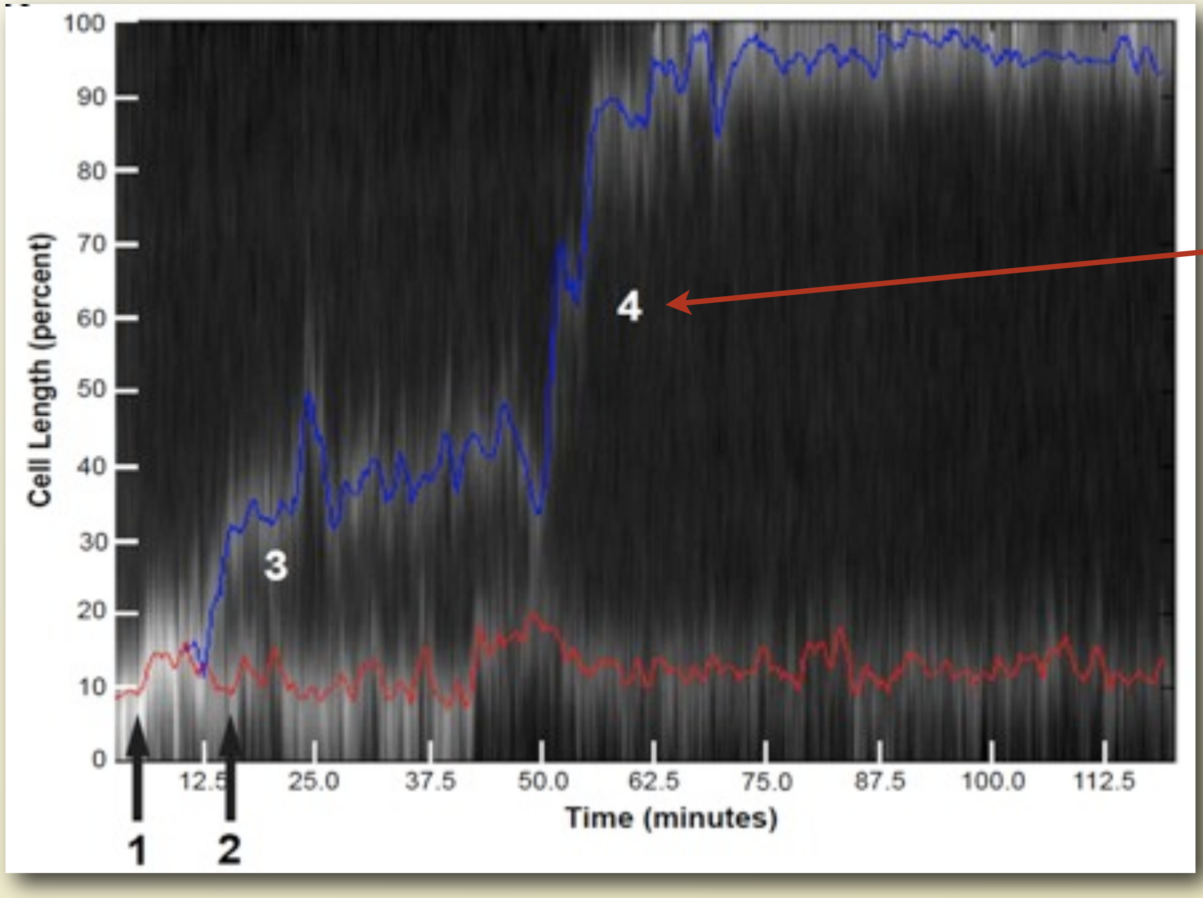
Caulobacter crescentus

Courtesy of C. W. Shebelut, J. M. Guberman and Z. Gitai

Chromosomal segregation in C. crescentus and V. cholerae

Caulobacter crescentus





We are interested in the 4th stage

How does the chromosome (ori) scoot across the cell?

Courtesy of C. W. Shebelut, J. M. Guberman and Z. Gitai

Vibrio cholerae

Courtesy of Popular Logistics

Fogel & Waldor *Genes & Dev.* **20**, 3269–3282 (2006)

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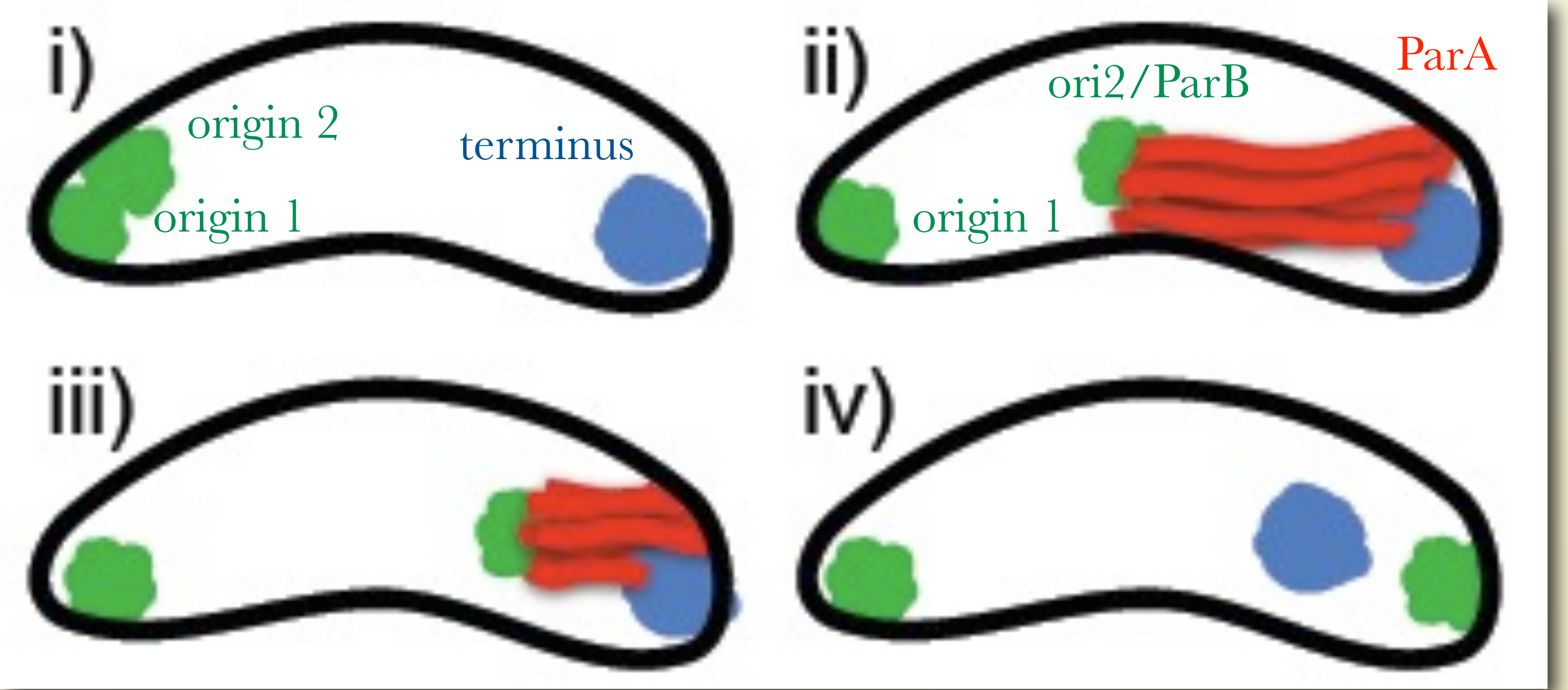
A CLOSER LOOK AT THE PROCESS

FOGEL & WALDOR *Genes & Dev.* **20**, 3269–3282 (2006)

Replication

ParB on origin attaches to ParA



ParA disassembles and origin moves

Origin and terminus switch places

- Origin is decorated with ParB which binds to and hydrolyses ParA
- ParA filament structure depolymerises and drags ParB along

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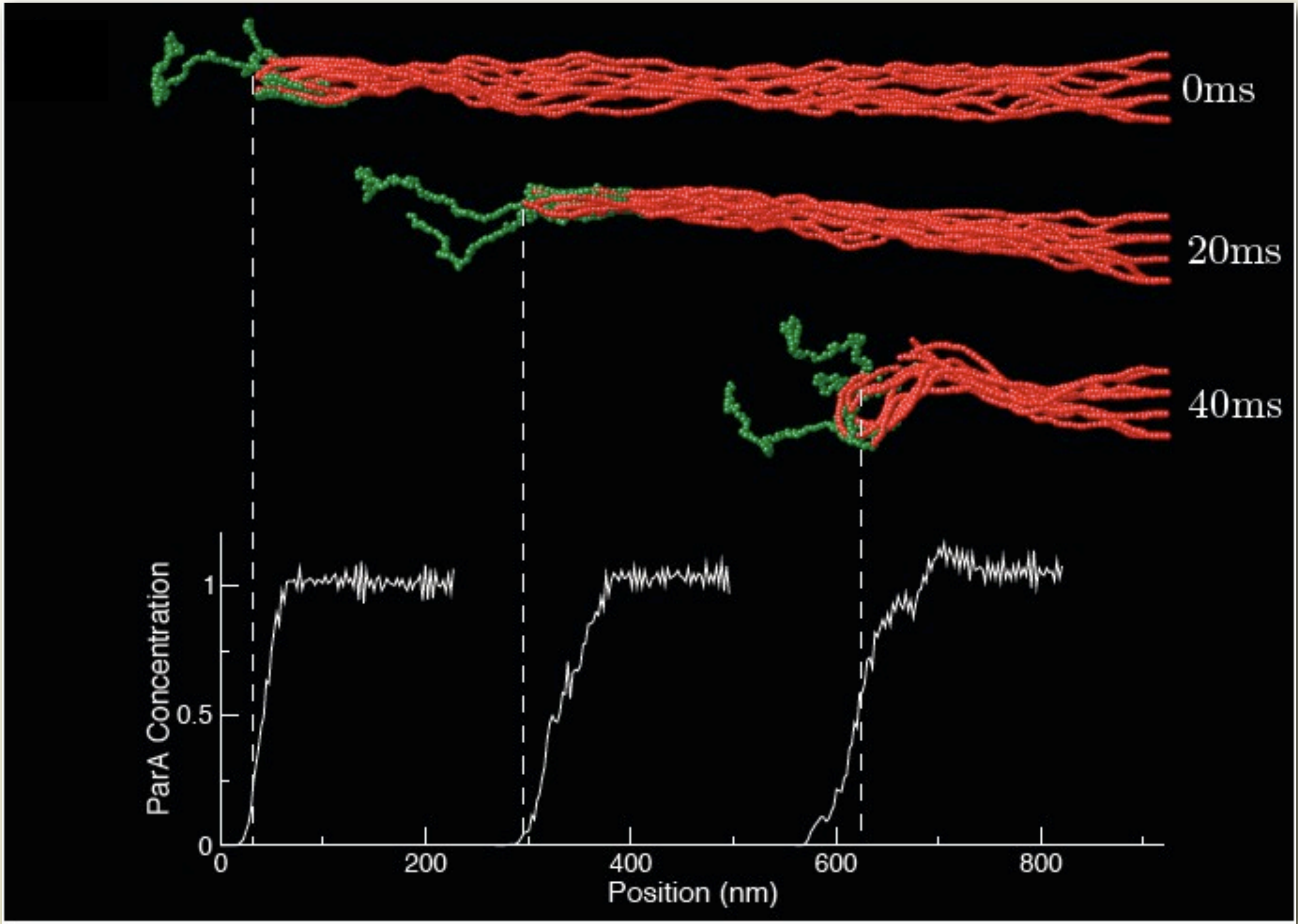
CONCENTRATION GRADIENT DRIVES MOTION



- System uses depolymerisation to create a steady-state concentration gradient to move up

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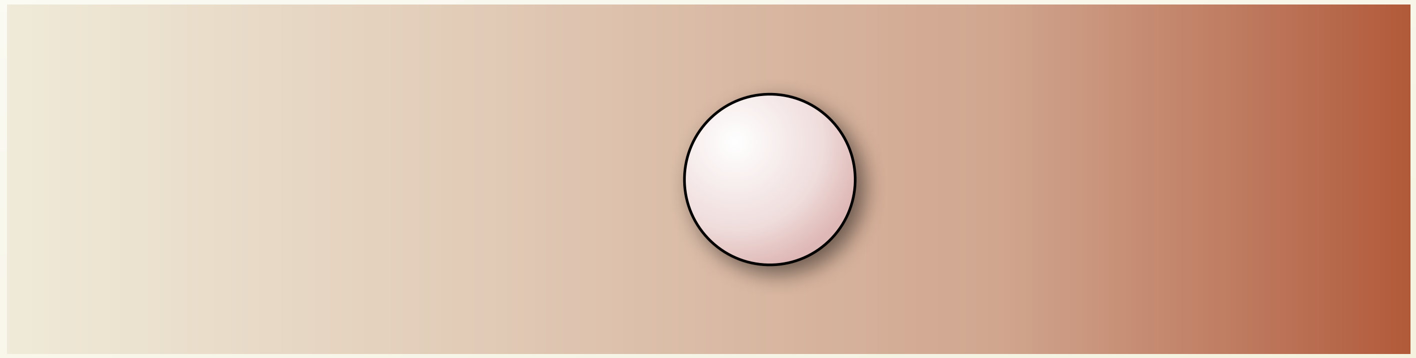
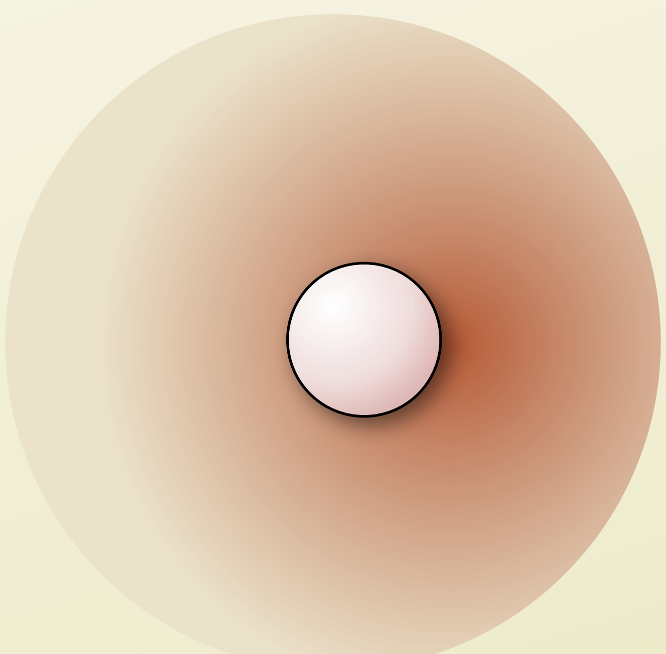
BANIGAN ET AL *submitted* (2011)

Particle motion in a concentration gradient

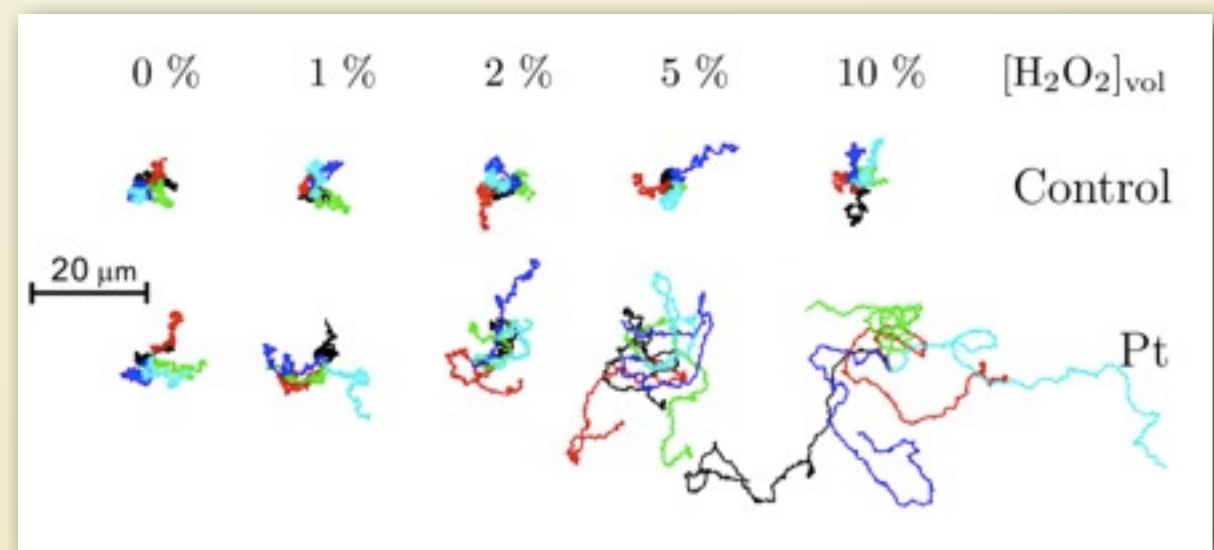


Particle interacts with the concentration field and moves down the gradient if it is repelled / up the gradient if it is attracted

In self-diffusiophoresis the particle itself generates the concentration gradient



Example: catalytic decomposition of H_2O_2



Howse et al *Phys. Rev. Lett.* **99**, 048102 (2007)

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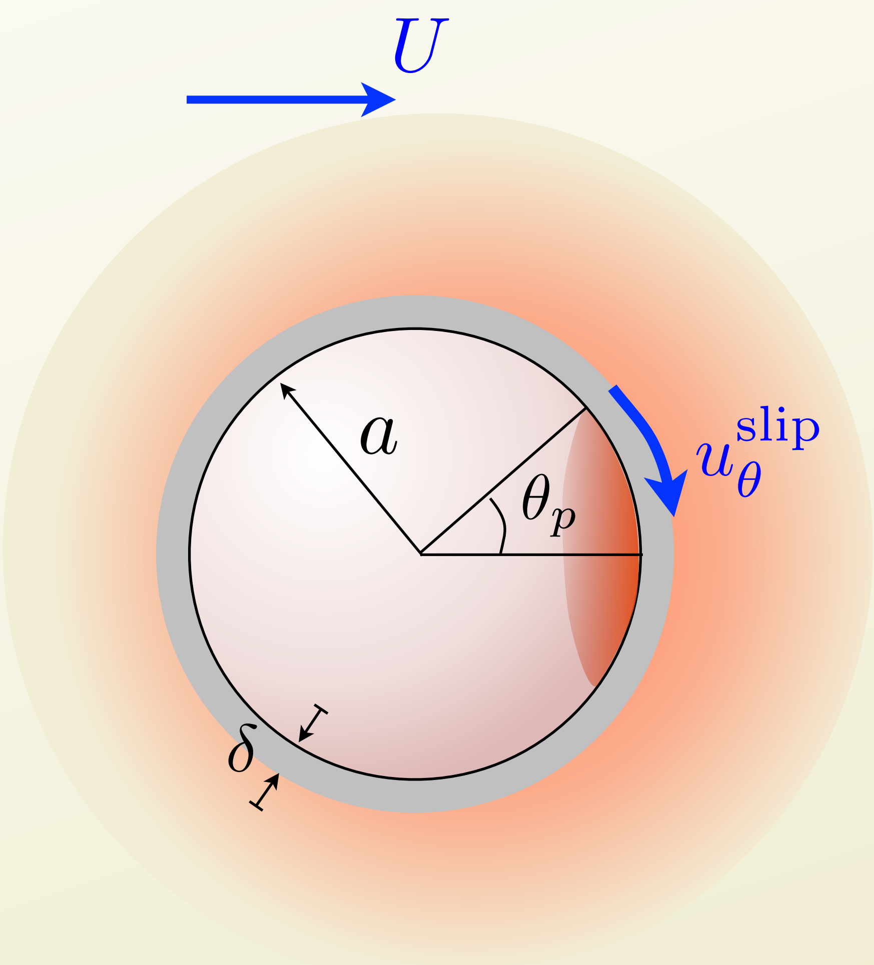
Anderson *Ann. Rev. Fluid Mech.* **21**, 61–99 (1989)
Golestanian, Liverpool & Ajdari *Phys. Rev. Lett.* **94**, 220801 (2005)

A PARED DOWN MODEL



Rest frame of the particle

- Spherical particle
- Axisymmetry, steady state
- Single solute species
- Purely radial potential with compact support
- Zero Reynolds number

Golestanian, Liverpool & Ajdari *Phys. Rev. Lett.* **94**, 220801 (2005)
Golestanian, Liverpool & Ajdari *New J. Phys.* **9**, 126 (2007)

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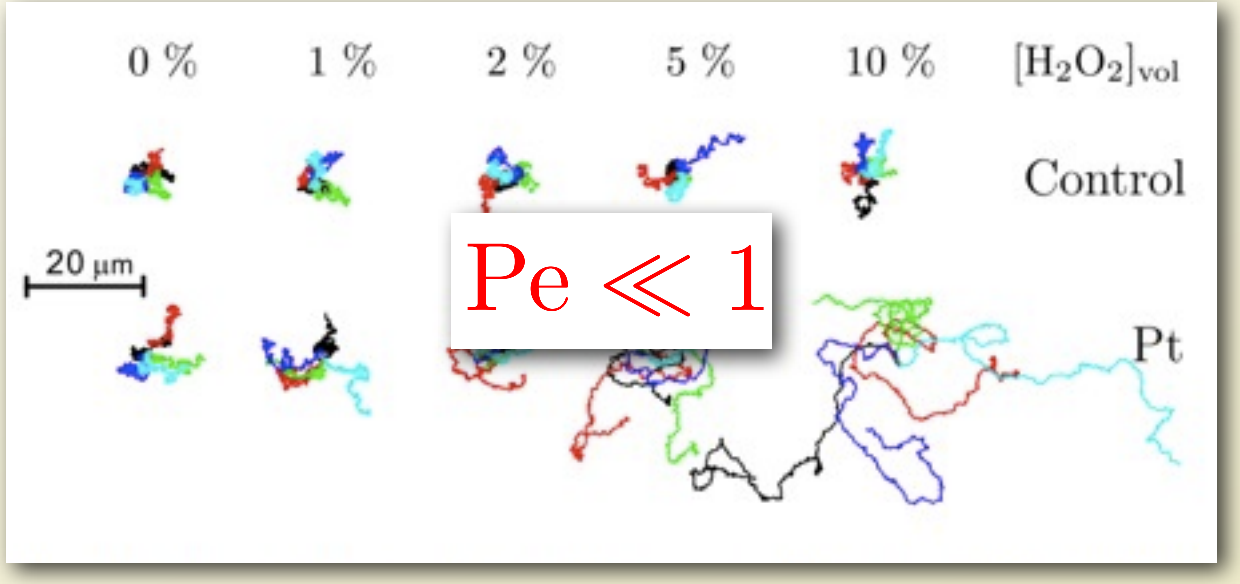
PARTICLE MOTION IN A CONCENTRATION GRADIENT

Motion involves a balance between diffusion and advection

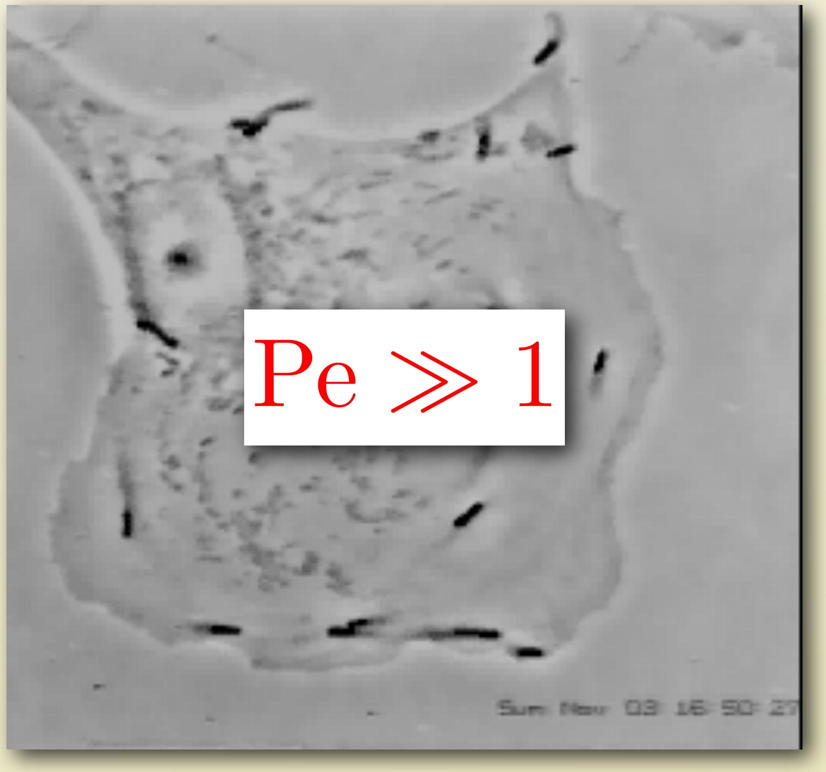
$$\mathrm{Pe} = \frac{\text{advection}}{\text{diffusion}} = \frac{U a}{D}$$

Diffusion dominated



HOWSE ET AL *Phys. Rev. Lett.* **99**, 048102 (2007)

Advection dominated



Courtesy of Julie Theriot
http://cmgm.stanford.edu/theriot/movies.htm

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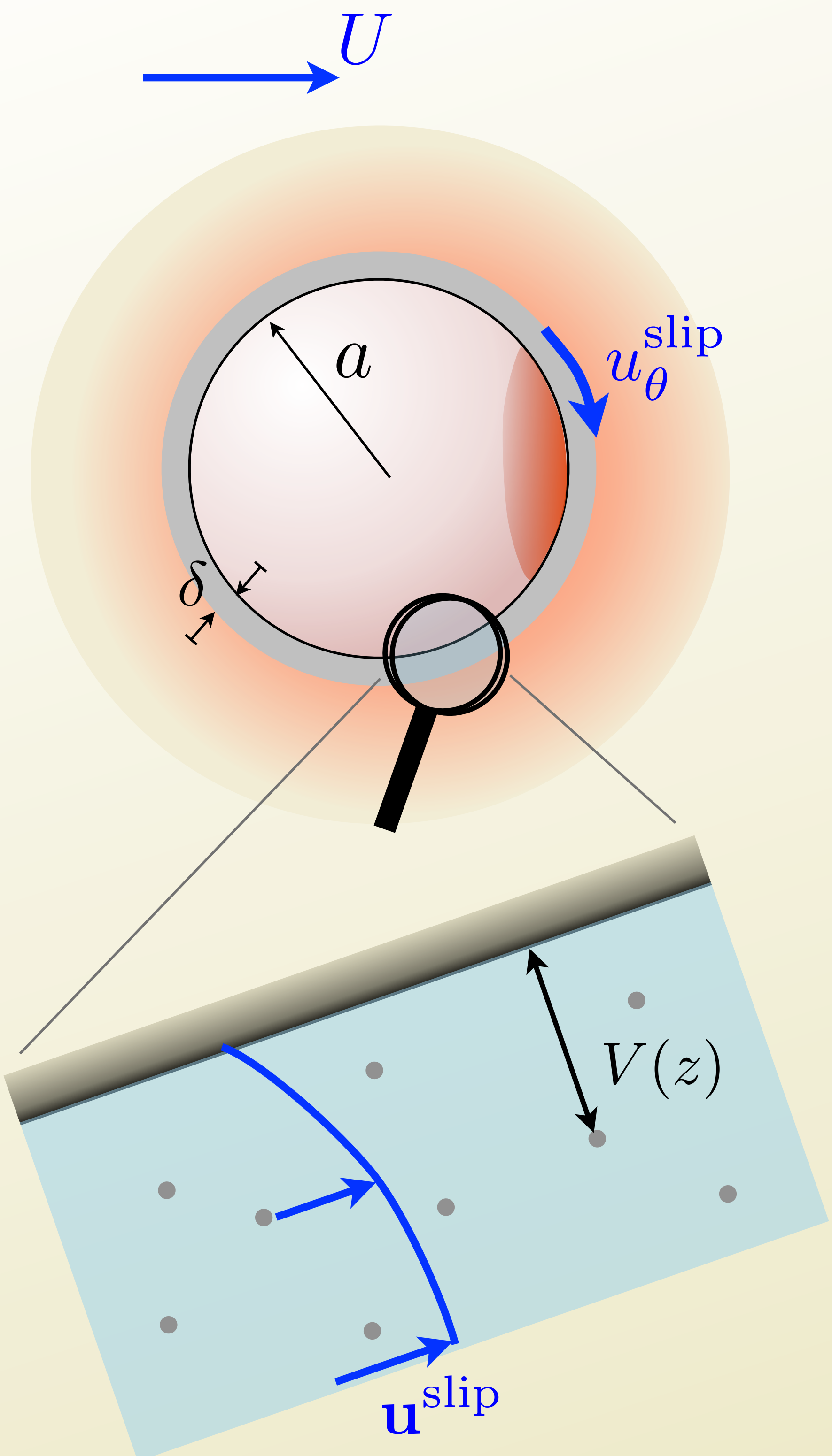
Boundary layer analysis



The interaction occurs close to the surface in a boundary layer of thickness δ

Solve Stokes in the upper half space

$$\mathbf{0} = -\nabla p + \mu \nabla^2 \mathbf{u} - c\,\nabla V \qquad 0 = \nabla \cdot \mathbf{u}$$

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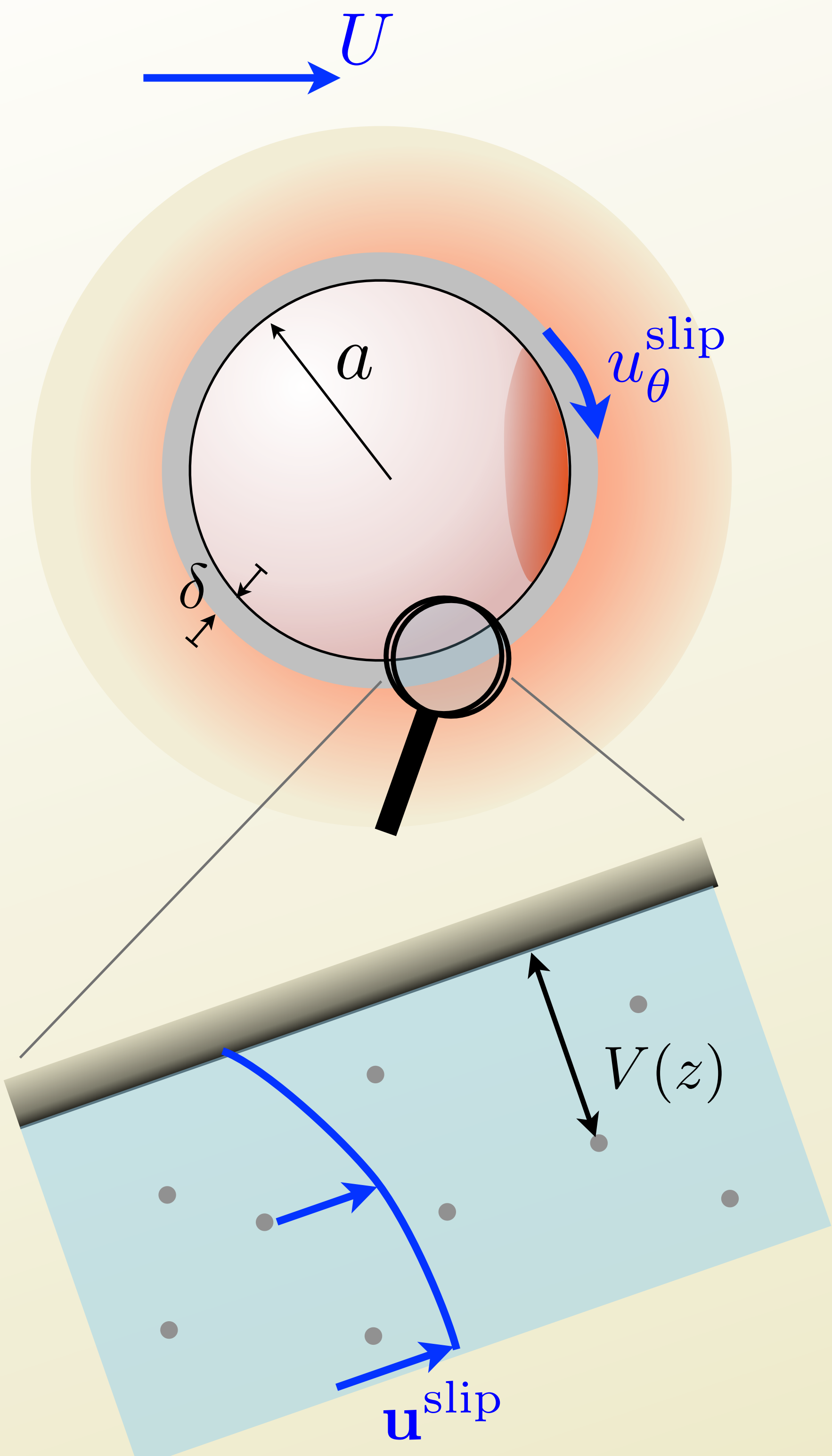
Anderson *Ann. Rev. Fluid Mech.* **21**, 61–99 (1989)
Jülicher, Prost *Eur. Phys. J. E* **29**, 27–36 (2009)

Boundary layer analysis



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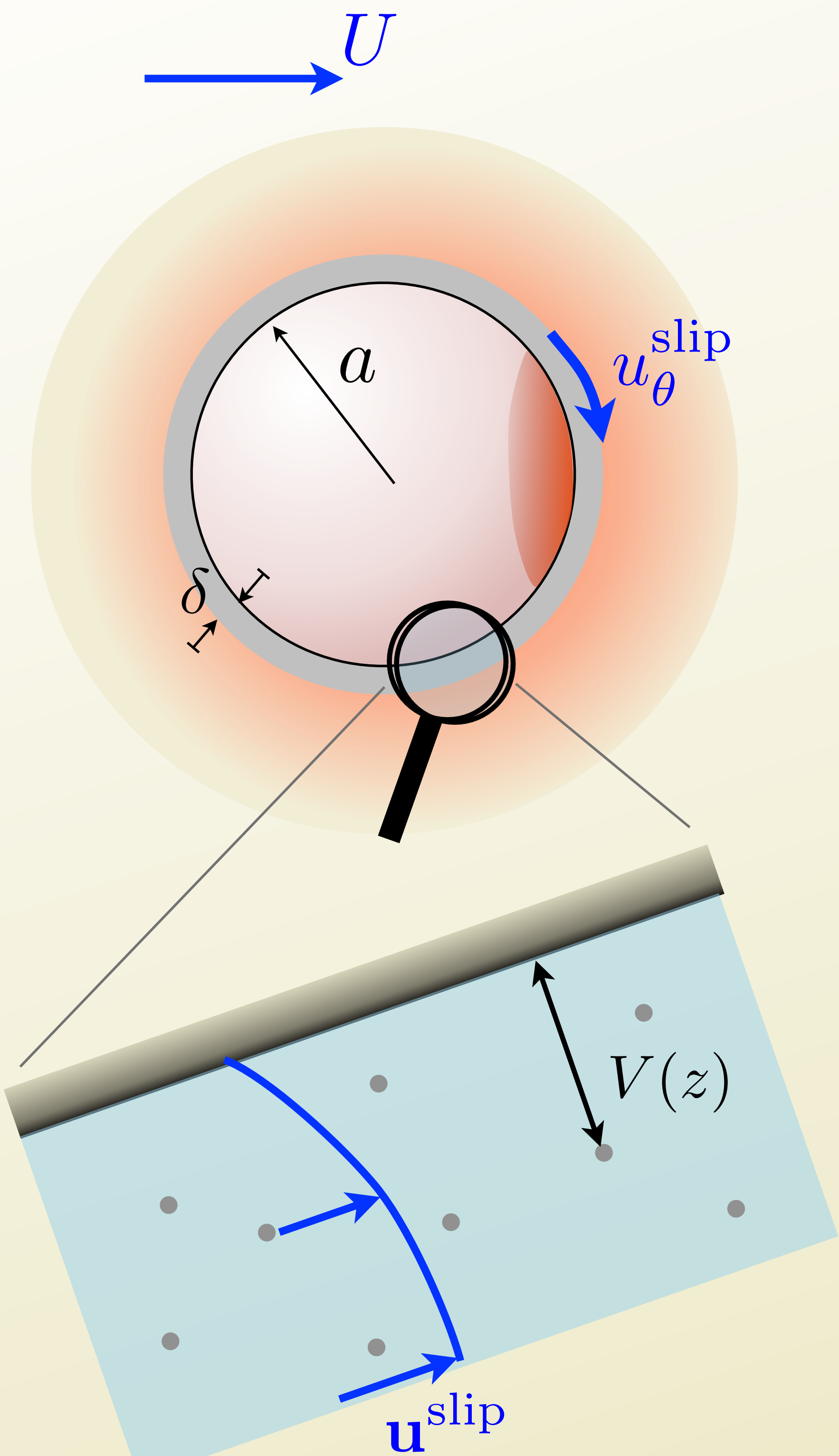
Tangential slip velocity

$$\mathbf{u}^{\text{slip}} = -\frac{\delta^2}{\mu} \int_0^1 \mathrm{d}z\, \tfrac{1}{2} z^2\, \nabla_{\|} c(z, \mathbf{x})\, \partial_z V(z)$$

proportional to tangential concentration gradients

Anderson *Ann. Rev. Fluid Mech.* **21**, 61–99 (1989)
Jülicher, Prost *Eur. Phys. J. E* **29**, 27–36 (2009)

Boundary layer analysis



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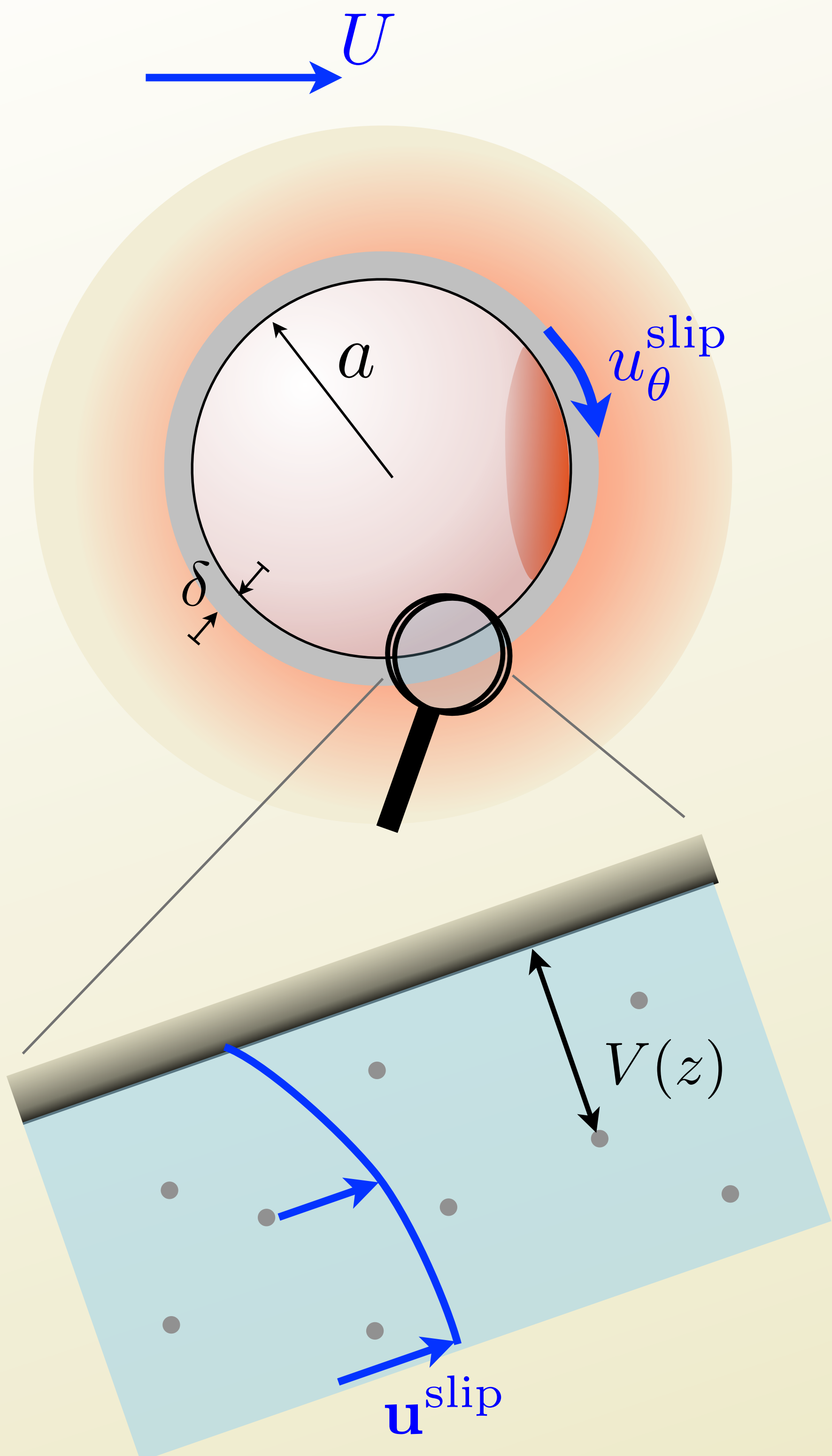
Boltzmann distribution $\quad c(z, \mathbf{x}) = c(1, \mathbf{x}) \, \mathrm{e}^{-V(z)/k_B T}$

Anderson *Ann. Rev. Fluid Mech.* **21**, 61–99 (1989)
Jülicher, Prost *Eur. Phys. J. E* **29**, 27–36 (2009)

BOUNDARY LAYER ANALYSIS



The interaction occurs close to the surface in a boundary layer of thickness δ

Solve Stokes in the upper half space

$$\mathbf{0} = -\nabla p + \mu \nabla^2 \mathbf{u} - c\,\nabla V \qquad 0 = \nabla \cdot \mathbf{u}$$

Tangential slip velocity

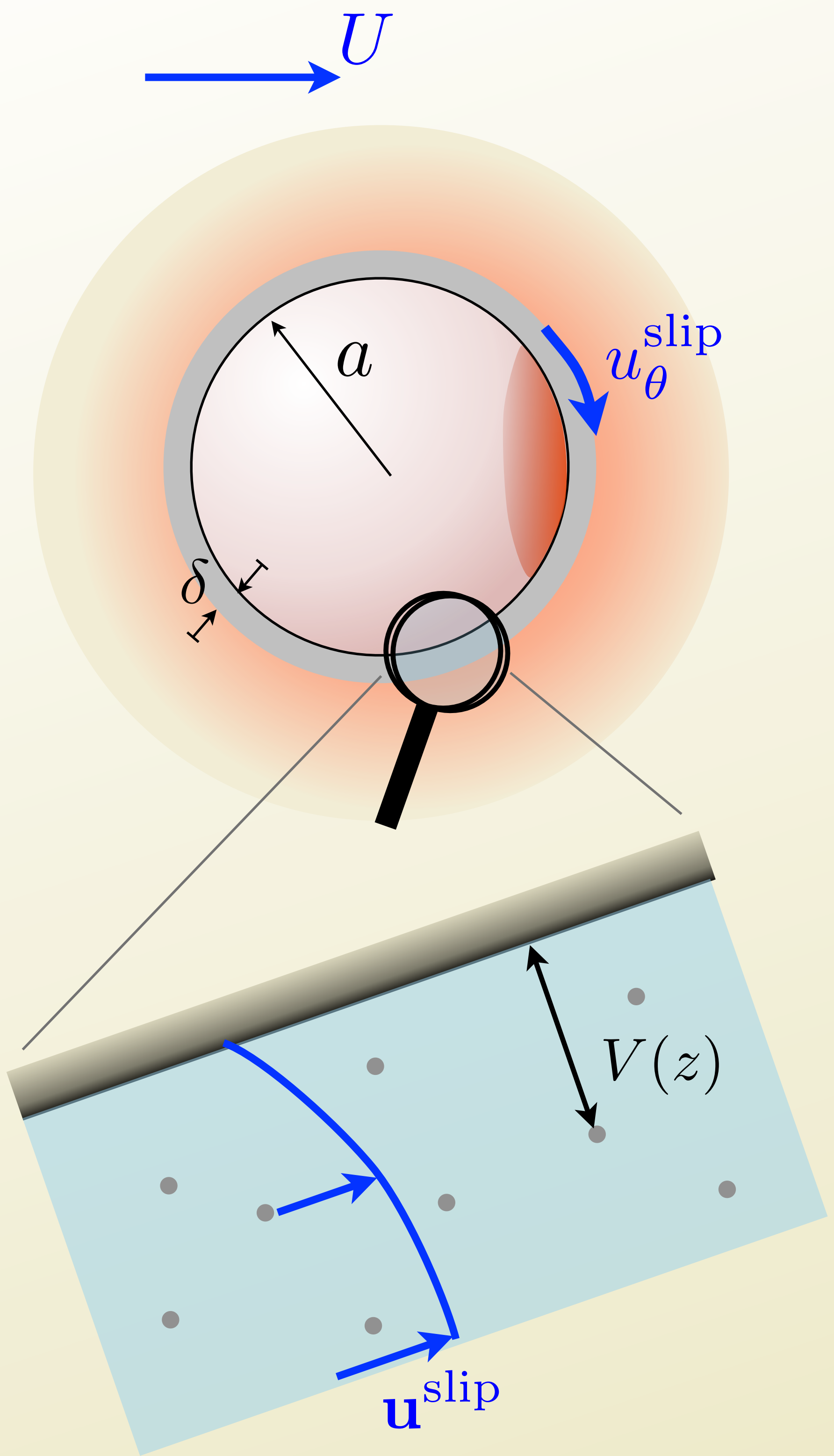
$$\boxed{\mathbf{u}^{\text{slip}} = m^D \nabla_\parallel c\big|_{z=1}}$$

diffusiophoretic mobility (Derjaguin)

$$m^D = \frac{k_B T}{\mu}\delta^2 \int_0^1 \mathrm{d}z\, z\left[1 - \mathrm{e}^{-V/k_B T}\right]$$

ANDERSON *Ann. Rev. Fluid Mech.* **21**, 61–99 (1989)
JÜLICHER, PROST *Eur. Phys. J. E* **29**, 27–36 (2009)

SQUIRMERS



Slip velocity provides an inner boundary condition for the exterior flow

General solution provided by Lighthill's squirmer model

$$u_r = U\left[1 - \left(\tfrac{a}{r}\right)^3\right]\cos(\theta) + \sum_{l=2}^{\infty} B_l\left[\left(\tfrac{a}{r}\right)^l - \left(\tfrac{a}{r}\right)^{l+2}\right]P_l\big(\cos(\theta)\big)$$

$$u_\theta = -U\sin(\theta) + \sum_{l=2}^{\infty} B_l\left[\tfrac{l-2}{2}\left(\tfrac{a}{r}\right)^l - \tfrac{l}{2}\left(\tfrac{a}{r}\right)^{l+2}\right]V_l\big(\cos(\theta)\big)$$

Matching the boundary condition gives the speed

$$\boxed{U = \frac{2m^D}{3a}c_1}$$

first Legendre coefficient

$$c\big|_{r=a+\delta} = \sum_{l=0}^{\infty} c_l P_l\big(\cos(\theta)\big)$$

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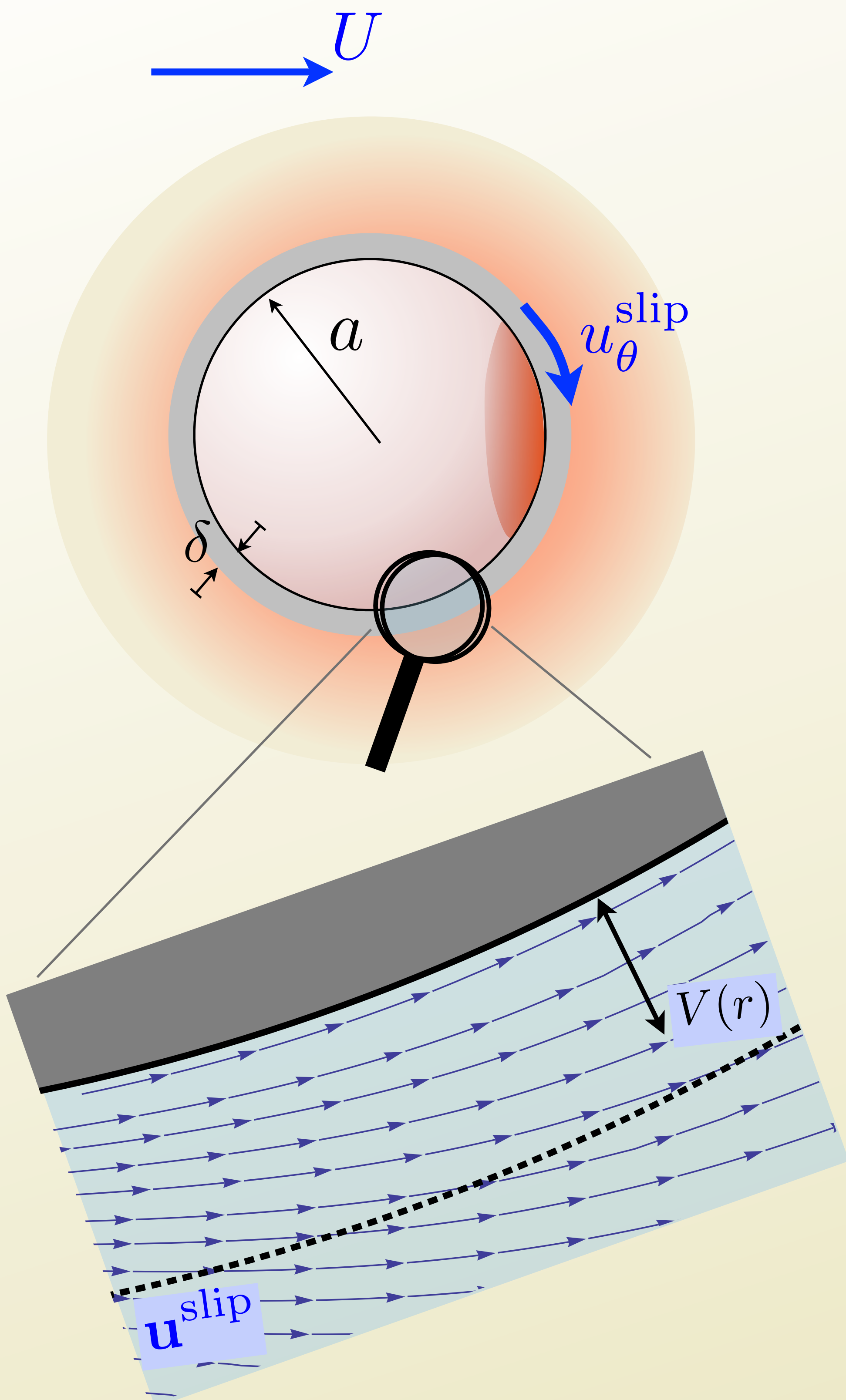
Blake *J. Fluid Mech* **46**, 199–208 (1971)
Lighthill *Commun. Pure Appl. Math.* **5**, 109–118 (1952)

SOLUTE PROFILE



Outside the boundary layer the solute is conserved

$$\cancel{\partial_t c + \mathbf{u}\cdot\nabla c} - D\nabla^2 c = 0 \qquad \boxed{\mathrm{Pe} \ll 1}$$

Rate of transport across the boundary layer equals rate of production

$$\int_{r=a+\delta} \left(\cancel{u_r^{\text{slip}}} c - D\partial_r c\right) = \int_{r=a} \alpha$$

solve pointwise

$$c(r,\theta) = \sum_{l=0}^{\infty} \frac{a+\delta}{(l+1)D}\, \alpha_l \left(\frac{a+\delta}{r}\right)^{l+1} P_l(\cos(\theta))$$

ANDERSON *Ann. Rev. Fluid Mech.* **21**, 61–99 (1989)
GOLESTANIAN, LIVERPOOL & AJDARI *Phys. Rev. Lett.* **94**, 220801 (2005)
GOLESTANIAN, LIVERPOOL & AJDARI *New J. Phys.* **9**, 126 (2007)

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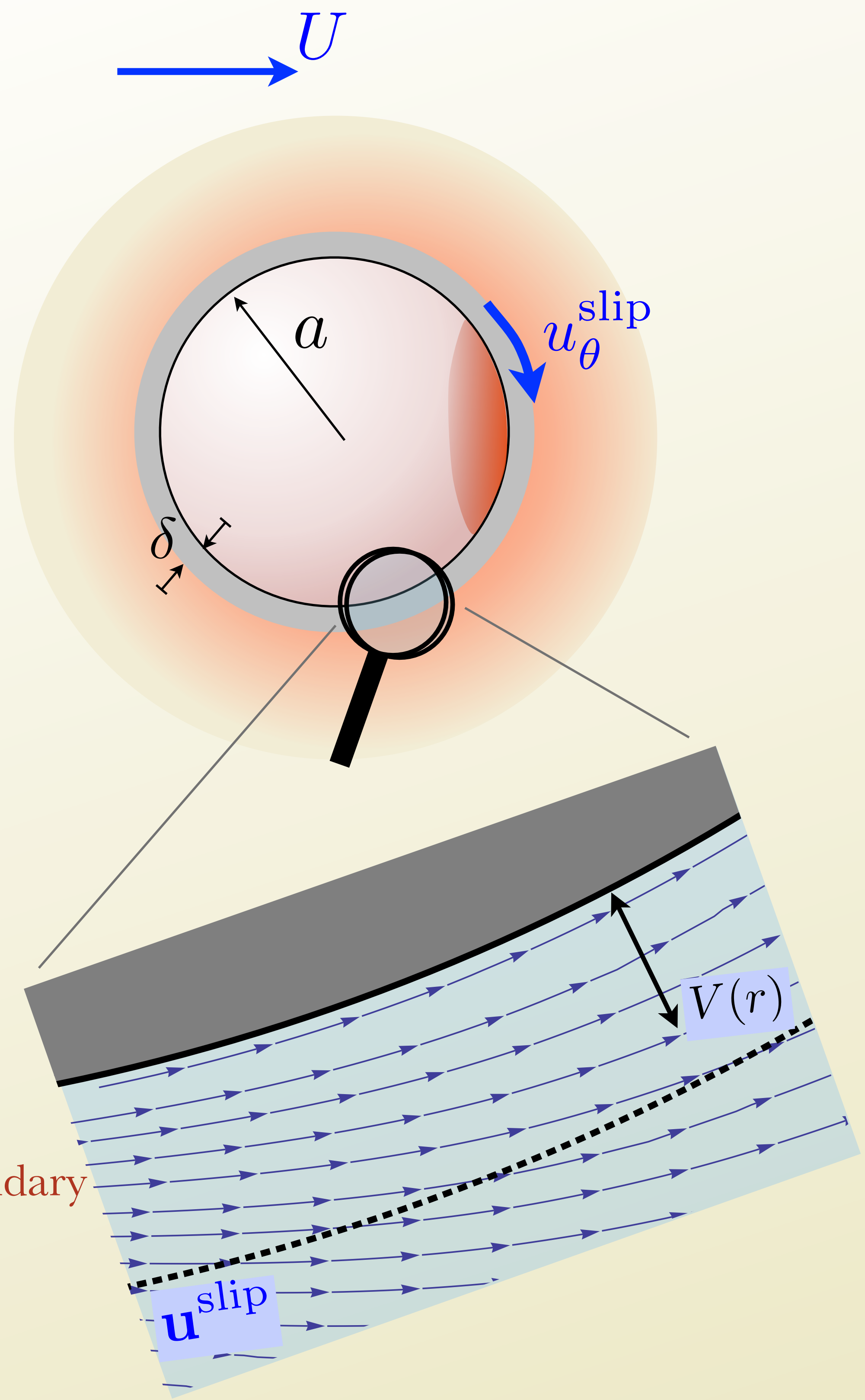
DIGRESSION



Boundary layer analysis neglects

- fluid continuity; radial slip

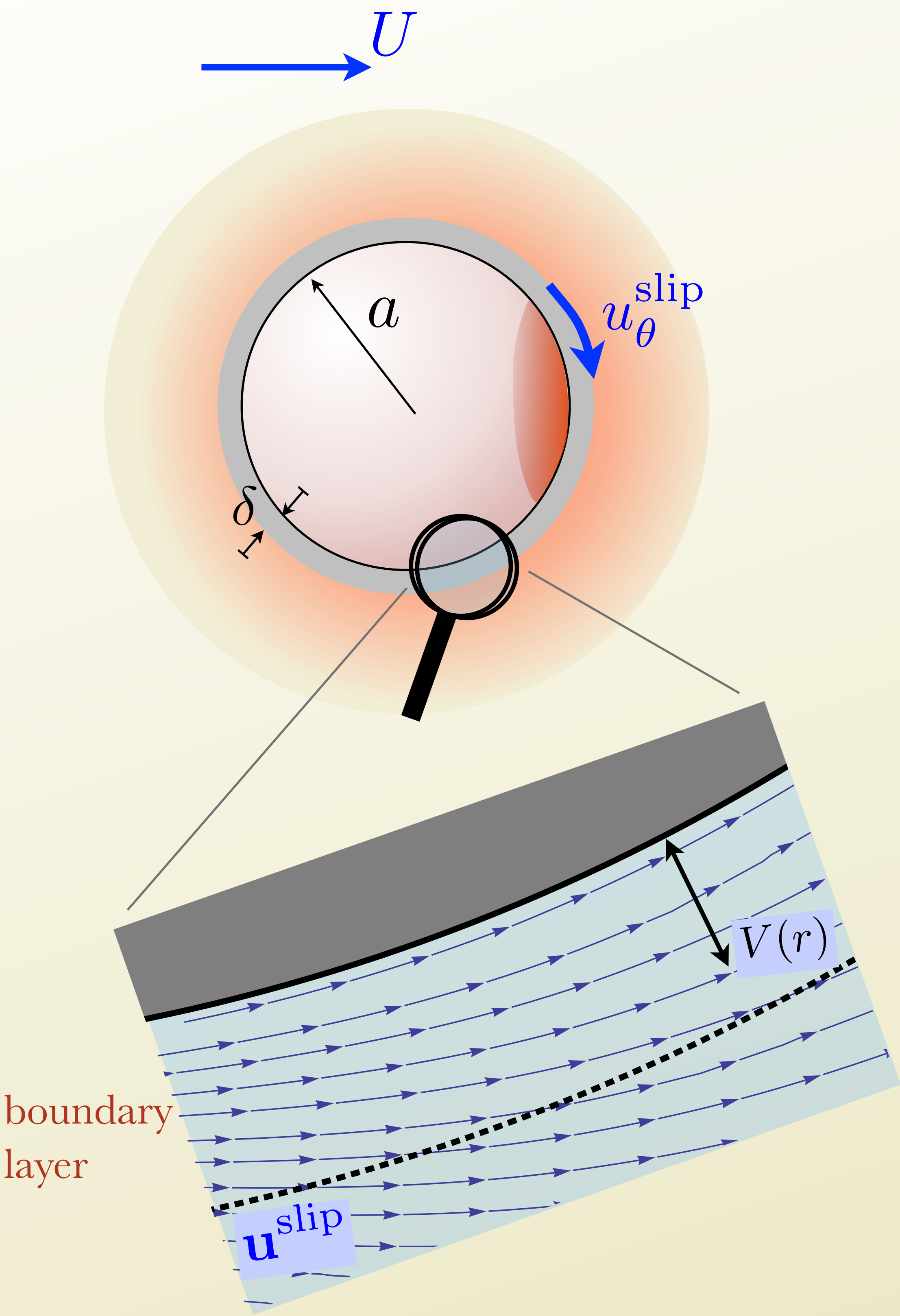
$$\frac{1}{r^2}\partial_r\left(r^2 u_r\right) + \frac{1}{r\sin(\theta)}\partial_\theta\left(\sin(\theta)u_\theta\right) = 0$$

not constant

Scaling $\quad u_r^{\rm slip} \sim \frac{\delta}{a} u_\theta^{\rm slip}$

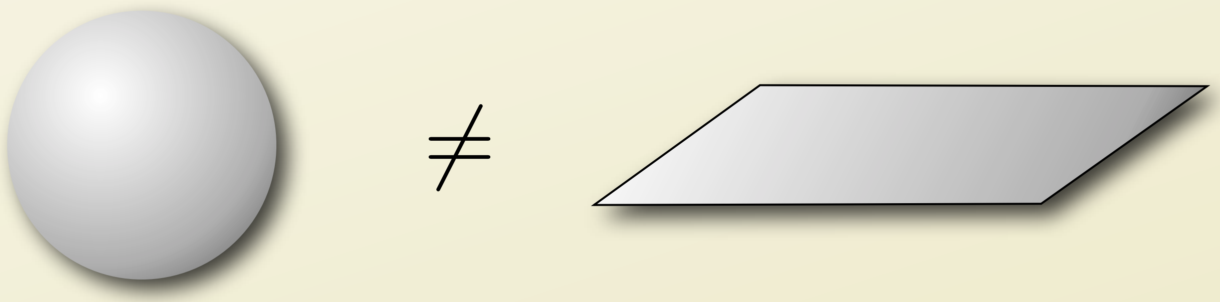
generically small

Digression



Boundary layer analysis neglects

- fluid continuity; radial slip
- topology of the sphere



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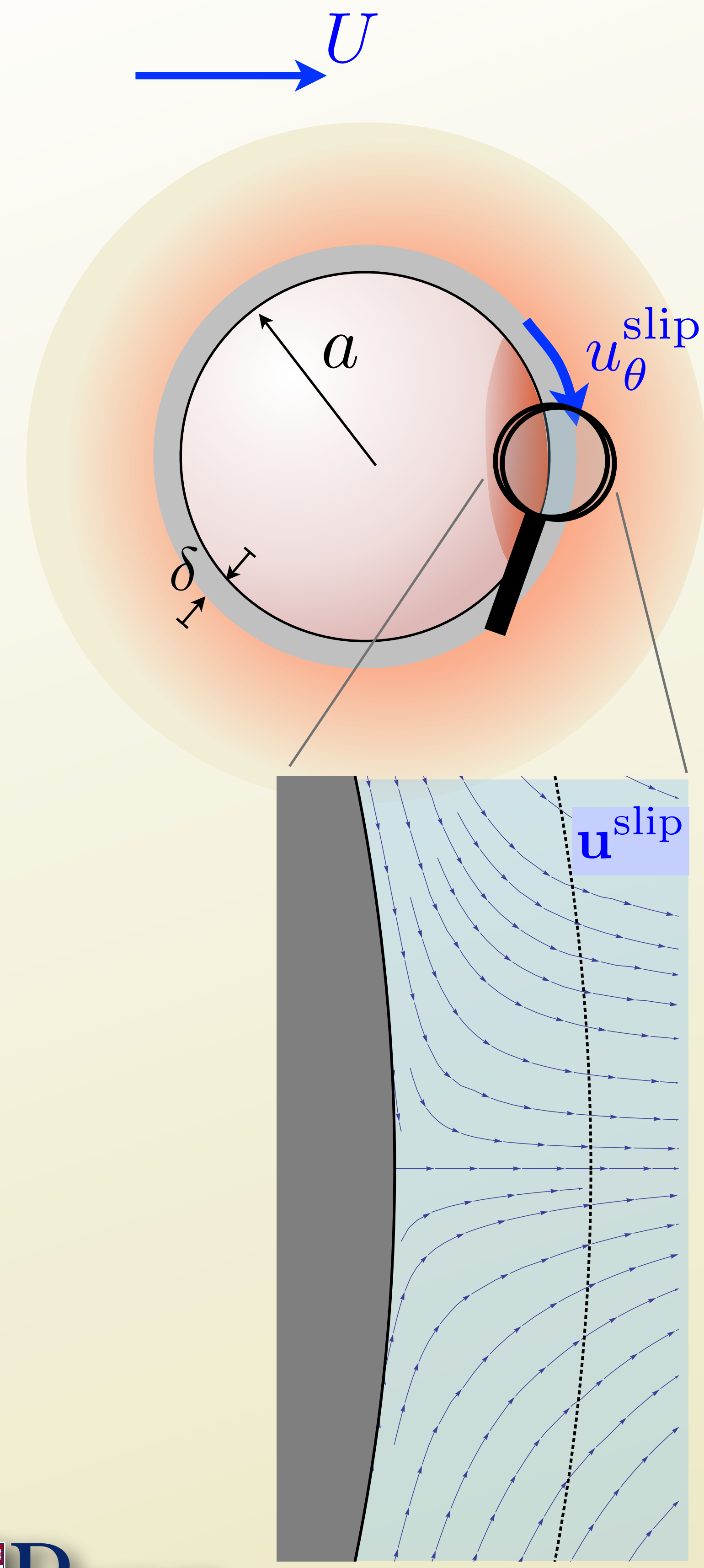

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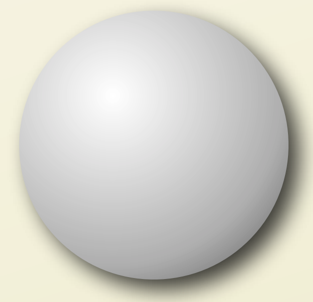

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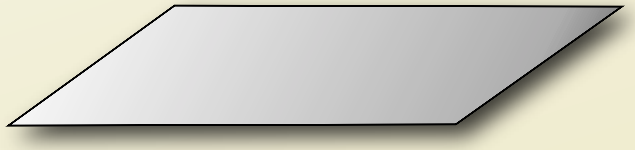

Neglecting the radial slip is not a good approximation near $\theta = 0,\ \pi$

consequence of topology rather than axisymmetry (Poincaré-Hopf)

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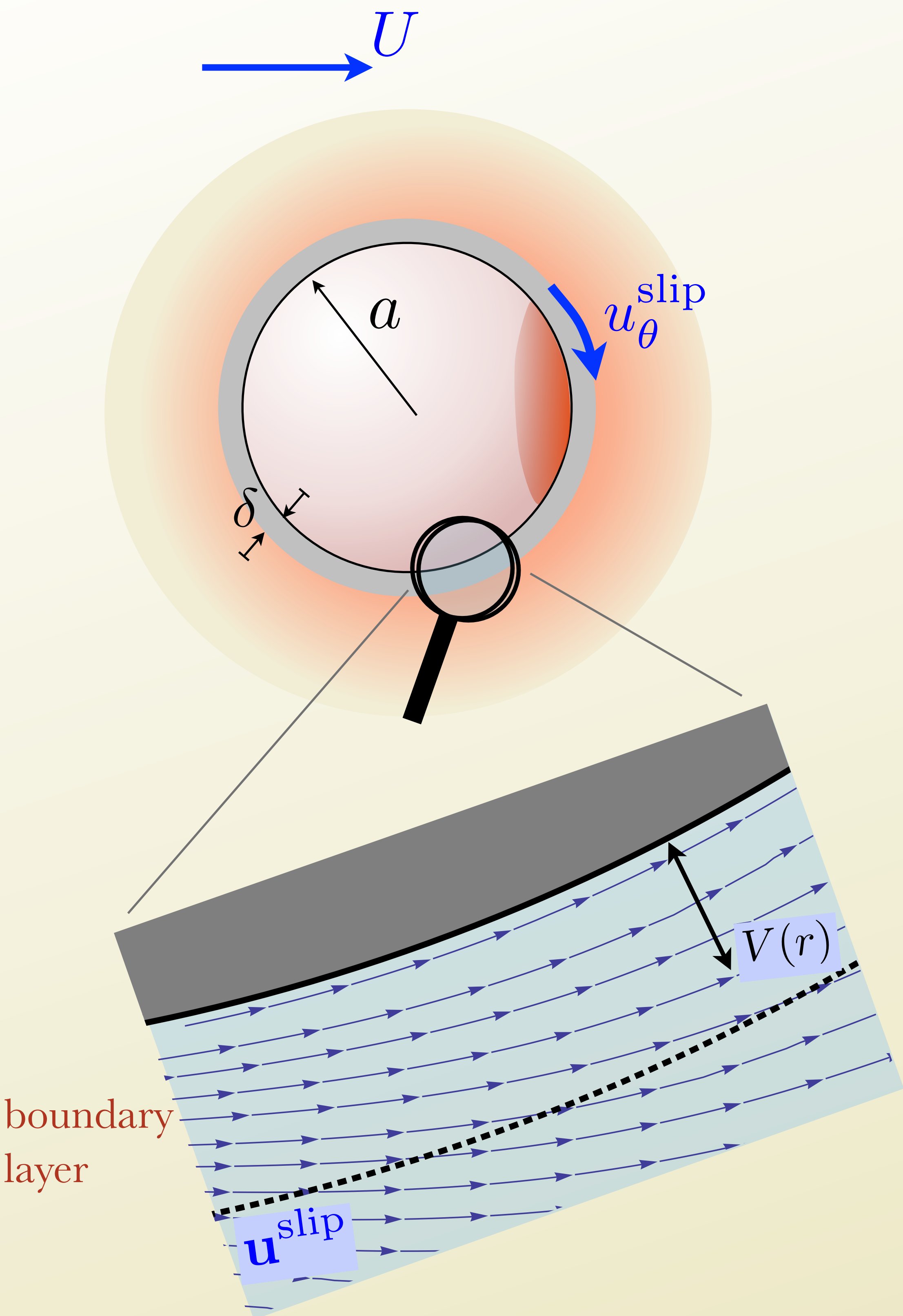
Digression



Boundary layer analysis neglects

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- topology of the sphere $\quad$ Poincaré-Hopf

Such a simple problem can be solved exactly

$$\mathbf{0} = -\nabla p + \mu \nabla^2 \mathbf{u} - \mathbf{e}_r \, c(r,\theta) \, \partial_r V \qquad 0 = \nabla \cdot \mathbf{u}$$

- Spherical particle
- Axisymmetry, steady state
- Single solute species
- Purely radial potential
- Zero Reynolds number
- Zero Péclet number

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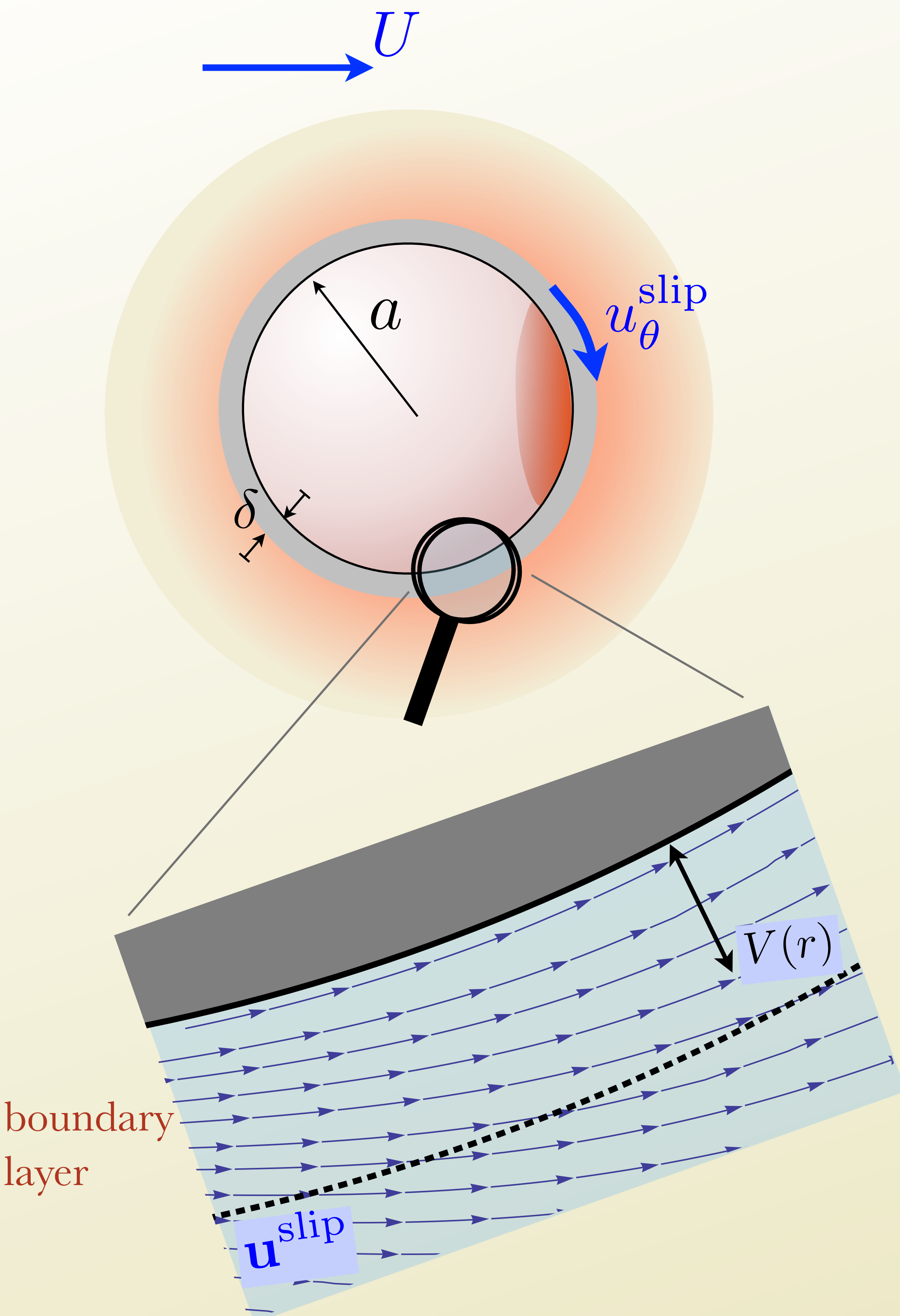
Digression



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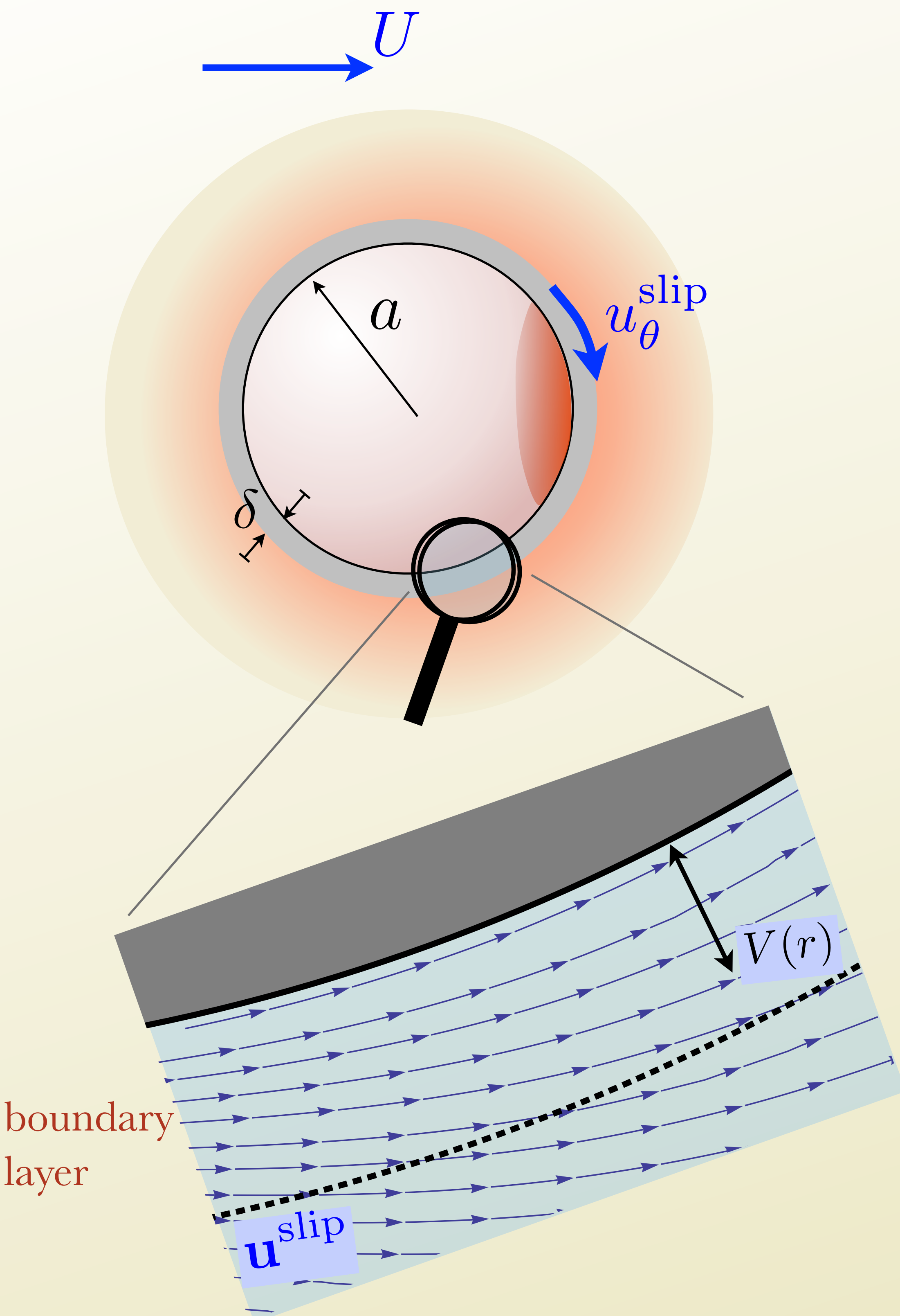
E.g., the speed is

$$U = \frac{2}{3a}\frac{k_B T}{\mu} c_1 \delta^2 \int_0^1 \mathrm{d}z\, z \Big(1 - \underbrace{\frac{\delta}{6a}\frac{3z + 2\delta z^2/a}{(1+\delta z/a)^2}}_{\text{correction}}\Big) \big[1 - \mathrm{e}^{-V/k_B T}\big]$$

Digression



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$$U = \frac{2}{3a} \frac{k_B T}{\mu} c_1 \delta^2 \int_0^1 \mathrm{d}z\, z \left(1 - \frac{\delta}{6a} \frac{3z + 2\delta z^2/a}{(1+\delta z/a)^2}\right) \left[1 - \mathrm{e}^{-V/k_B T}\right]$$

scaling $\quad U \sim \dfrac{k_B T \alpha_1}{\mu D} \delta^2 \left(1 + \frac{\delta}{a}\right)$

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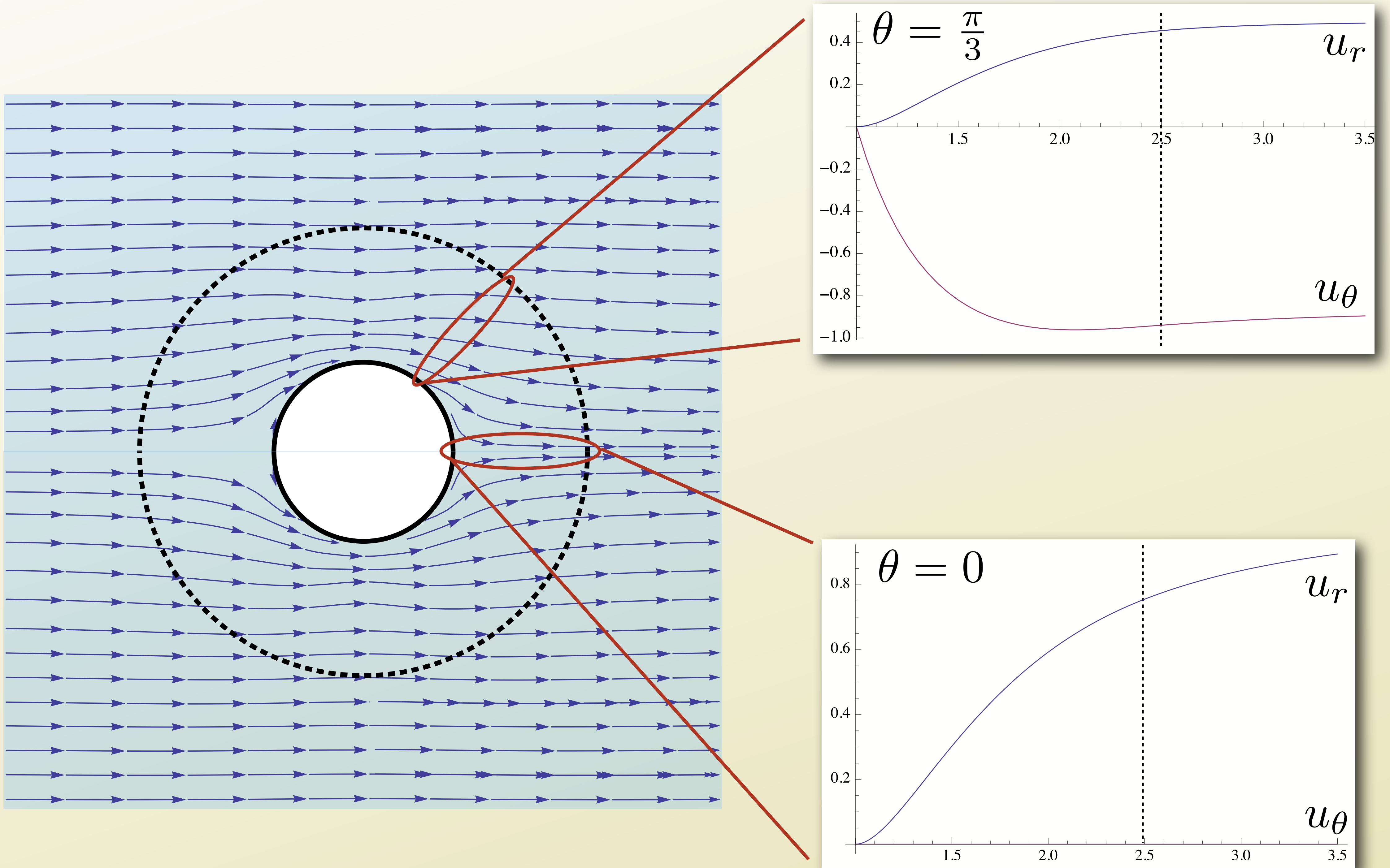
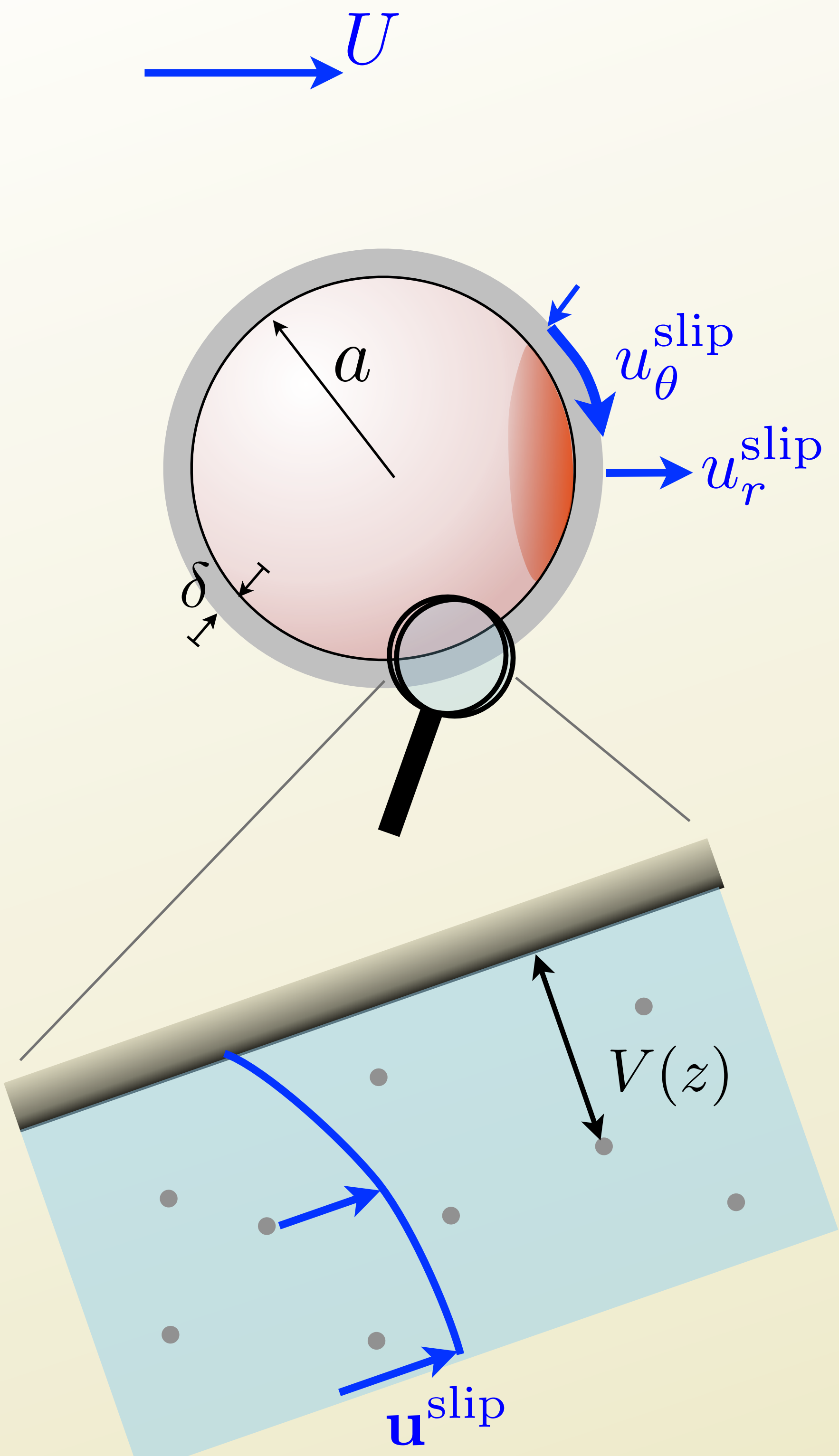
Sabass, Seifert *Phys. Rev. Lett.* **105**, 218103 (2010)

Boundary layer flow



What changes if the Péclet number is large?



Basic mechanism remains unchanged

- concentration gradients drive tangential flow in a thin boundary layer

postulate $\quad \mathbf{u}^{\text{slip}} = m^A \nabla_{\|} c$

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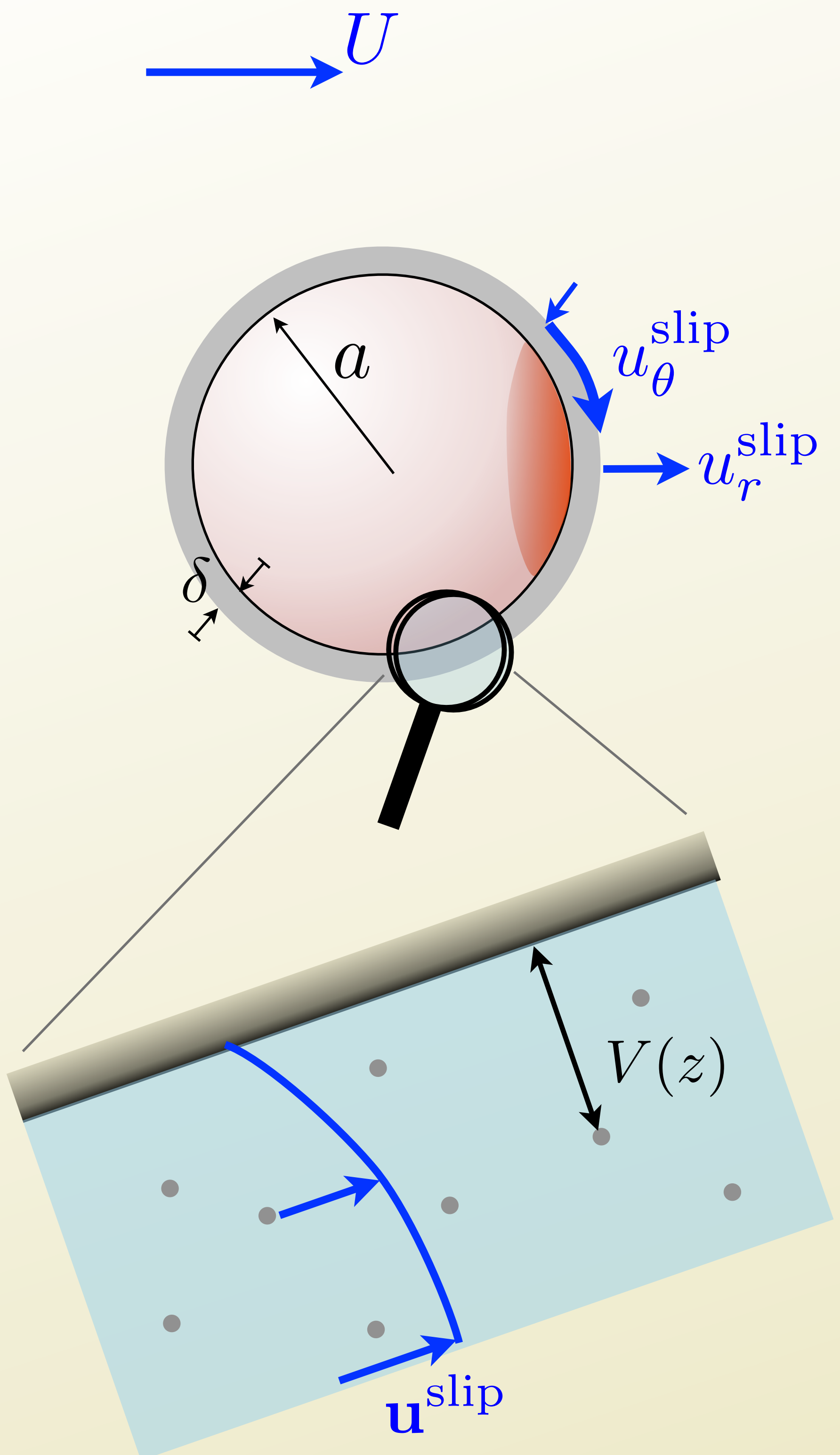

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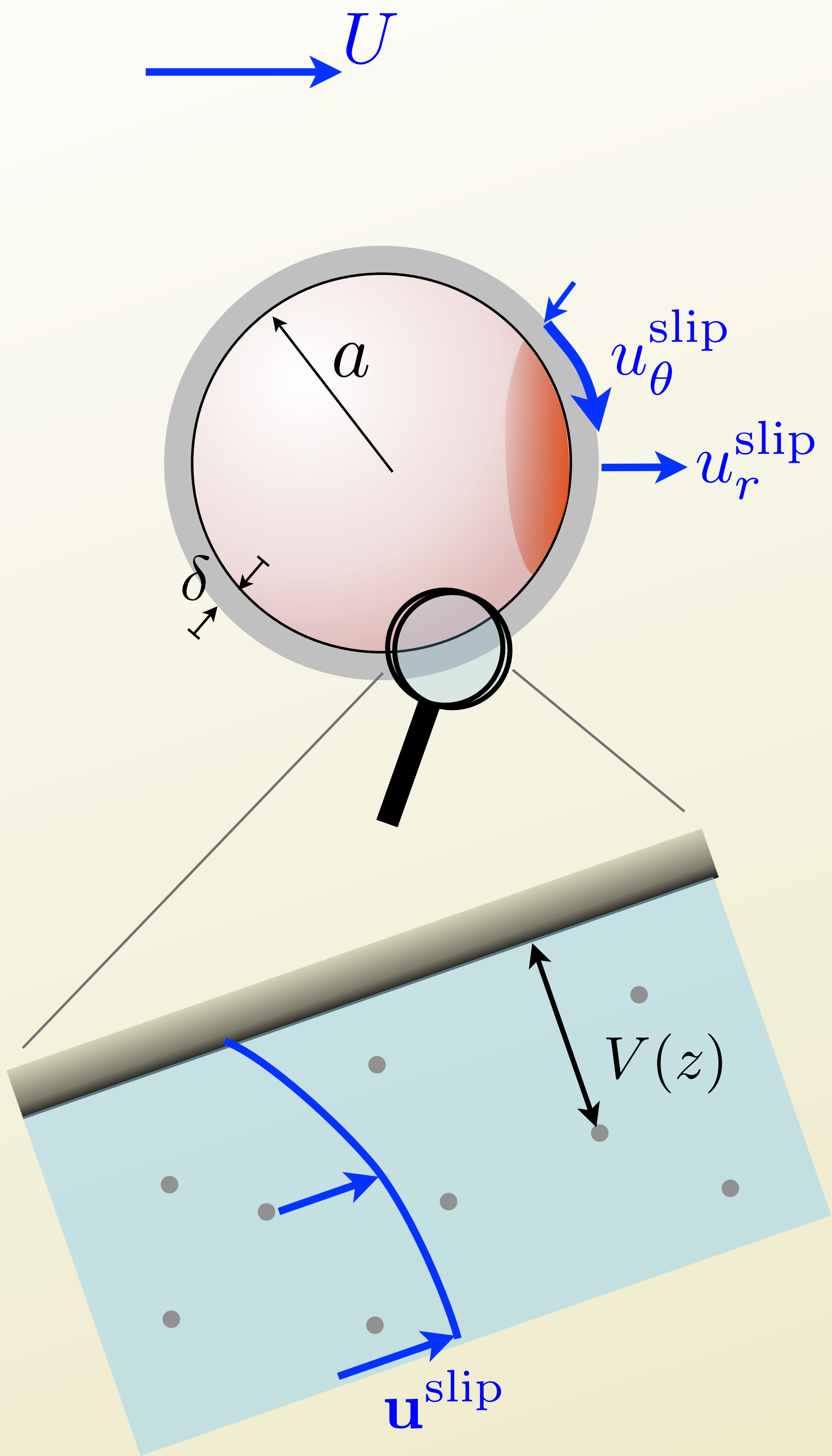
postulate $\quad \mathbf{u}^{\text{slip}} = m^A \nabla_{\parallel} c$

But ...

- if the solute does not diffuse then it will only be found where it is produced
- tangential slip only generated within the active patch
- radial influx at the boundary and outflux from the interior

Penn

SOLUTE TRANSPORT



Outside the boundary layer the solute is conserved

$$\partial_t c + \mathbf{u} \cdot \nabla c - \cancel{D\nabla^2 c} = 0 \qquad \boxed{\mathrm{Pe} \gg 1}$$

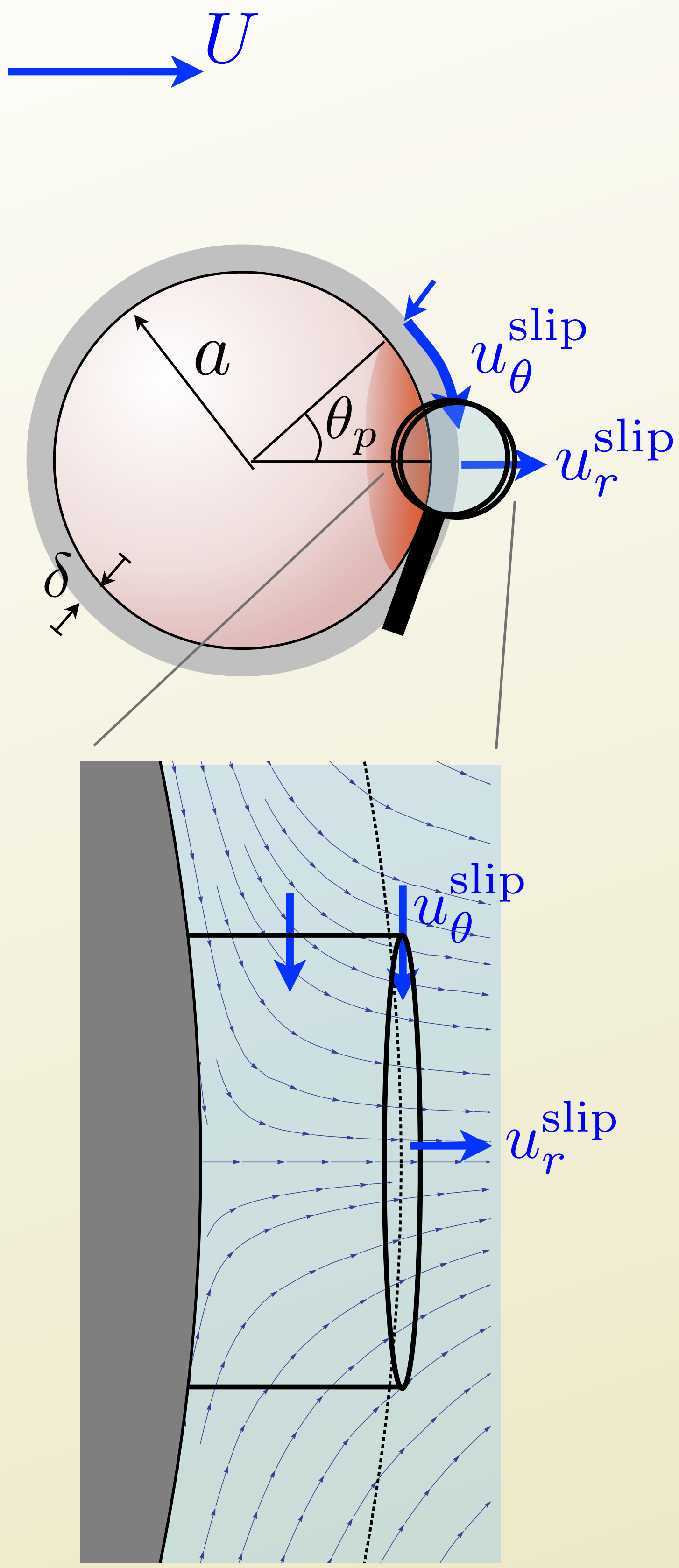
Rate of transport across the boundary layer equals rate of production

$$\int_{r=a+\delta} \left(u_r^{\mathrm{slip}} c - \cancel{D\partial_r c}\right) = \int_{r=a} \alpha$$

radial slip is important

Penn

Radial slip



Radial outflux

$$2\pi(a+\delta)^2 \int_0^{\theta_q} \mathrm{d}\theta \,\sin(\theta)\, u_r^{\mathrm{slip}}$$

Tangential influx

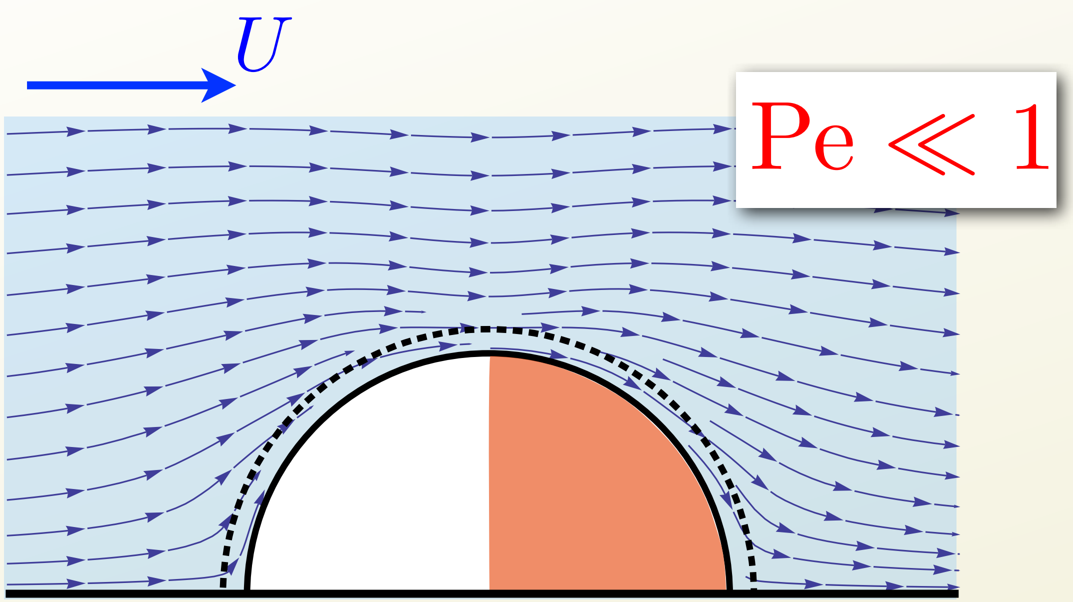
$$-2\pi \int_a^{a+\delta} \mathrm{d}r \, r \sin(\theta)\, u_\theta(r,\theta)$$

$$\approx -2\pi a \delta \beta \,\sin(\theta)\, u_\theta^{\mathrm{slip}}$$

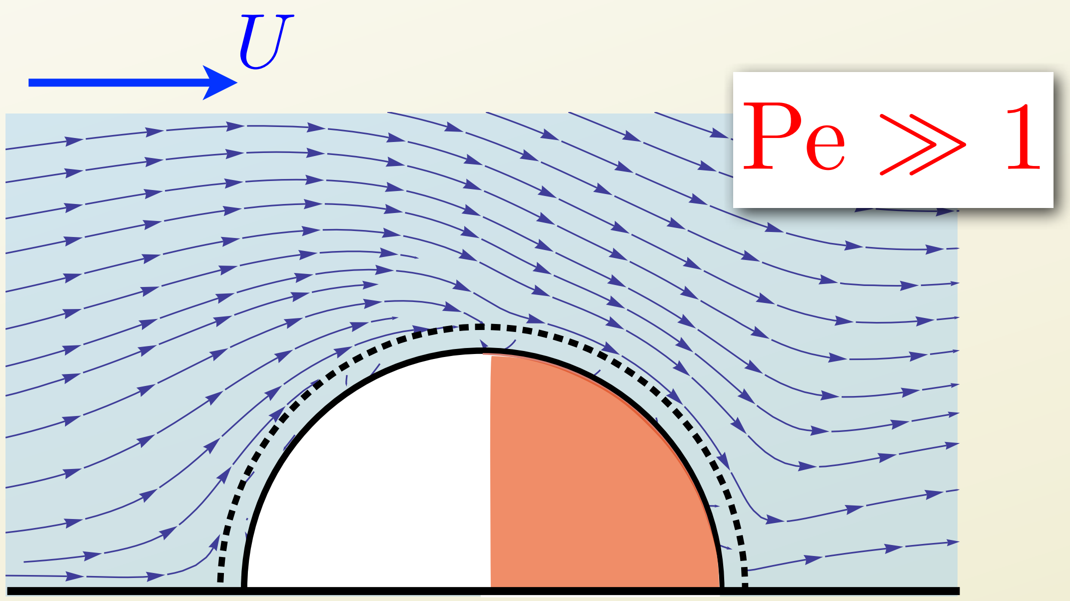
$\mathcal{O}(1)$

conserved away from $\theta = 0$

So, what's different?



$$U \sim \frac{m^D \bar{\alpha}}{D} \sin^2(\theta_p)$$



$$U \sim \left(\frac{m^A \bar{\alpha}}{\delta \ln\left(\frac{a}{\delta}\right)} \right)^{1/2} \sin^3\left(\tfrac{1}{2}\theta_p\right)$$

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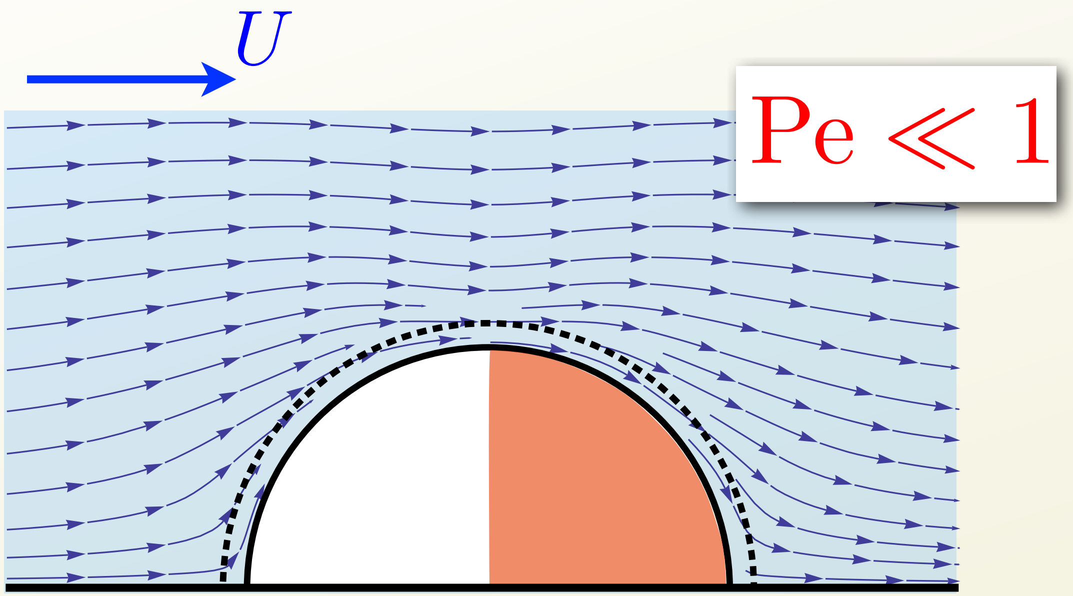
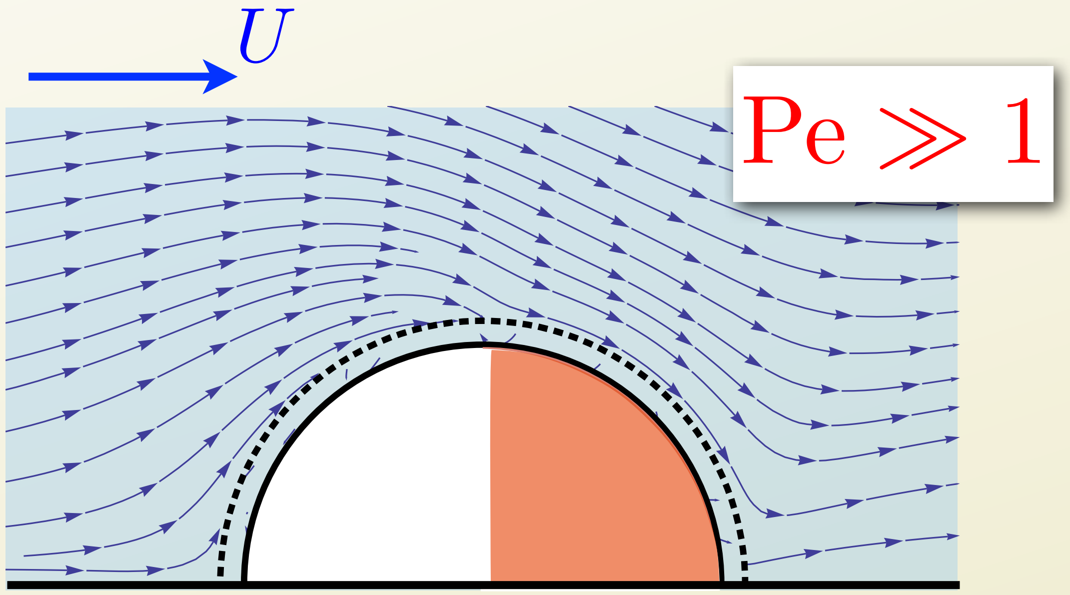
So, what's different?





$$U \sim \frac{m^D \bar{\alpha}}{D} \sin^2(\theta_p)$$

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Transport balance $$\int_{r=a+\delta} \left(u_r^{\text{slip}} c - D\partial_r c\right) = \int_{r=a} \alpha$$

$\propto |u_\theta^{\text{slip}}|$

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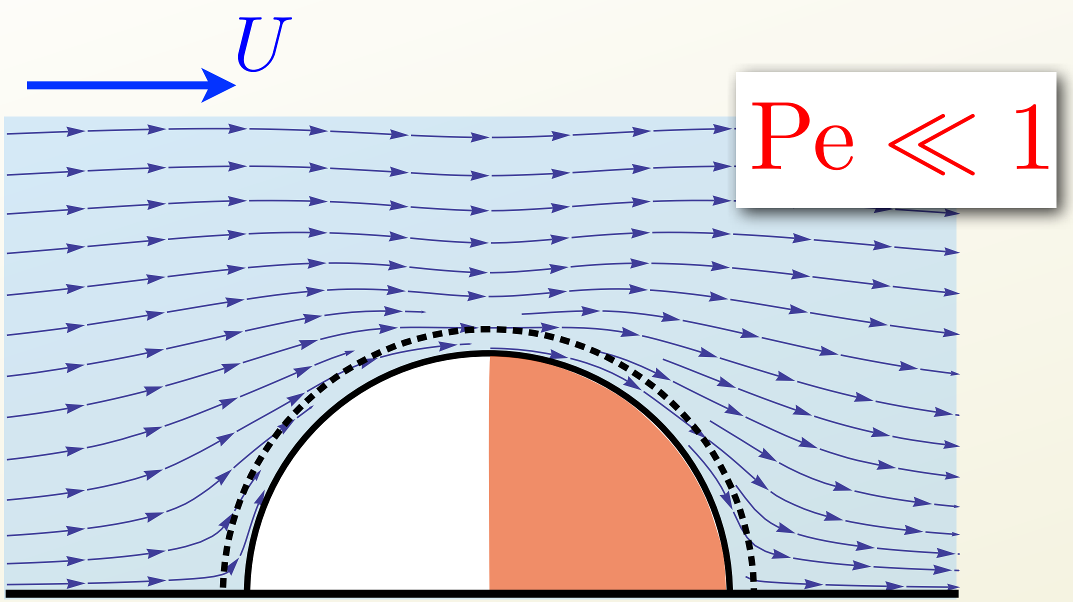
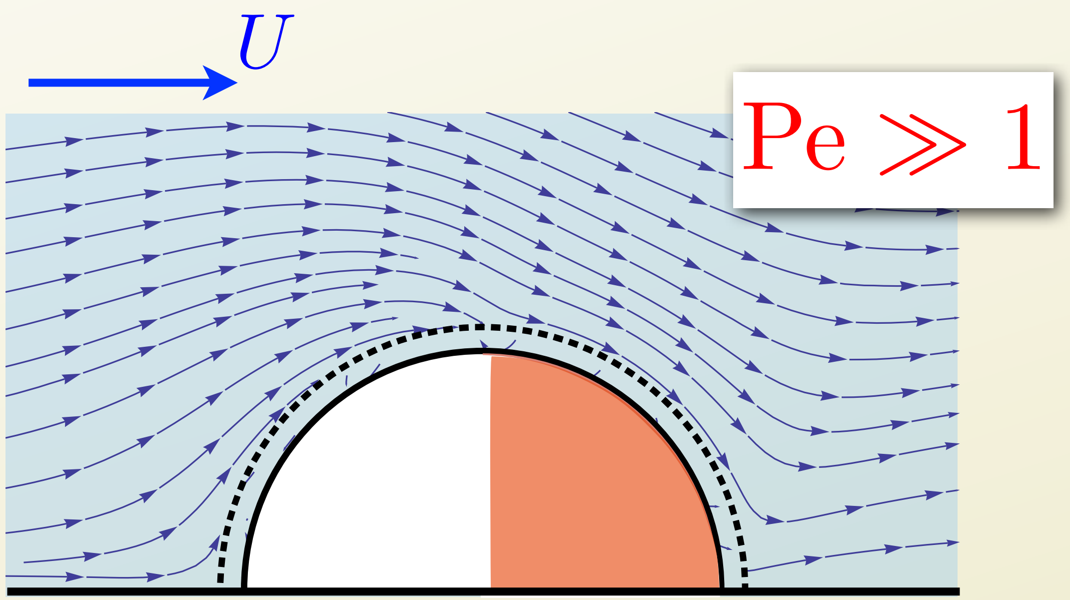
So, what's different?





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Transport balance $$\int_{r=a+\delta} \left(u_r^{\text{slip}} c - D\partial_r c\right) = \int_{r=a} \alpha$$

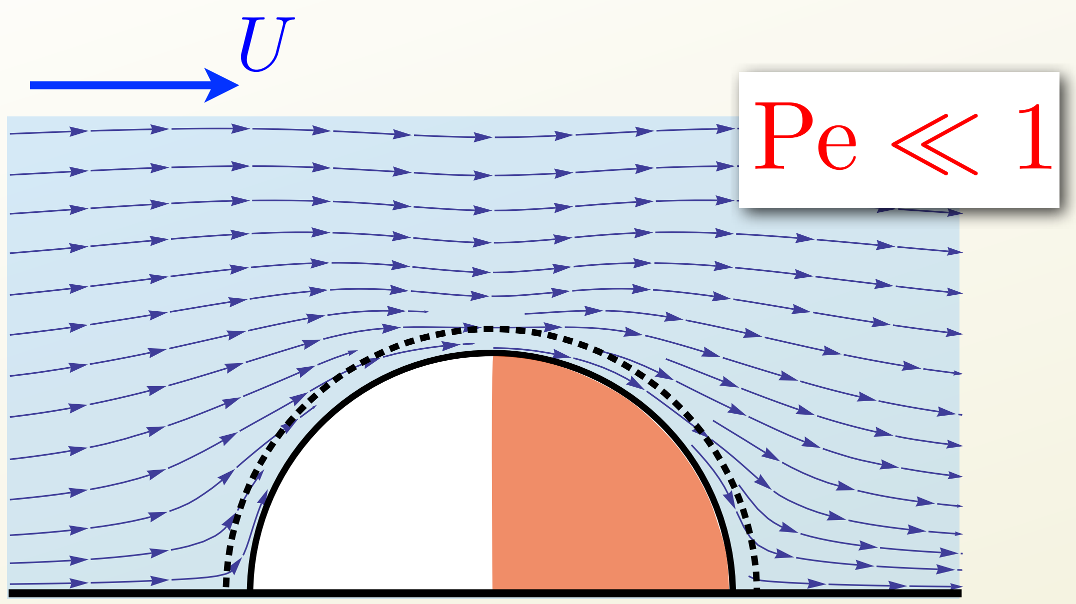
$\propto |\sin(\theta) u_\theta^{\text{slip}}|$

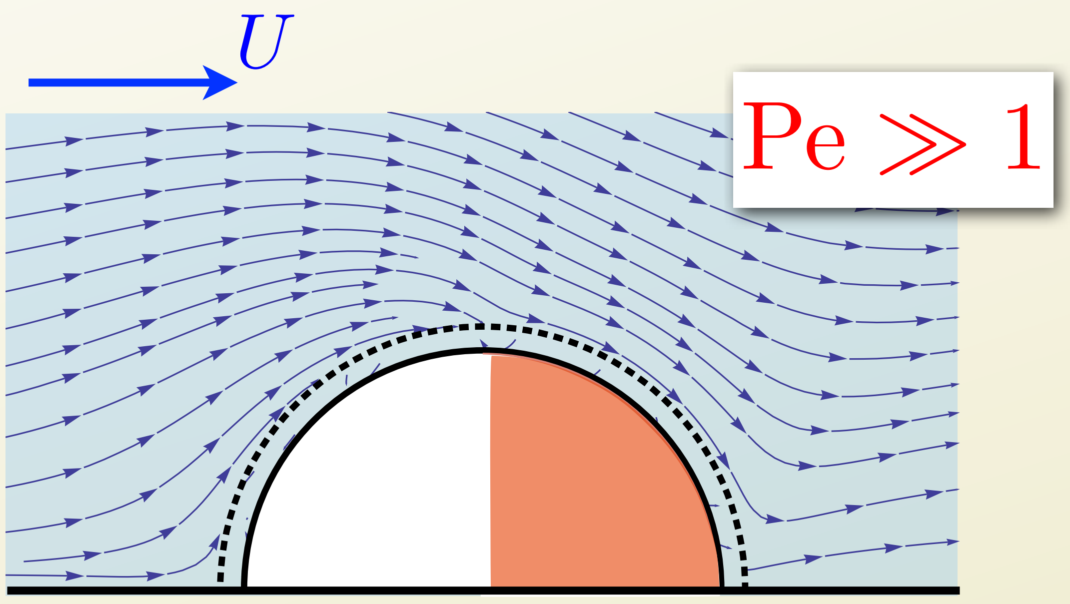
by fluid continuity

$\propto |u_\theta^{\text{slip}}|$

by postulate

So, what's different?





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$$U \sim \left(\frac{m^A \bar{\alpha}}{\delta \ln\left(\frac{a}{\delta}\right)} \right)^{1/2} \sin^3\left(\tfrac{1}{2}\theta_p\right)$$

$$U \sim \alpha_1 \sim \int_{\cos(\theta_p)}^{1} \mathrm{d}s\, s\bar{\alpha} \sim \sin^2(\theta_p)$$

first Legendre
coefficient of activity

$$U \sim \int_0^{\theta_p} \mathrm{d}\theta\, \sin^2(\theta) u_\theta^{\mathrm{slip}} \sim \sin^2\left(\tfrac{1}{2}\theta_p\right)$$

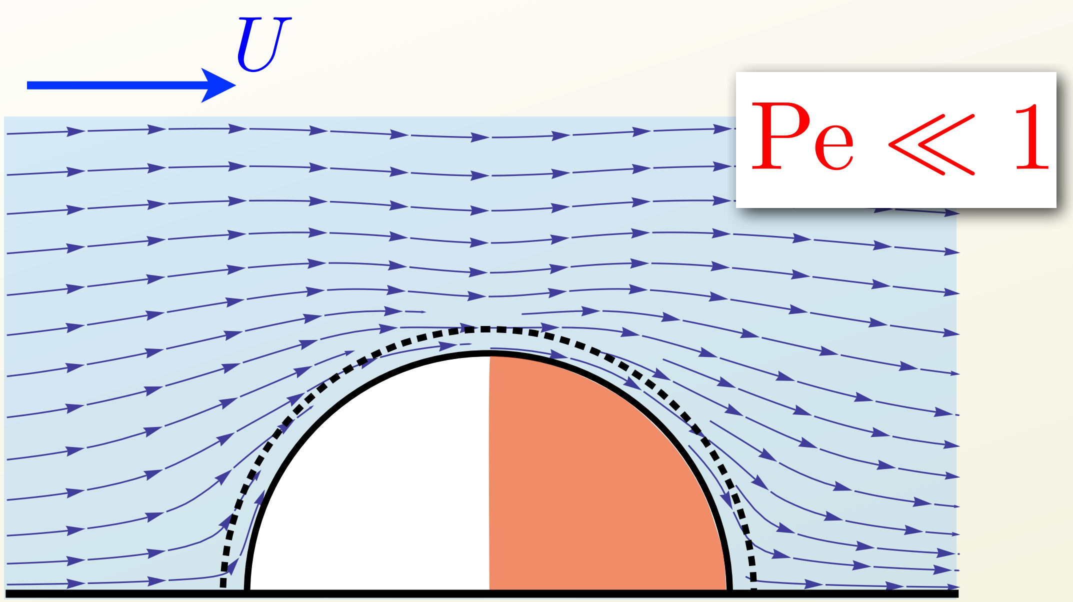
by Stone & Samuel

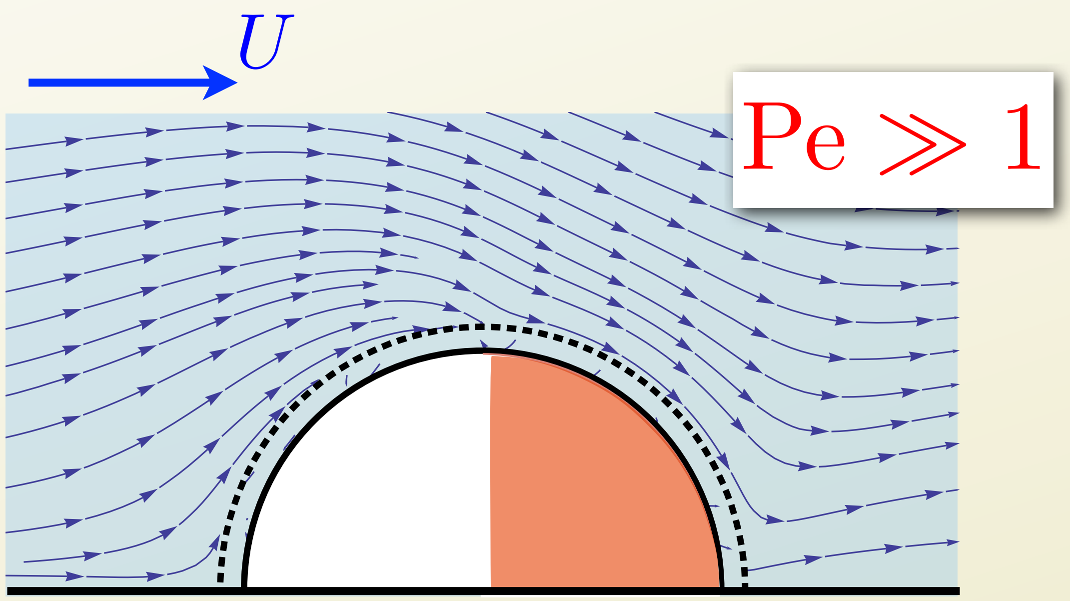
tangential flux
is conserved

Penn

Stone & Samuel *Phys. Rev. Lett.* **77**, 4102–4104 (1996)

So, what's different?

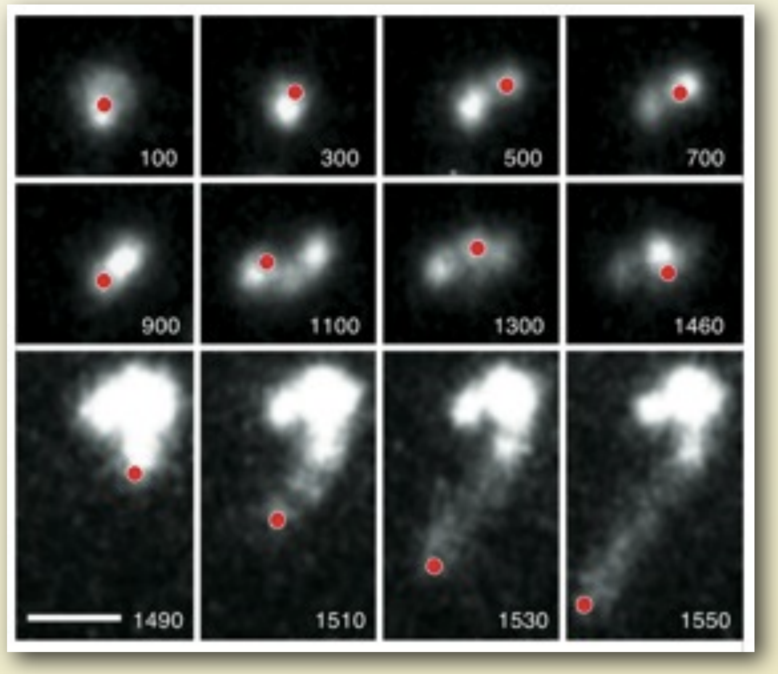




$$U \sim \frac{m^D \bar{\alpha}}{D} \sin^2(\theta_p)$$

$$U \sim \left(\frac{m^A \bar{\alpha}}{\delta \ln\left(\frac{a}{\delta}\right)} \right)^{1/2} \sin^3\left(\tfrac{1}{2}\theta_p\right)$$

does not vanish for
total coverage $\theta_p \to \pi$

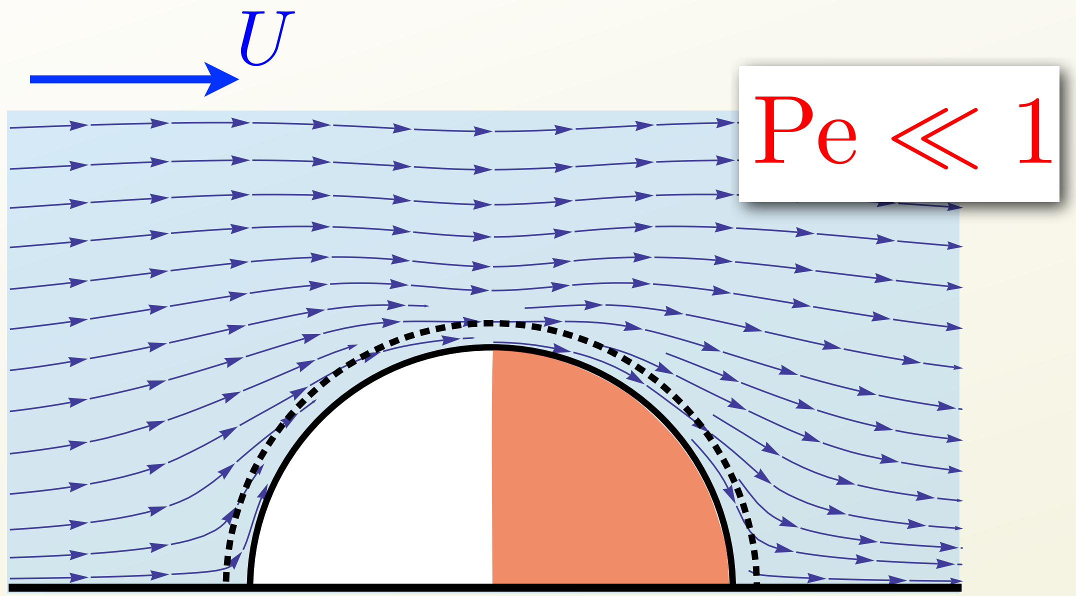


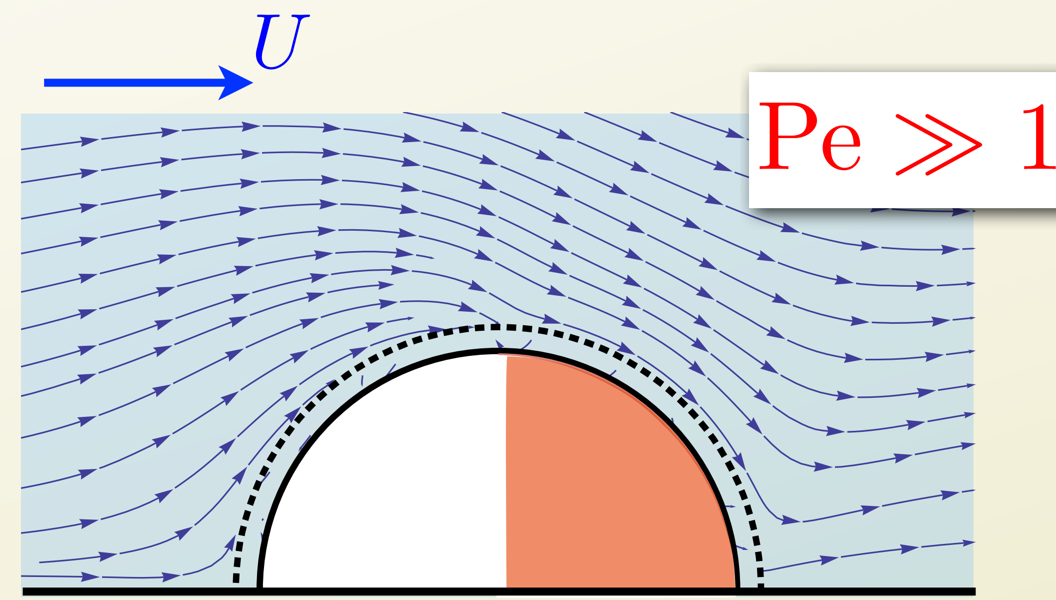
Cameron et al *Proc. Natl. Acad. Sci. USA* **96**, 4908–4913 (1999)

- is state of total coverage unstable?
- if so, what is the critical Péclet number for the instability?

Penn

Different scenarios





$$U \sim \frac{m^D \bar{\alpha}}{D} \sin^2(\theta_p)$$

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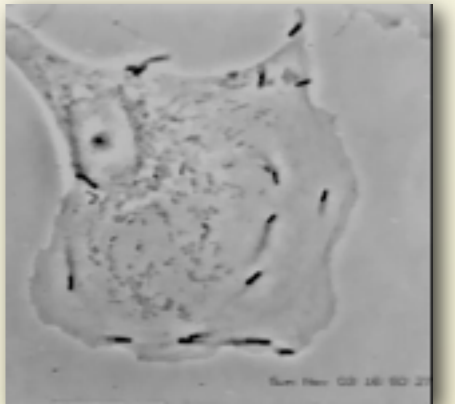

Four scenarios

repulsive producer
$V > 0, \ \alpha > 0$

attractive producer
$V < 0, \ \alpha > 0$

repulsive consumer
$V > 0, \ \alpha < 0$

attractive consumer
$V < 0, \ \alpha < 0$

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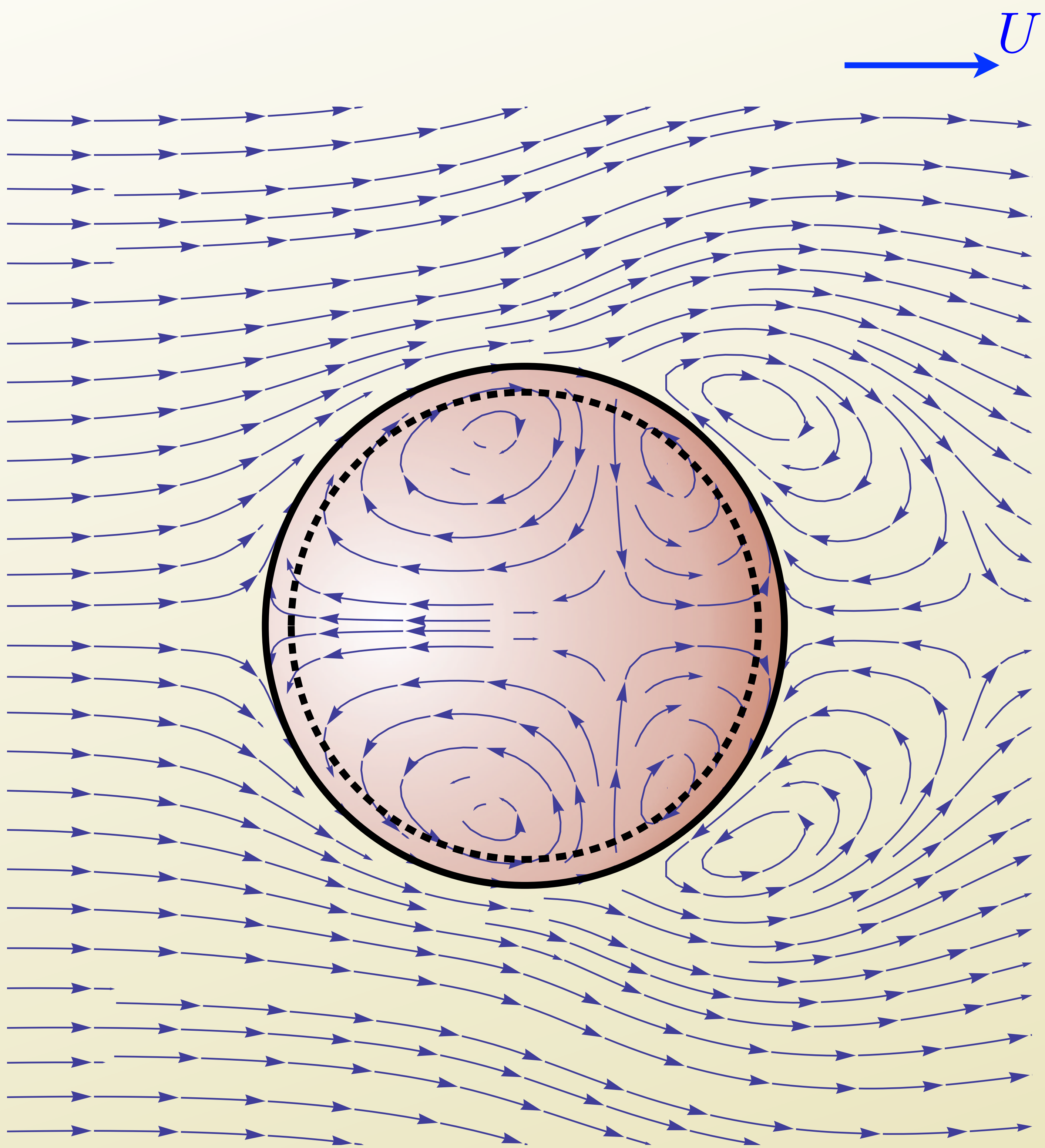
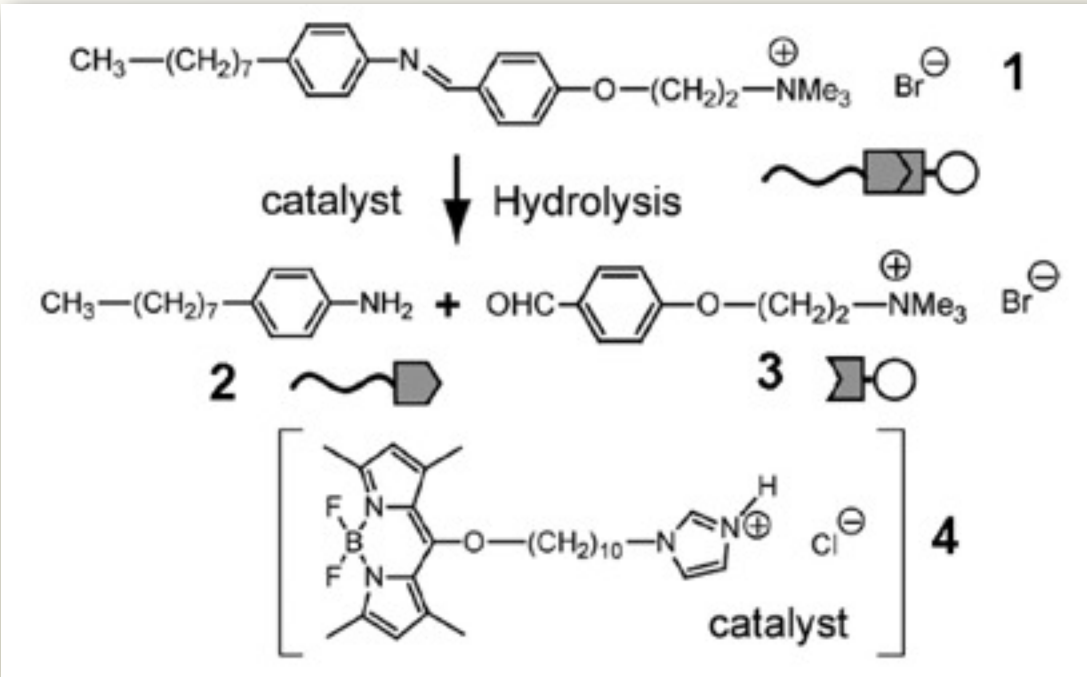
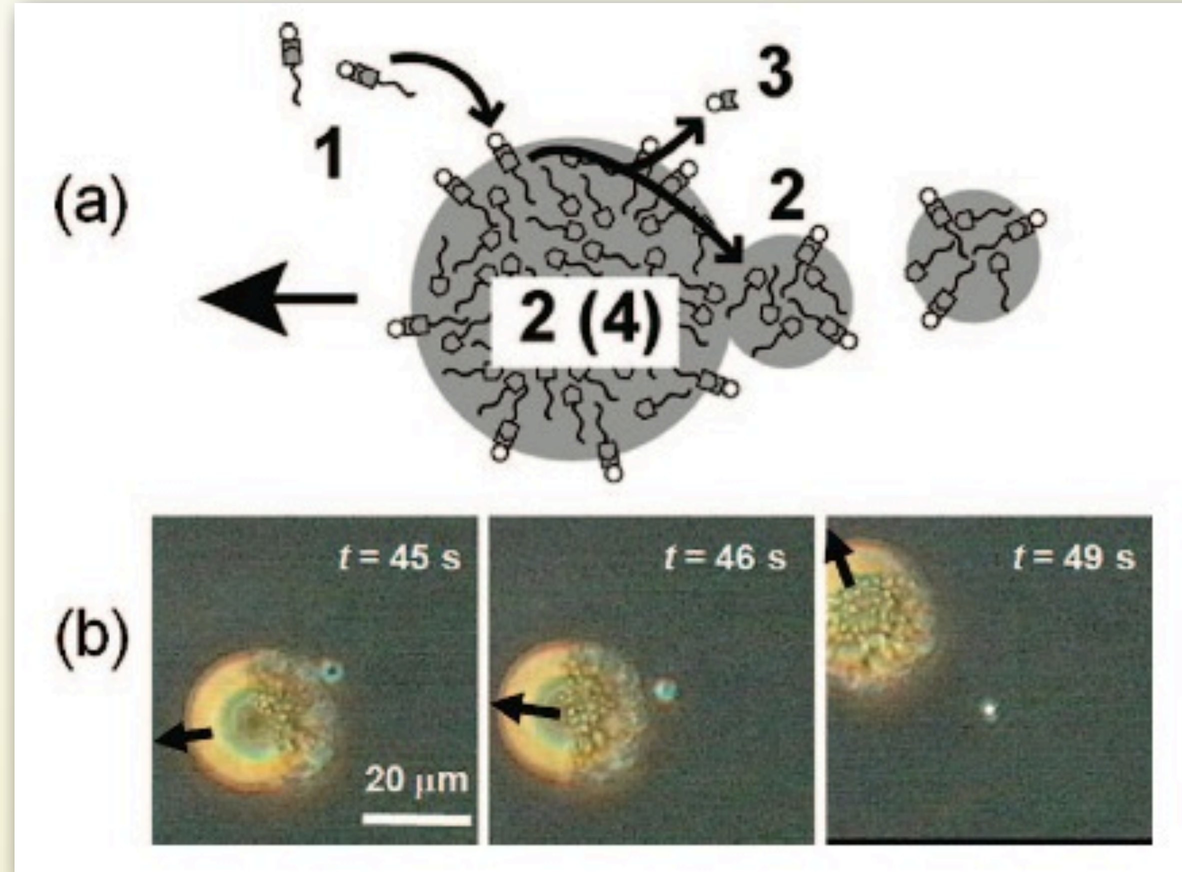
Fluid droplets



Fluid droplets

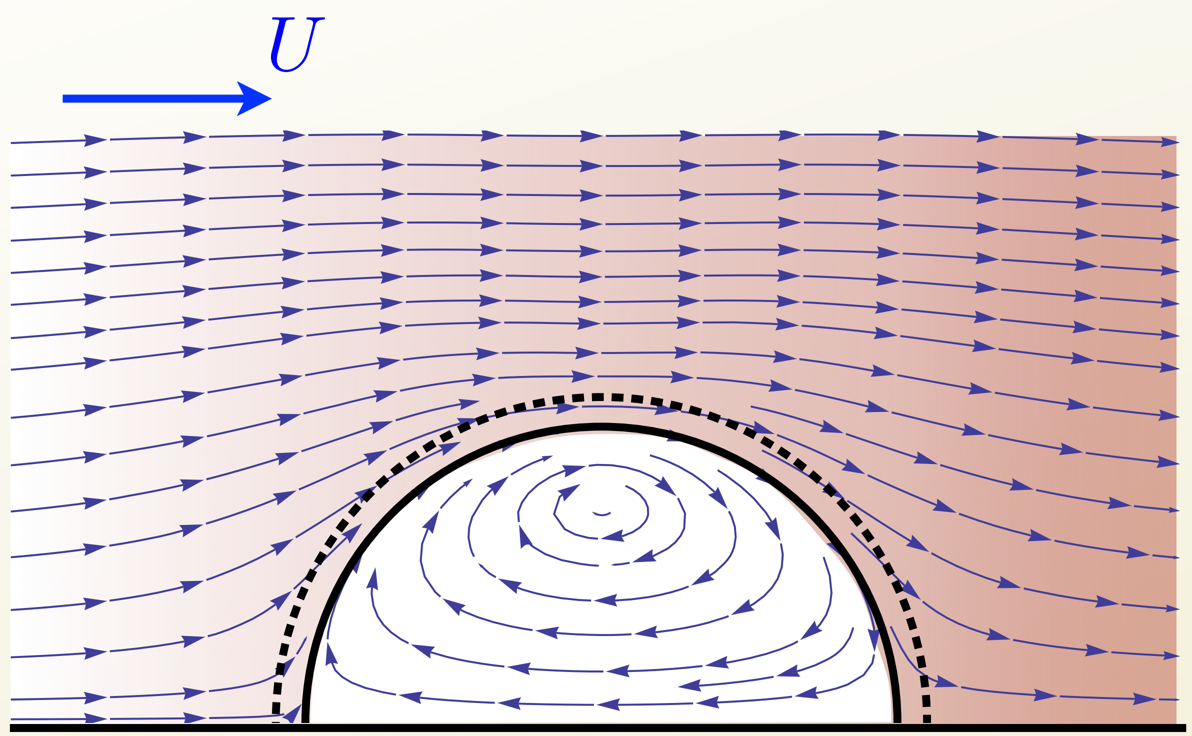




- Droplets containing a catalyst dispersed in a bulk fuel
- Fuel hydrolysed at the surface
- Waste product accumulates on the surface and is released at the rear
- Self-maintained surface tension gradients drive Marangoni flows

Toyota et al *J. Am. Chem. Soc.* **131**, 5012–5013 (2009)

Fluid droplets



Rest frame of the particle

- Spherical particle
- Axisymmetry, steady state
- Single solute species
- Purely radial potential with compact support
- Zero Reynolds number
- Zero Péclet number

Stress balance at the interface

$$\left(\sigma_{ab} - \tilde{\sigma}_{ab}\right) n_b - \gamma (2H)\, n_a + \nabla_a \gamma = 0$$

external fluid stress

internal fluid stress

surface normal

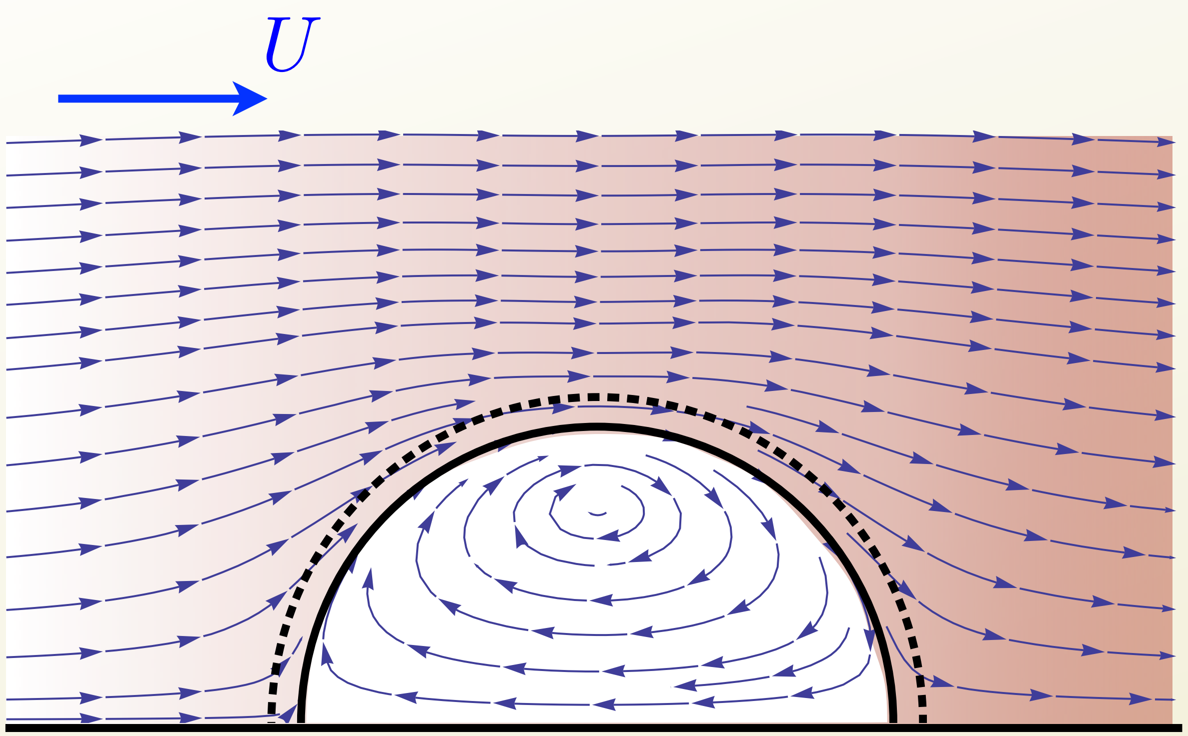
surface tension

mean curvature

Marangoni stress

Levich, Krylov *Ann. Rev. Fluid Mech.* **1**, 293–316 (1969)
Levich, Kuznetsov *Dokl. Akad. SSSR* **146**, 145–147 (1962)
Rednikov, Ryazantsev, Verlarde *Phys. Fluids* **6**, 452–468 (1994)

Fluid droplets



Rest frame of the particle

- Spherical particle
- Axisymmetry, steady state
- Single solute species
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- Zero Reynolds number
- Zero Péclet number

Solve as before; e.g., the speed is

$$U = \frac{k_B T}{3\mu} \frac{\mu + \tilde{\mu}}{\mu + \frac{3}{2}\tilde{\mu}} \frac{c_1 \delta^2}{a} \int_0^1 \mathrm{d}z \left(\left(2 + \frac{\tilde{\mu}}{\mu + \tilde{\mu}} \right) z + \frac{\mu}{\mu + \tilde{\mu}} \frac{a}{\delta} - \frac{\delta}{2a} \frac{\tilde{\mu}}{\mu + \tilde{\mu}} \frac{3z^2 + 2\delta z^3/a}{(1 + \delta z/a)^2} \right) \left[1 - \mathrm{e}^{-V/k_B T} \right]$$

limit $\frac{\mu}{\tilde{\mu}} \longrightarrow 0$
(solid particle)

$$U \sim \frac{k_B T \alpha_1}{\mu D} \delta^2$$

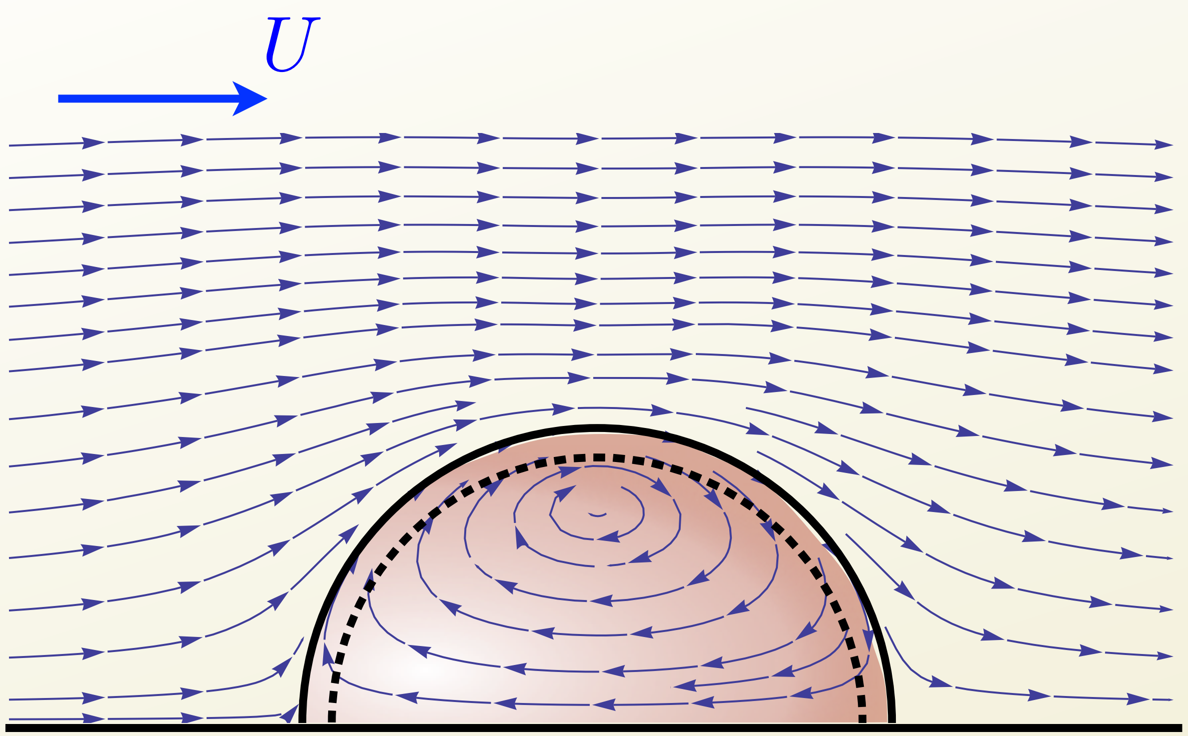
independent of particle size

$$U \sim \frac{k_B T \alpha_1}{\mu D} a\delta$$

proportional to particle radius

limit $\frac{\tilde{\mu}}{\mu} \longrightarrow 0$
(gas bubble)

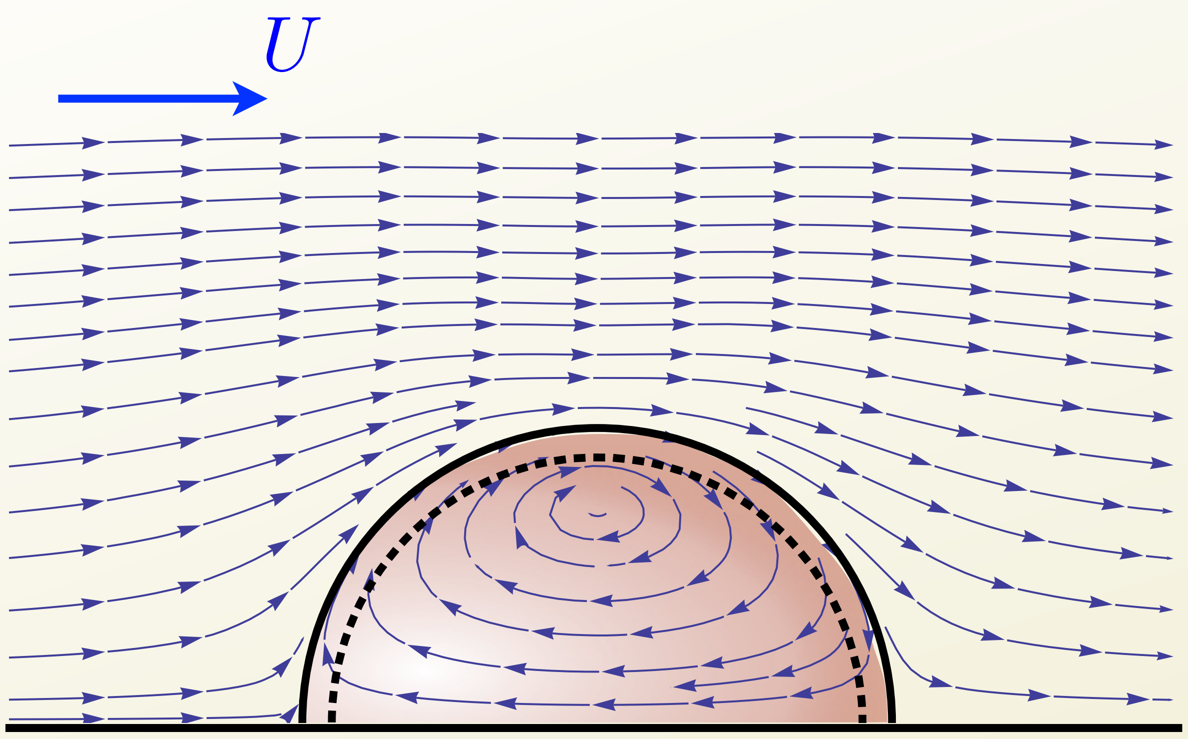
Activity on the inside



Rest frame of the particle

- No need for a favourable environment -- take everything you need with you!
- “Clean” system; everything is internal
- Only interaction between droplets is hydrodynamic

ACTIVITY ON THE INSIDE



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Solve as before; e.g., the speed is

$$U = \frac{k_B T}{3\mu} \frac{\mu}{\mu + \frac{3}{2}\tilde{\mu}} \tilde{c}_1 \tilde{\delta} \int_0^1 \mathrm{d}z \left(1 - 5\frac{\tilde{\delta}}{a} z + 6\left(\frac{\tilde{\delta}}{a}\right)^2 z^2 - 2\left(\frac{\tilde{\delta}}{a}\right)^3 z^3\right)\left[1 - \mathrm{e}^{-\tilde{V}/k_B T}\right]$$

Scaling $$U \sim \frac{k_B T \alpha_1}{\mu D} \frac{\mu}{\mu + \frac{3}{2}\tilde{\mu}} a\tilde{\delta}$$

same as the "gas bubble"

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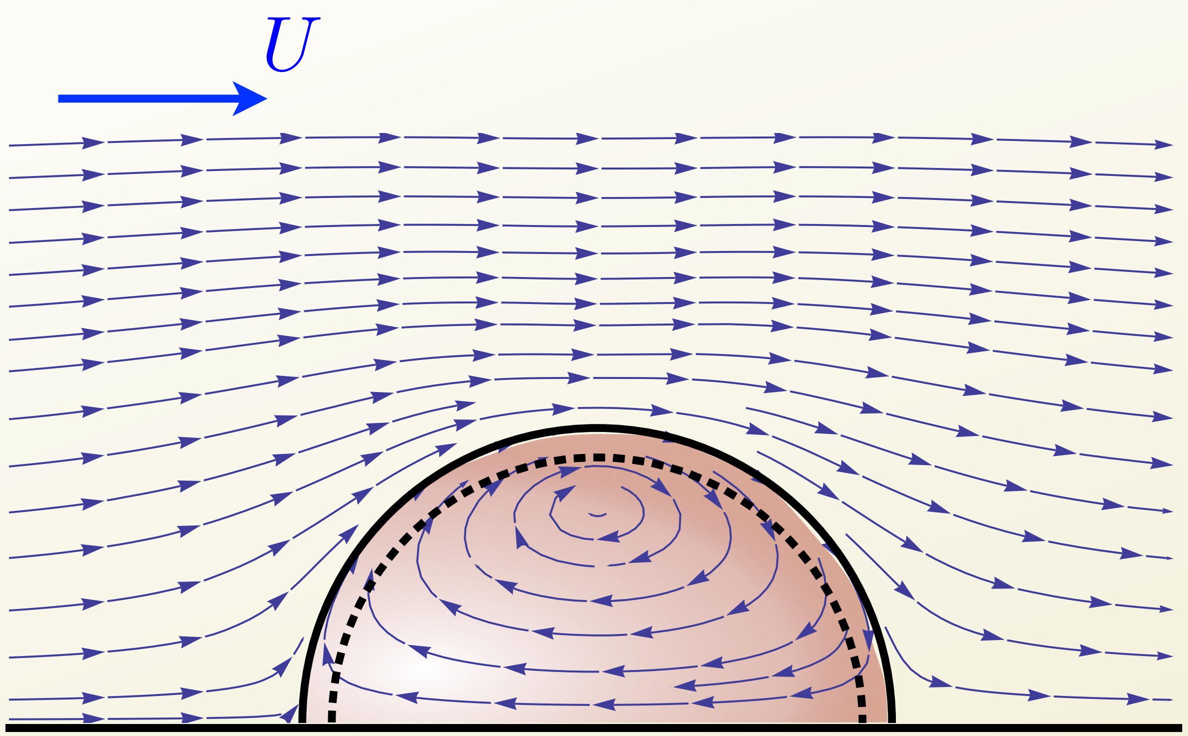
Surface tension gradients



Rest frame of the particle

- Activity due to a surface active catalyst
- Surface adsorbed species lower the surface tension
- Chemical reaction near surface produces local heating; lowers surface tension
- Non-uniform surface tension drives Marangoni flows

Stress balance at the interface

$$(\sigma_{ab} - \tilde{\sigma}_{ab})\, n_b - \gamma(2H)\, n_a + \nabla_a \gamma = 0$$

external fluid stress

internal fluid stress

surface normal

surface tension

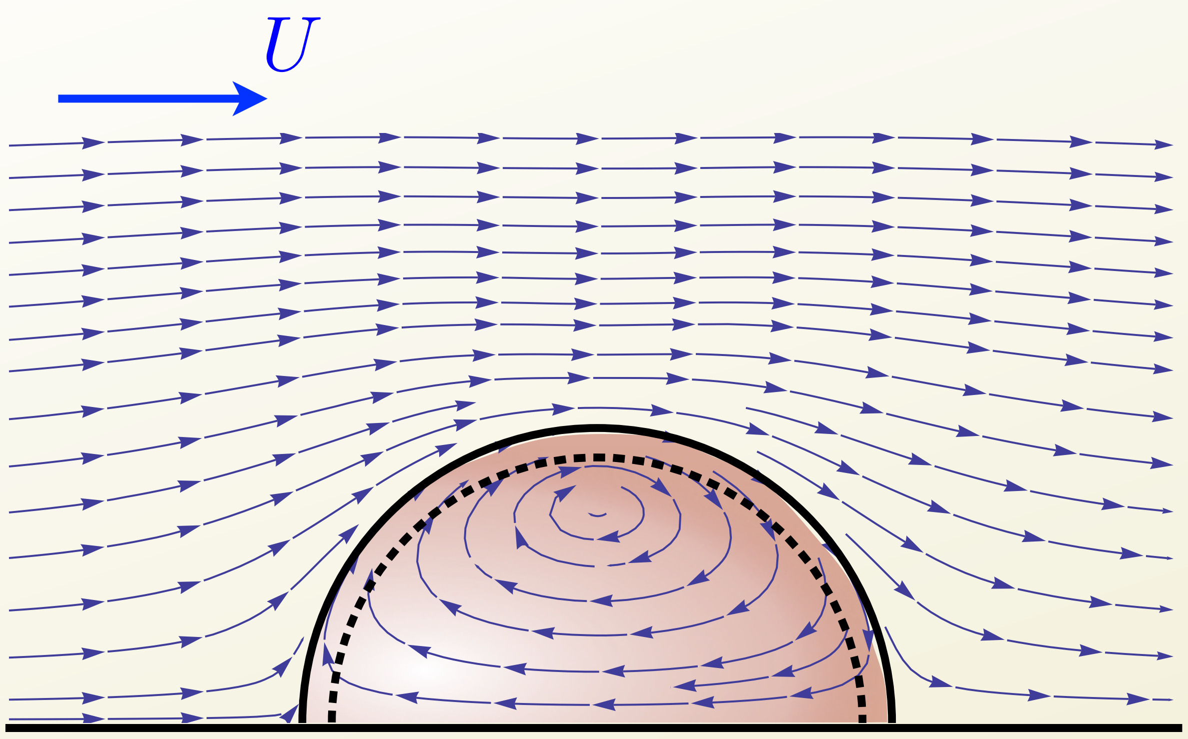
mean curvature

Marangoni stress

Penn

Pearson *J. Fluid Mech.* **4**, 489–500 (1958)
Sternling, Scriven *AIChE J.* **5**, 514–523 (1959)
Levich, Krylov *Ann. Rev. Fluid Mech.* **1**, 293–316 (1969)

Surface tension gradients



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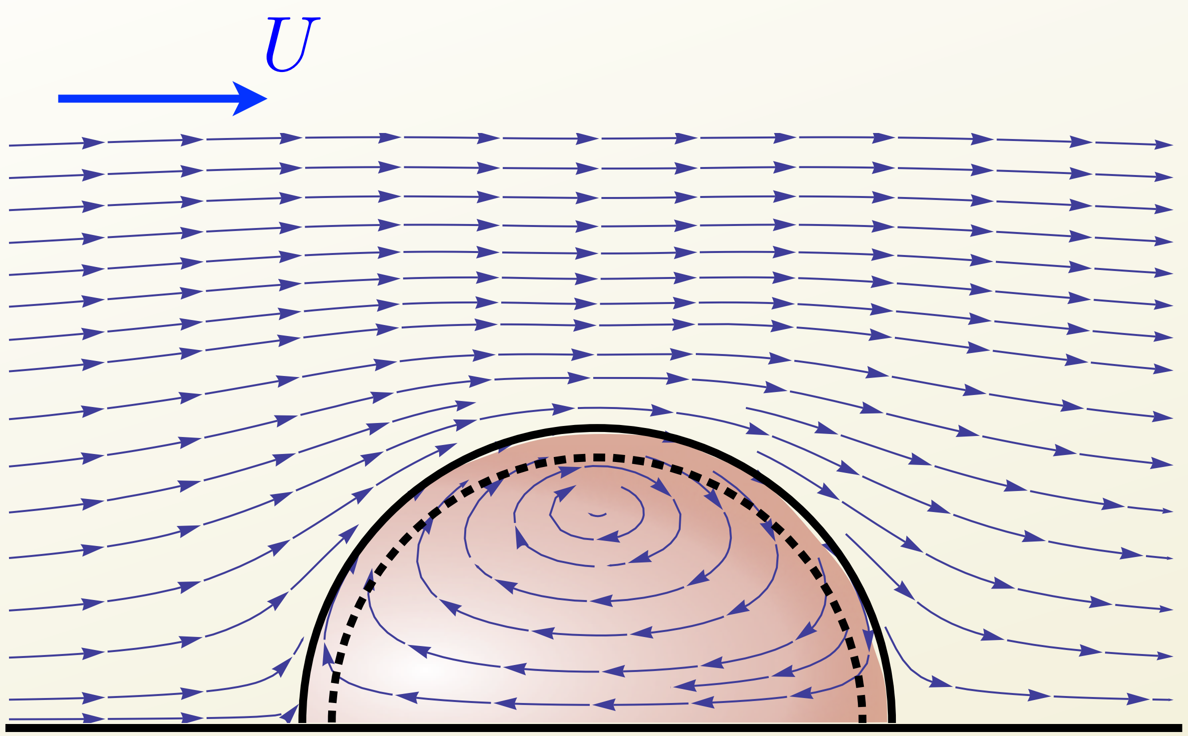
Additional contribution to the speed $$\frac{\mu+\tilde{\mu}}{3\mu\left(\mu+\frac{3}{2}\tilde{\mu}\right)}\gamma_1$$

typically surface tension is **lowered** so that γ_1 is **negative**
Marangoni flows then **oppose** self-diffusiophoresis

Surface tension gradients



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ratio $$\frac{\text{Marangoni}}{\text{diffusiophoresis}} \sim \frac{\mu+\tilde{\mu}}{\mu}\frac{D\gamma_1}{k_B T\alpha_1 a\delta}$$

can this be made small?

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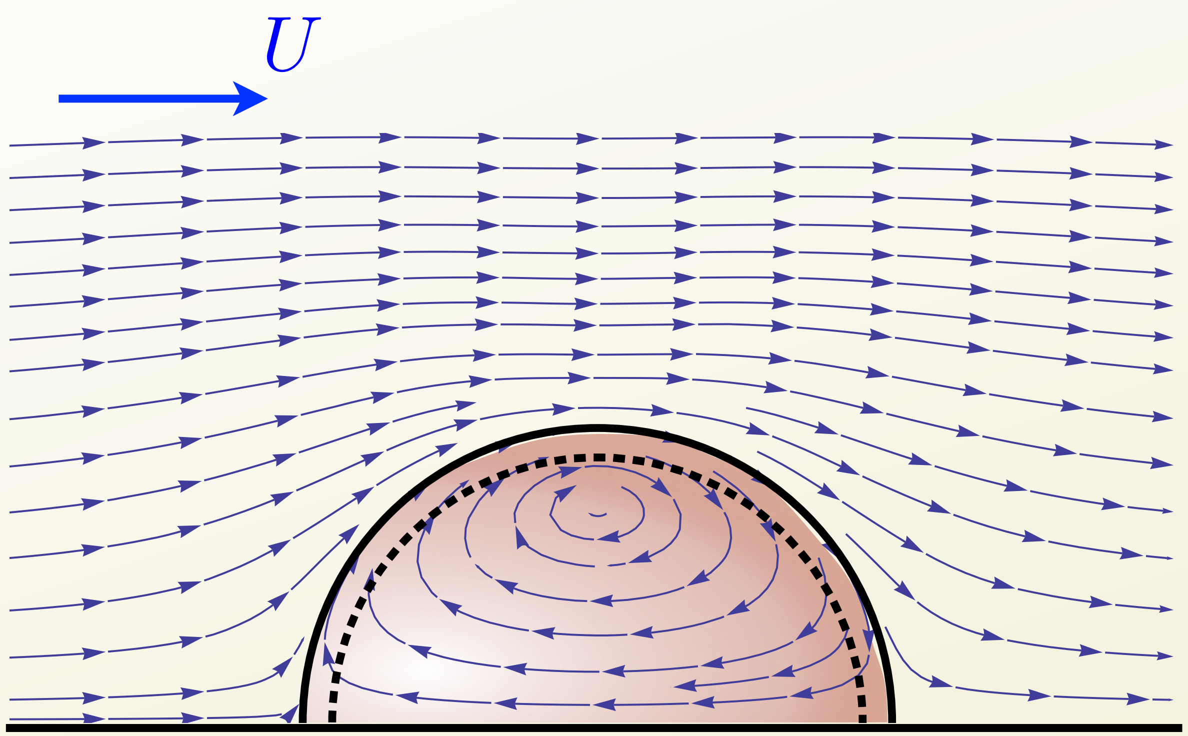
Droplet deformation



Rest frame of the particle

- Particle is a fluid droplet -- no reason why it won't deform
- Normal stress balance is really an equation for the drop shape

droplet remains approximately spherical provided $\dfrac{\mu U}{\gamma} \ll 1$

Stress balance at the interface

$$\left(\sigma_{ab} - \tilde{\sigma}_{ab}\right) n_b - \gamma(2H)\, n_a + \nabla_a \gamma = 0$$

external fluid stress

internal fluid stress

surface normal

surface tension

mean curvature

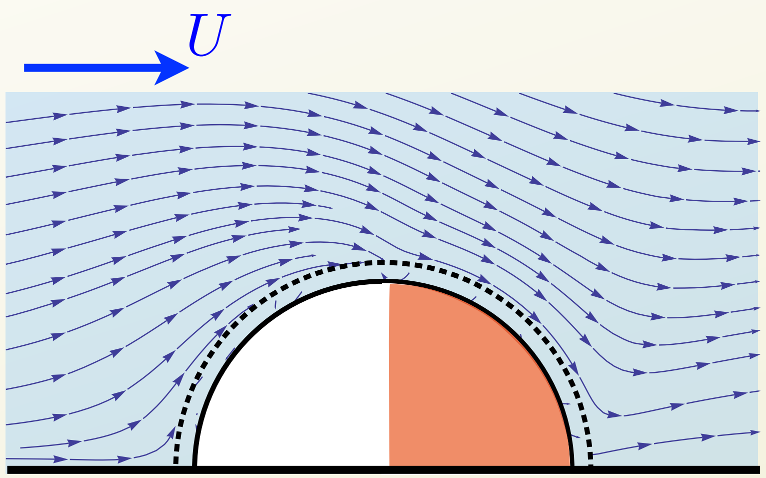
Marangoni stress

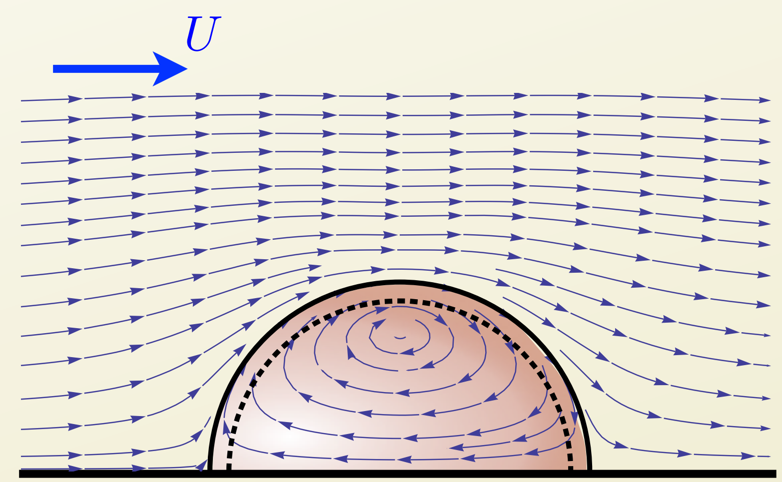
Penn

Taylor *Proc. Roy. Soc. Lond. A* **146**, 501–523 (1934)
Taylor, Acrivos *J. Fluid Mech.* **18**, 466–476 (1964)
Stone, Leal *J. Fluid Mech.* **220**, 161–186 (1990)

Thanks!

Andrea Liu, Timon Idema





- High Péclet number relevant to biological motility
- Different scaling with activity, dependence on coverage and successful strategies

- Fluid drops can move due to internal motor
- "Clean" system
- Faster than a solid particle

Acknowledgements

We are grateful to Ed Banigan and Kun-Chun Lee for beneficial discussions and to Randy Kamien for his support and encouragement

Funding

NSF DMR05-47230
UPENN-MRSEC DMR05-20020