COLLECTIVE RESPONSE AND EMERGENT STRUCTURES IN MICRO (NANO) SWIMMER SUSPENSIONS

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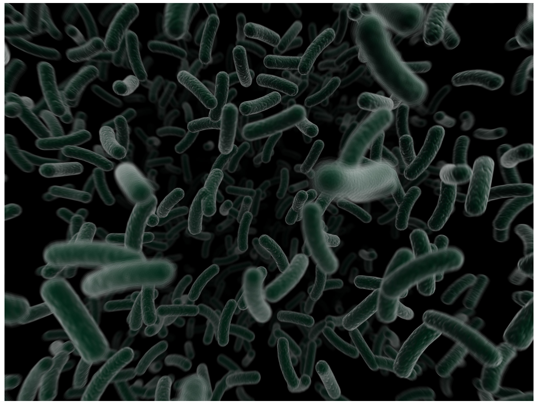
1. Introduction

Active systems: collections of elements able to convert internal energy into mechanical work (autonomous motion)

→ intrinsically out-of-equilibrium systems (even in a steady state, if any, and without external forcing)

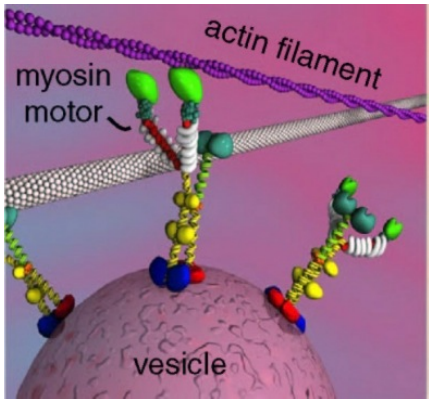
Examples

bacterial colonies

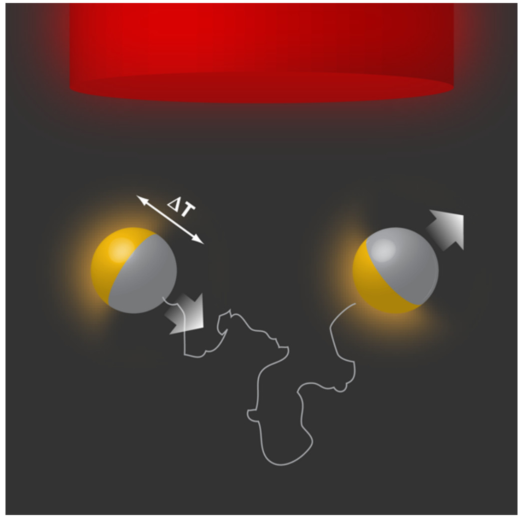


flocks of birds

molecular motors

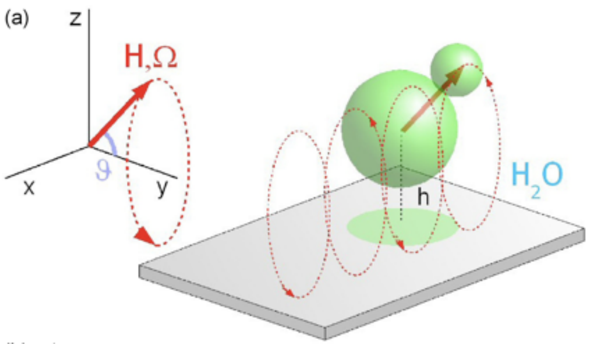


artificial self-propelled objects

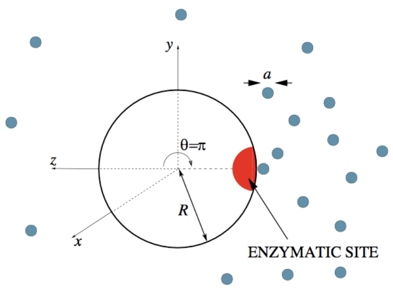


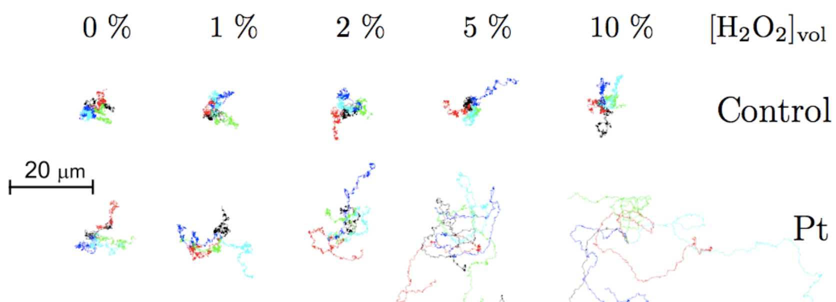
1. Introduction

Actuated colloids



Heterogeneous particles

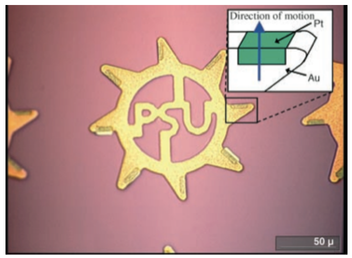


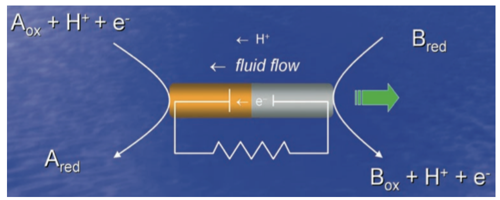


Adding reactivity:

new propelling mechanisms

Different sets of micro/nano robots





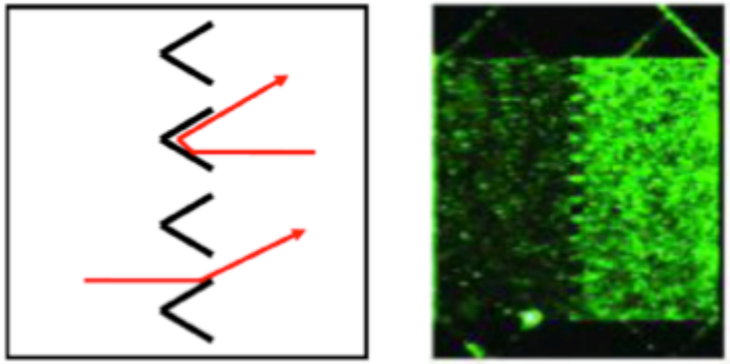
Confinement + asymmetric mobility

no deformation

Desired structures, adaptive, capable of self repair

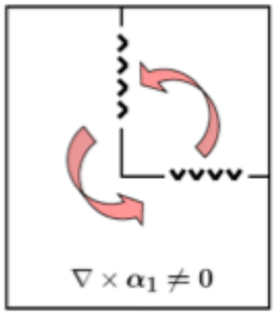
1. Introduction

Energy from small scales
Systems intrinsically out of equilibrium

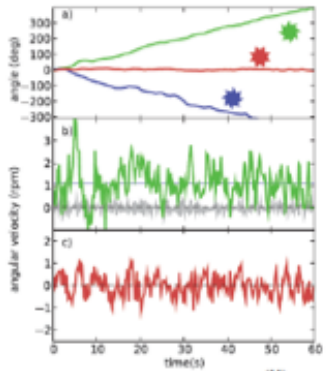


Galajda et al. J Bacter. (2007)

Non-equilibrium distributions
no detailed balance

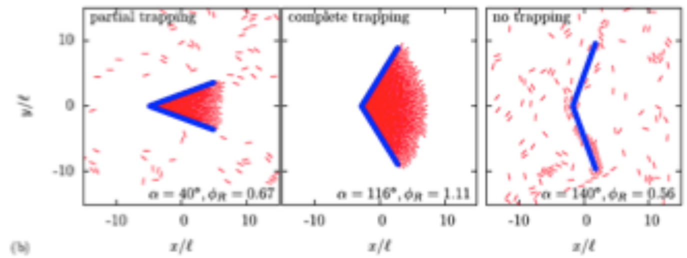


(i)



(ii)

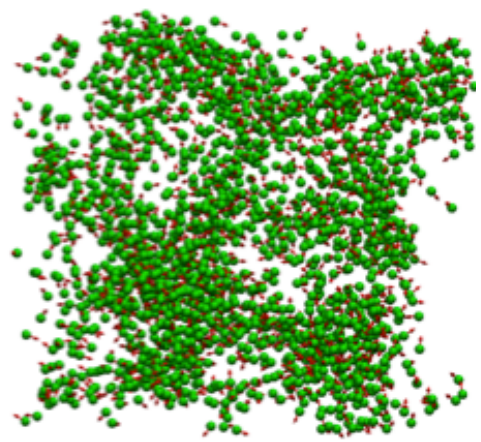
Ratchets



Kaiser et al. PRL (2012)

Emerging patterns
and phases

Di Leonardo et al. PNAS (2010)

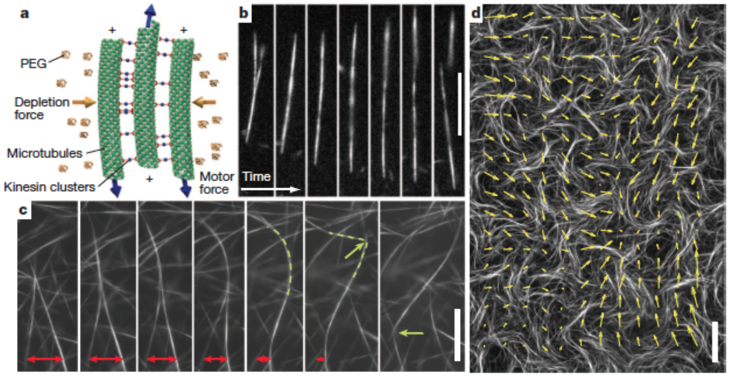


1. Introduction

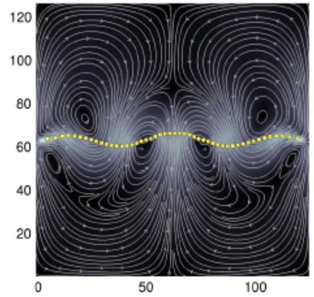
Jaramayan et al. PRL (2012)

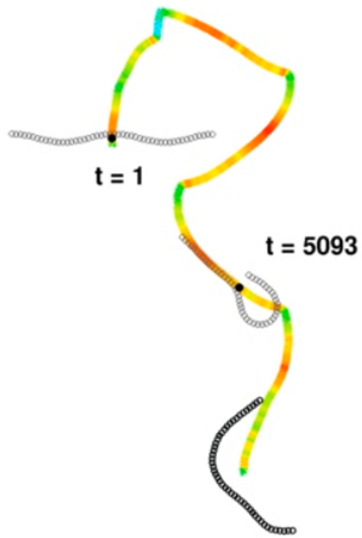
Internal activity
new materials

Biomimetic cilia
flagella

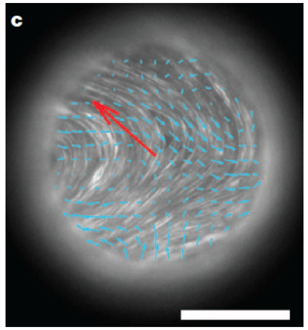


Sanchezet al. Nature (2013)





Active drops

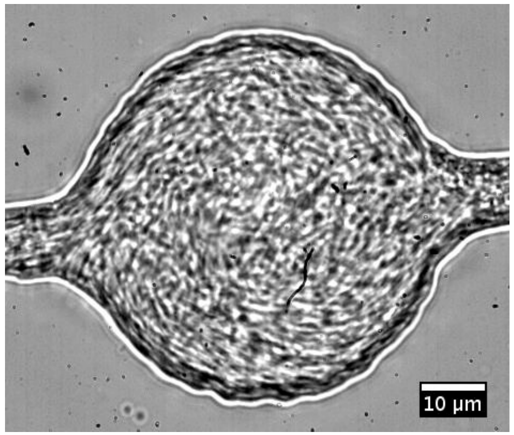


Left: Active Droplets

Right: Passive Droplets

10X Magnification

100μm bar



Microfluidic flows

Wioland et al. PRL (2013)

Desired structures, adaptive, capable of self repair

1. Introduction

Understanding motion at mesoscale

How does collective transport emerges in systems which
move due to internal consumption of energy?

Generic features due to
dynamic coupling to the environment?

Basic physical mechanisms
(over)simplified geometries / models
neglect any specific coupling

2. Molecular motors

Molecular motors on biopolymers
central role in transport and information exchange in cells

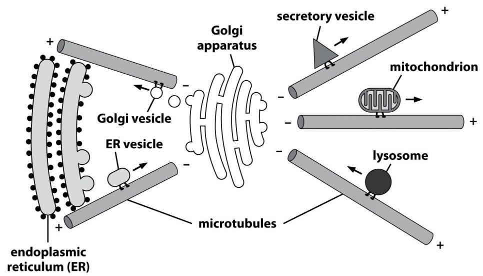


Figure 16.3 Physical Biology of the Cell (© Garland Science 2009)

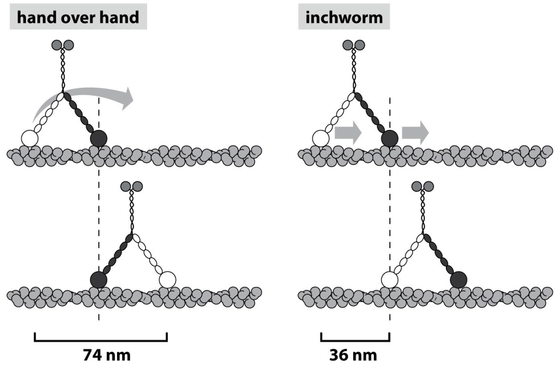


Figure 16.11a Physical Biology of the Cell (© Garland Science 2009)

Provide structure and activity to flagella

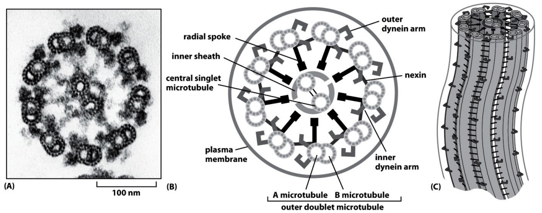


Figure 16.4 Physical Biology of the Cell (© Garland Science 2009)

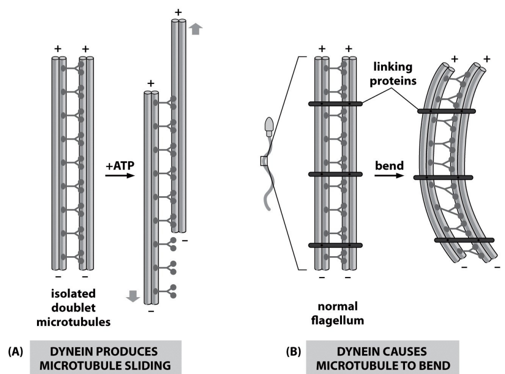


Figure 16.5 Physical Biology of the Cell (© Garland Science 2009)

How do they displace?
minimal step size
step and stop: large dispersion

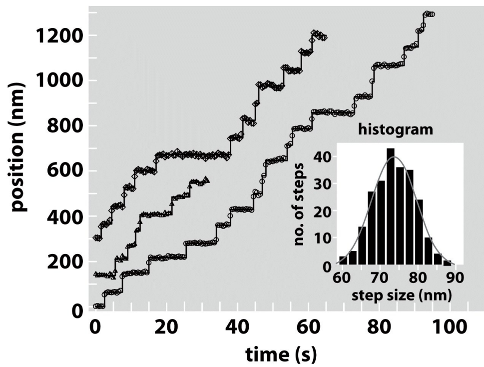
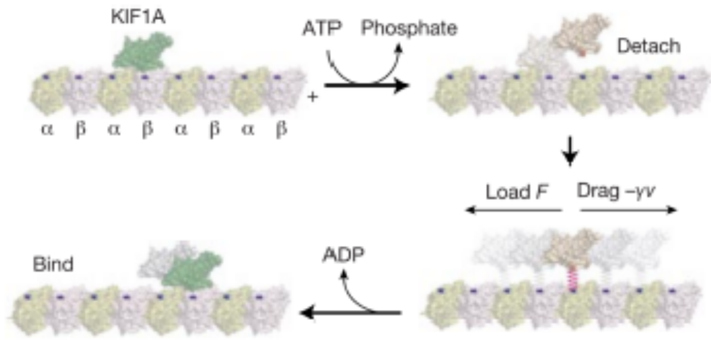


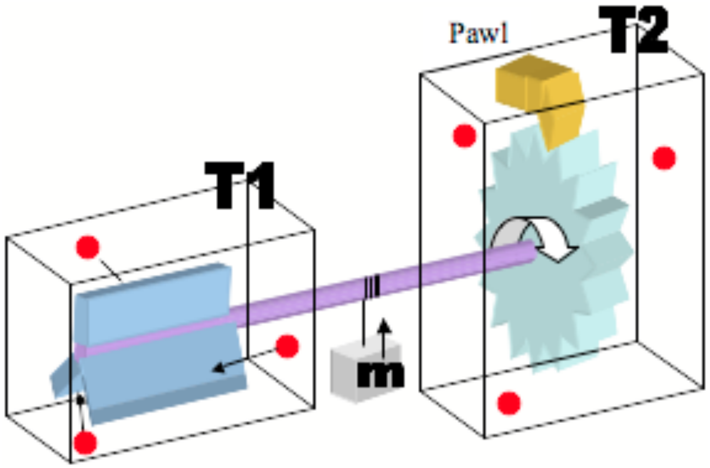
Figure 16.11b Physical Biology of the Cell (© Garland Science 2009)

2. Molecular motors

How do molecular motors move?
Brownian ratchet analogy

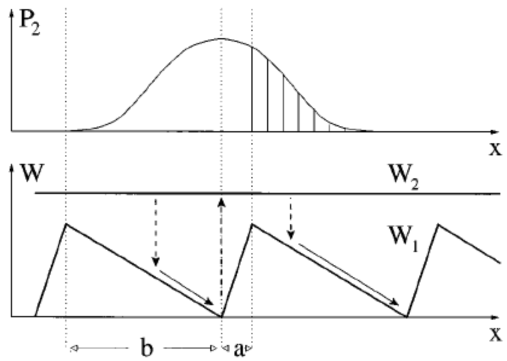


Asymmetry + no-detailed balance

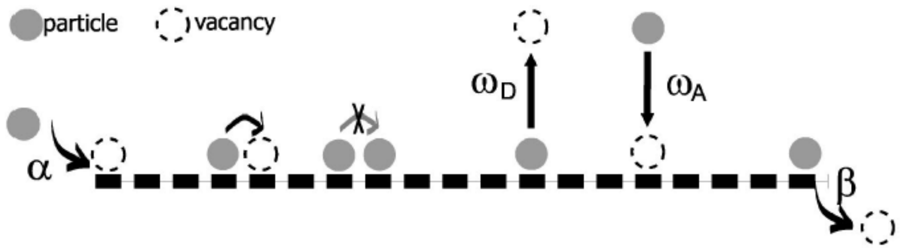


Smoluchowsky

Feynman

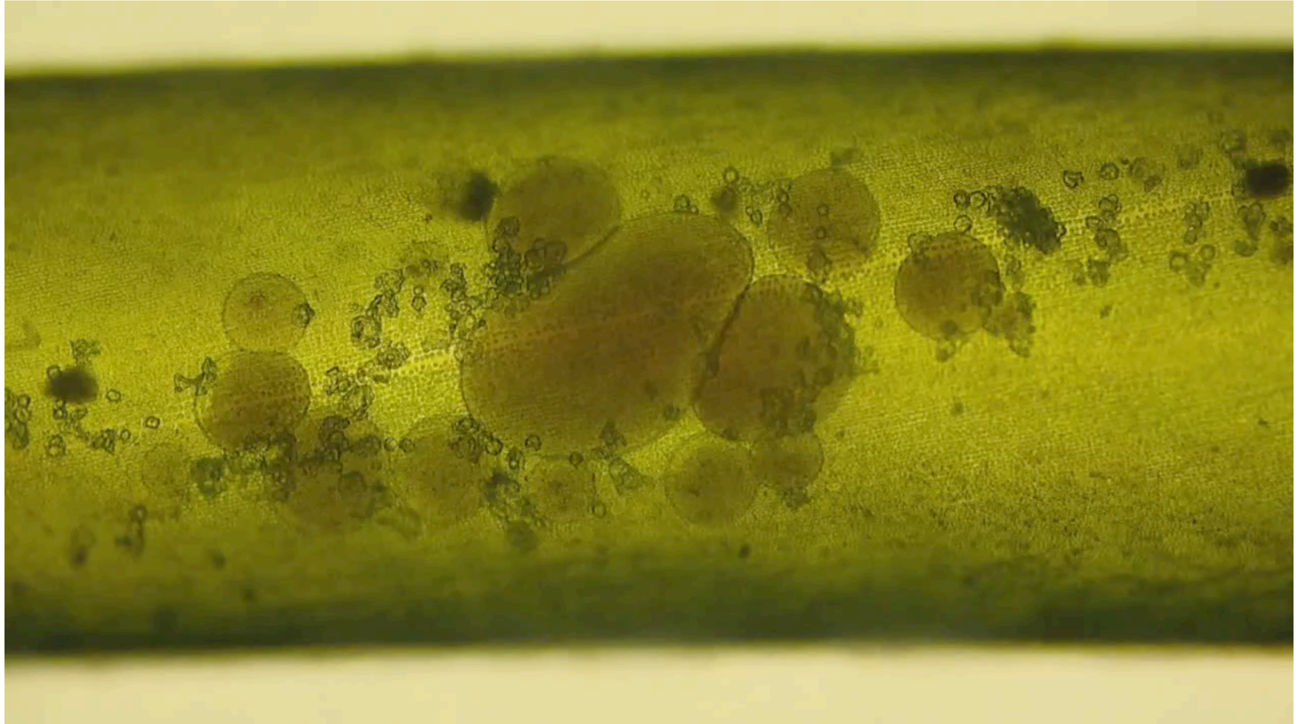


Asymmetry in jump rates
biases diffusion in less bound state



3. Collective transport

Inside cells
molecular motors
  cooperative transport
    role of embedding solvent?

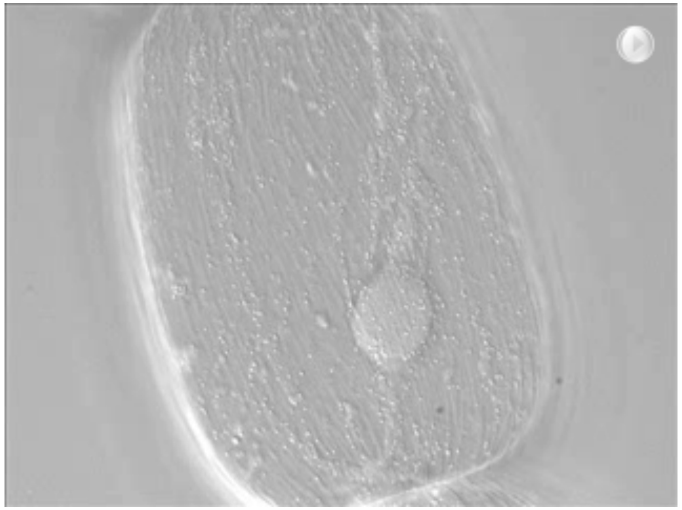


R. Goldstein

Relevance of hydrodynamic coupling?
  passive transport?

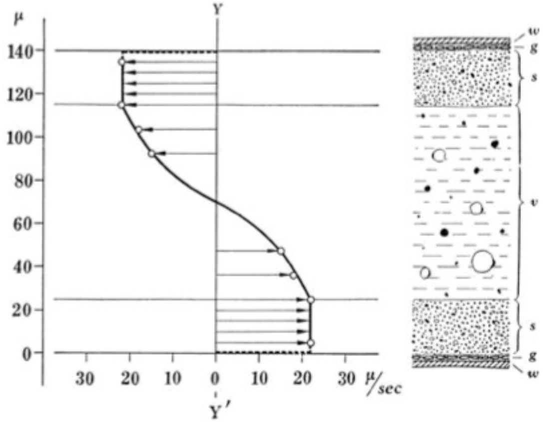
Forces small
  collective flow (restricted geometries)

Tradescantia virginiana



Cytoplasmatic streaming

Role actin/myosin

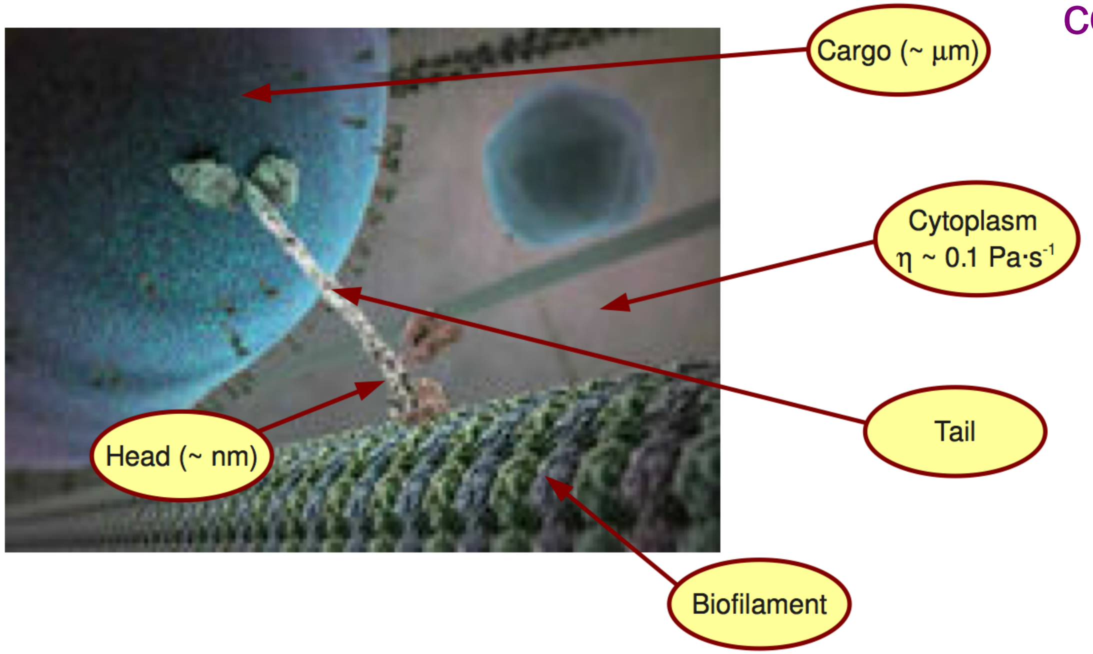


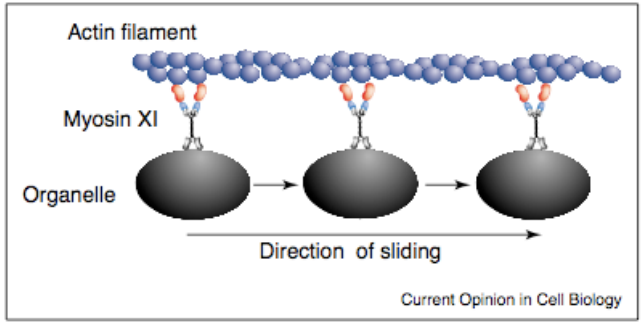
Shimmen 2007

3. Collective transport

Relevance of hydrodynamic coupling?
passive transport?

Forces small
collective flow (restricted geometries)





Cytoplasmatic streaming
induced by molecular motors?

3. Collective transport

Lowe-Andersen thermostat (LAT):
C.P.Lowe, Europhys. Lett. 47, 145, (1999).

$$\vec{r}_i(t+\Delta t) = \vec{r}_i(t) + \Delta t\vec{v}_i(t) + \frac{1}{2}\Delta t^2 f_i^C(t)$$

$$\vec{v}_i(t+\Delta t) = \vec{v}_i \qquad \Gamma\Delta t < \xi$$

$$\vec{v}_j(t+\Delta t) = \vec{v}_j \qquad \Gamma\Delta t < \xi$$

"Bath" collision:

$$\vec{v}_i(t+\Delta t) = \vec{v}_i + \frac{\mu_{ij}}{m_i}\left(\theta_{ij}\sqrt{\frac{kT}{\mu_{ij}}} - (\vec{v}_i - \vec{v}_j)\cdot\hat{r}_{ij}\right)\hat{r}_{ij} \qquad \Gamma\Delta t \geq \xi$$

$$\vec{v}_j(t+\Delta t) = \vec{v}_j - \frac{\mu_{ij}}{m_j}\left(\theta_{ij}\sqrt{\frac{kT}{\mu_{ij}}} - (\vec{v}_i - \vec{v}_j)\cdot\hat{r}_{ij}\right)\hat{r}_{ij} \qquad \Gamma\Delta t \geq \xi$$

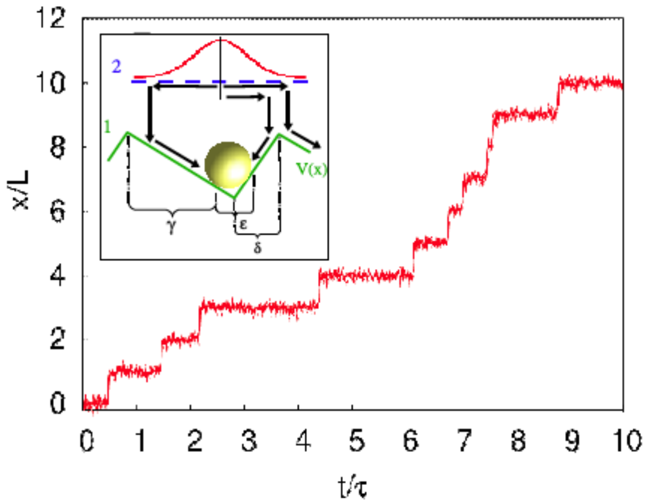
Here Γ is a bath collision frequency (plays a similar role to γ/m in DPD)

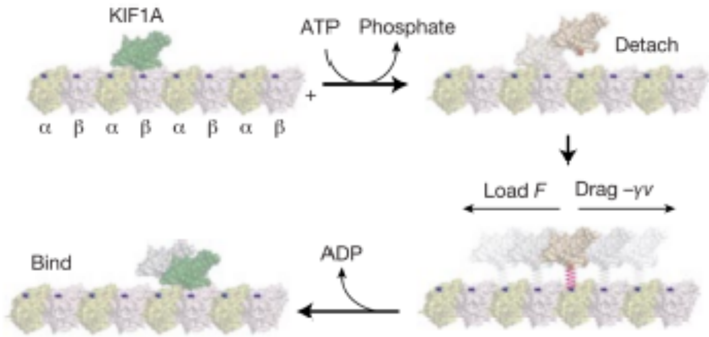
- Bath collisions are processed for all pairs with $r_{ij}<r_c$
- The current value of the velocity is always used in the bath collision (hence the lack of an explicit time on the R.H.S.)
- The quantity ξ is a random number uniformly distributed in the range 0-1
- Reduced mass for particles *i* and *j*, $\mu_{ij}=m_i m_j/(m_i+m_j)$

3. Collective transport

Motors step rather than slide

Need for a detailed description of motor displacement along a biofilaments?

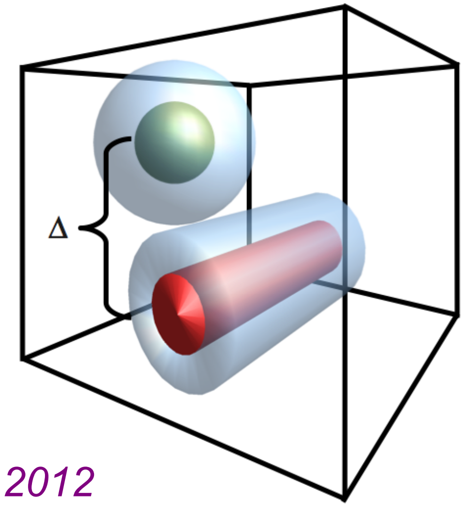




Ratchet model

Combined motion +
coupling to solvent

Simplified geometry



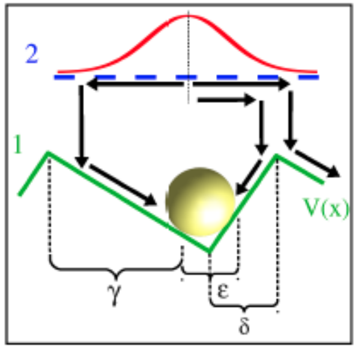
Malgaretti et al. PRL 2012

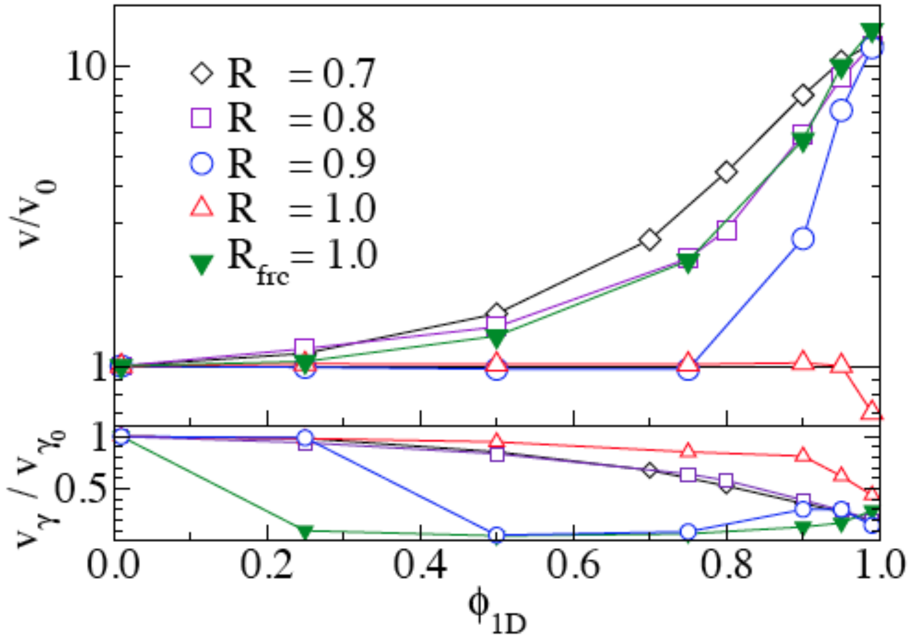
3. Collective transport

Correlated trajectories due to pull/push

Excluded volume

substrate periodicity

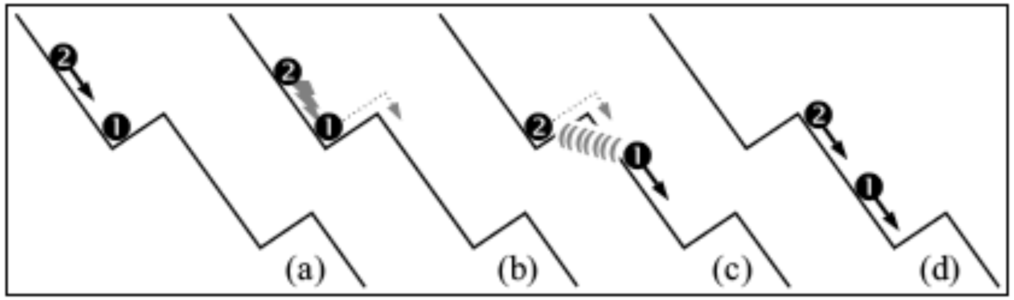




Relevance of forcing during free diffusion

Determine velocity enhacement

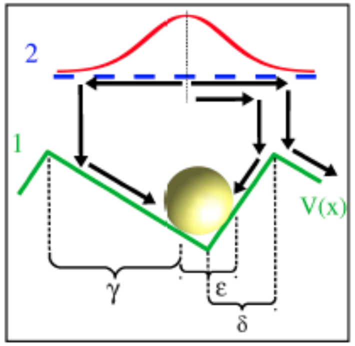
Commensurate motors do not see each other



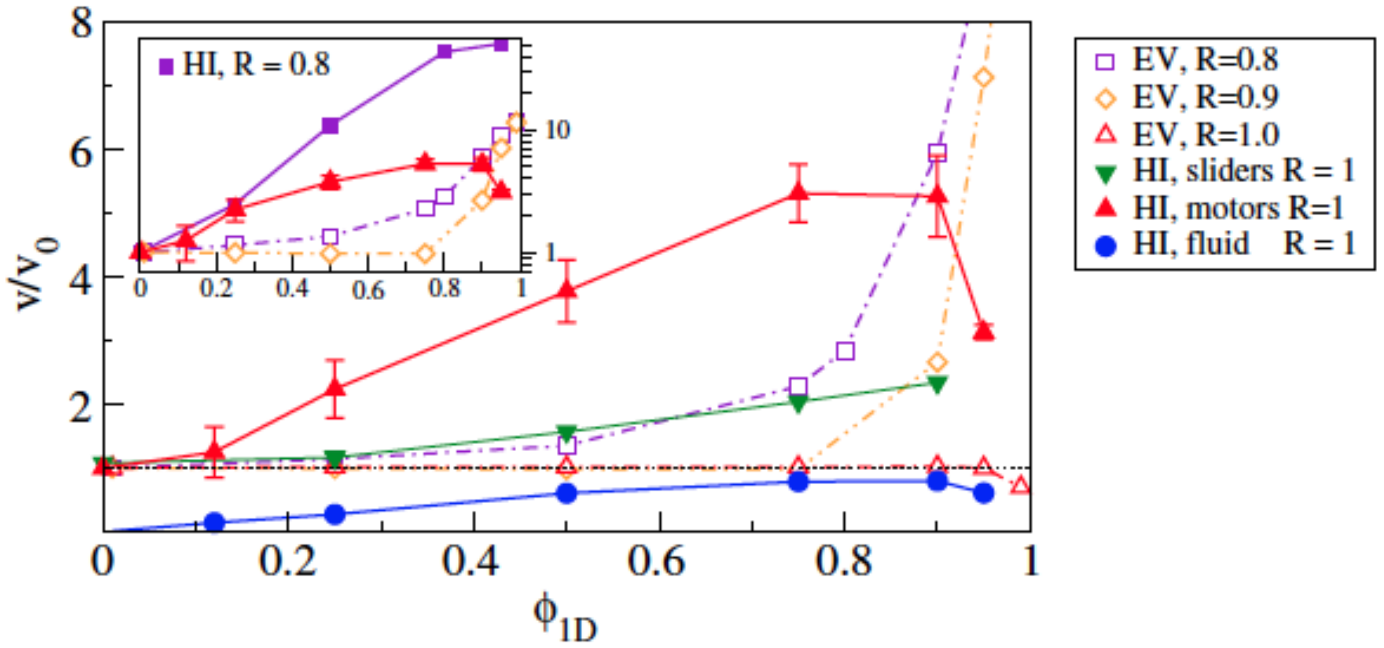
Forced colloids

Lutz et al 06

3. Collective transport



Minimize excluded volume interactions



Correlated trajectories due to
hydrodynamic pull/push

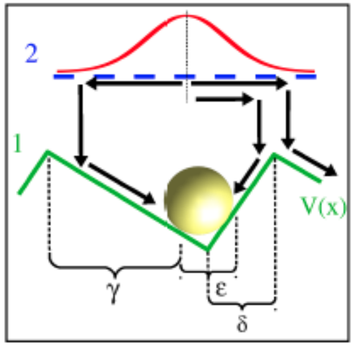
Bias due to induced flow during diffusion

Long-range hydrodynamic coupling subdominant

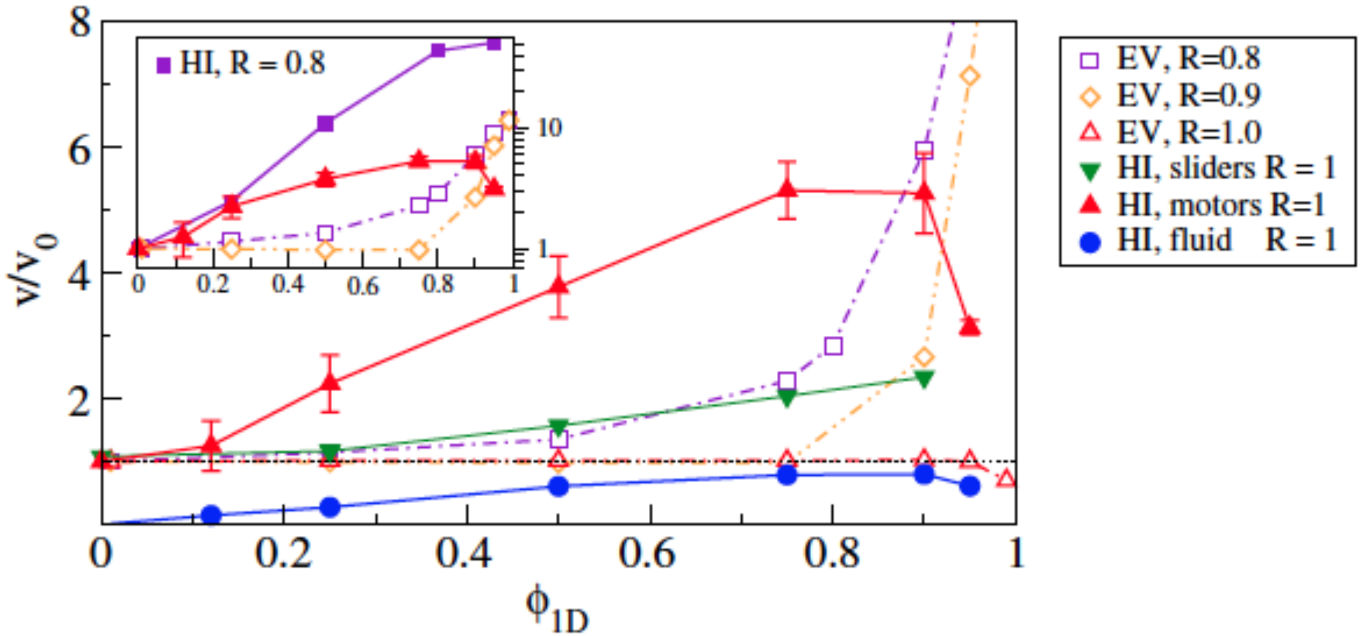
Forces small
motion due to collective flow

Hydrodynamics enhances transport
suspended particles

3. Collective transport



Minimize excluded volume interactions



$$v \simeq 2\frac{f}{6\pi\eta a}\rho p_\downarrow \frac{l-\delta}{l}\int_{2R}^{L/2} dr\,\frac{3a}{2r}$$

$$\Delta t \simeq \frac{1}{2}\frac{6\pi\eta a}{f(l-\delta)}$$

When does coupling induce significant displacement

$$v\Delta t \geq \delta$$

$$\bar{\phi}_{1D} \equiv 2\delta/(l-d)^2 \ln L/4a$$

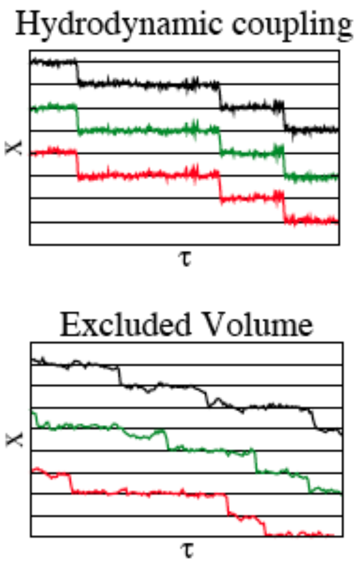
Hydrodynamic transient coupling

new mechanism for activated motion?

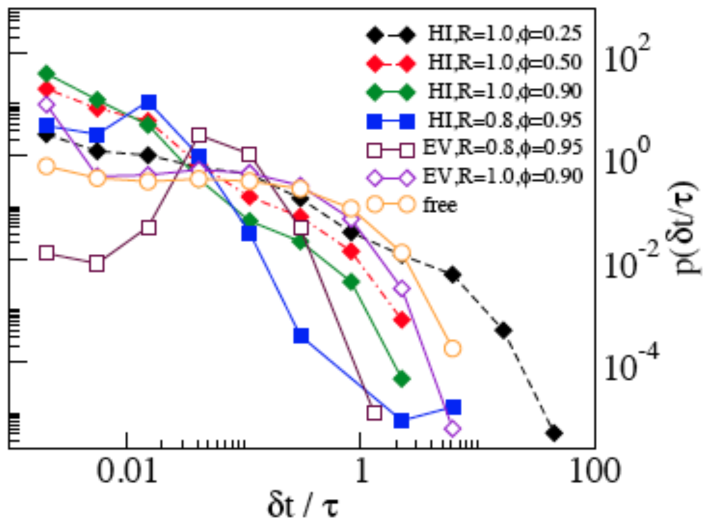
less sensitive to confinement

3. Collective transport

Correlated trajectories due to
hydrodynamic pull/push

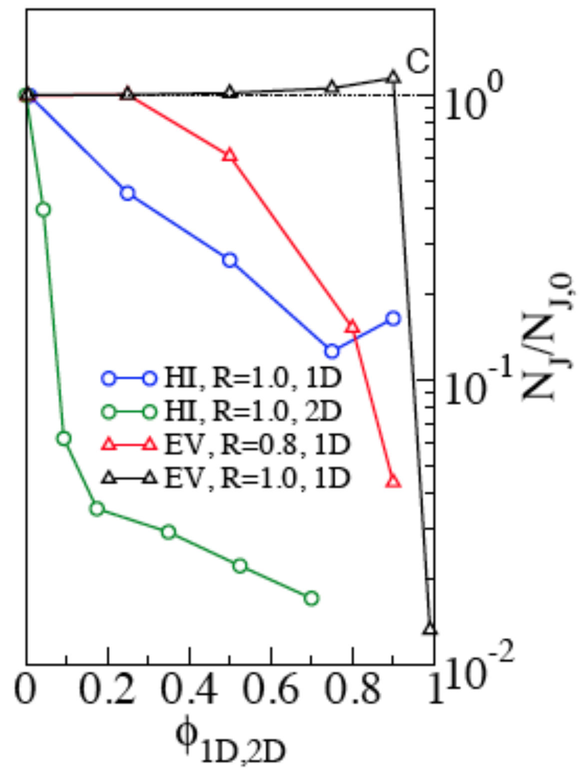


Dwelling times consecutive motors

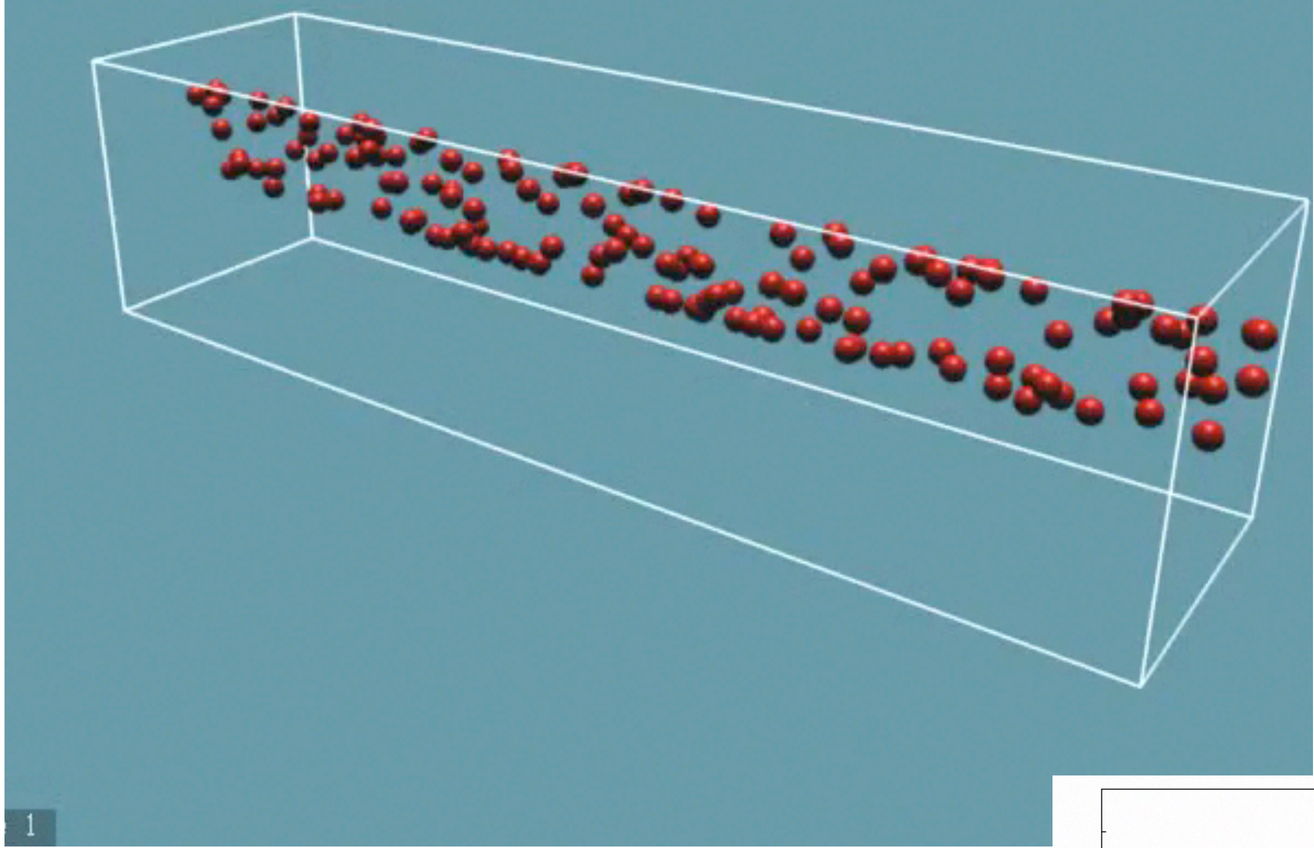


Correlated jumps

Decrease longer dwelling times

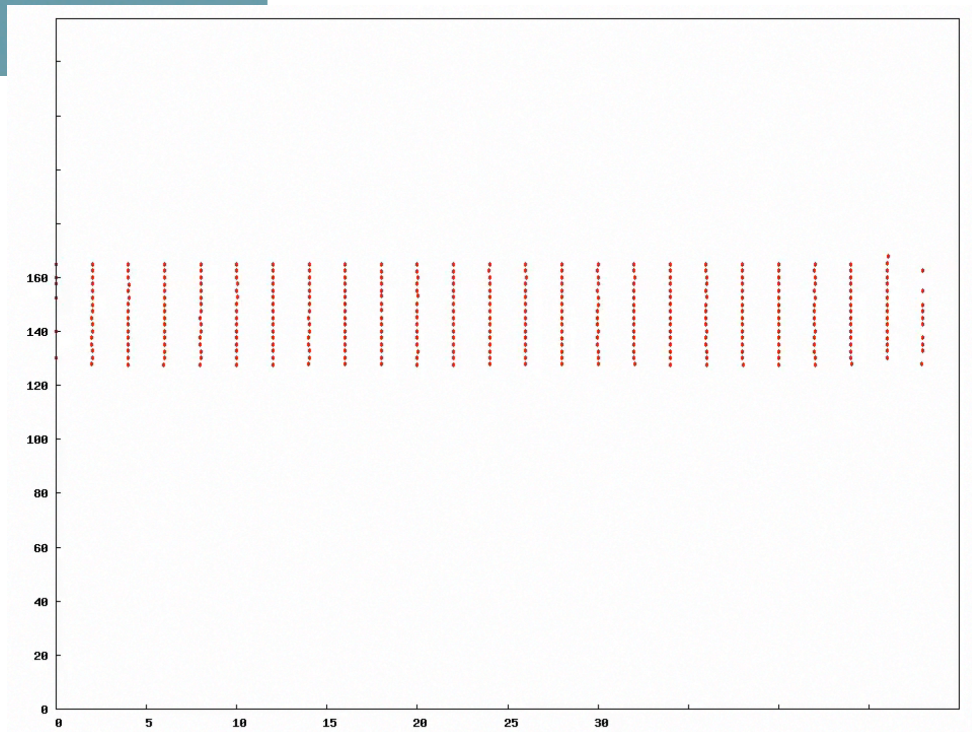


Hydrodynamic coupling
decrease in energy cost
energy used to move isolated motor
exploited by neighbours

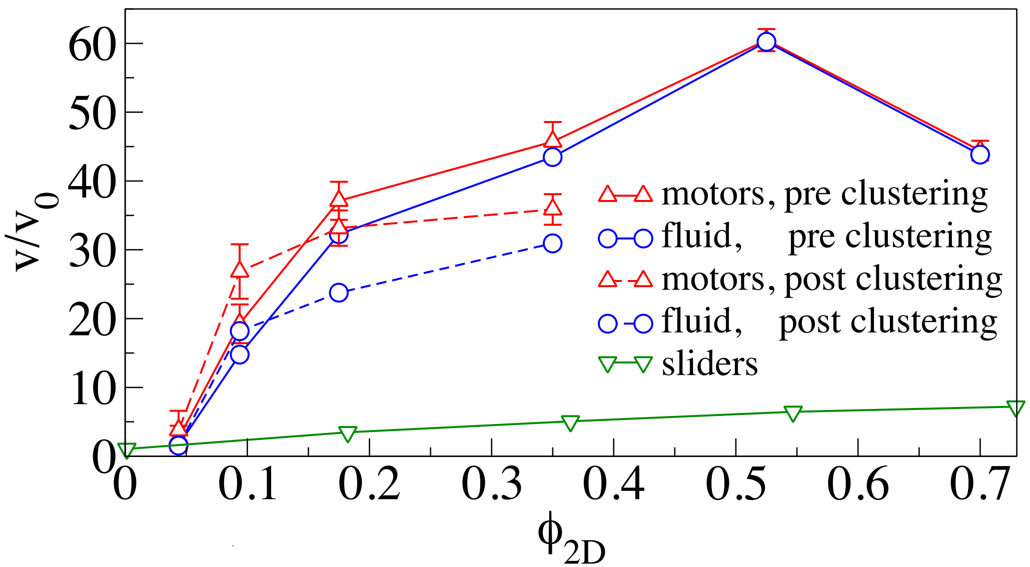


Cluster formation: ring stability?

Shock wave?

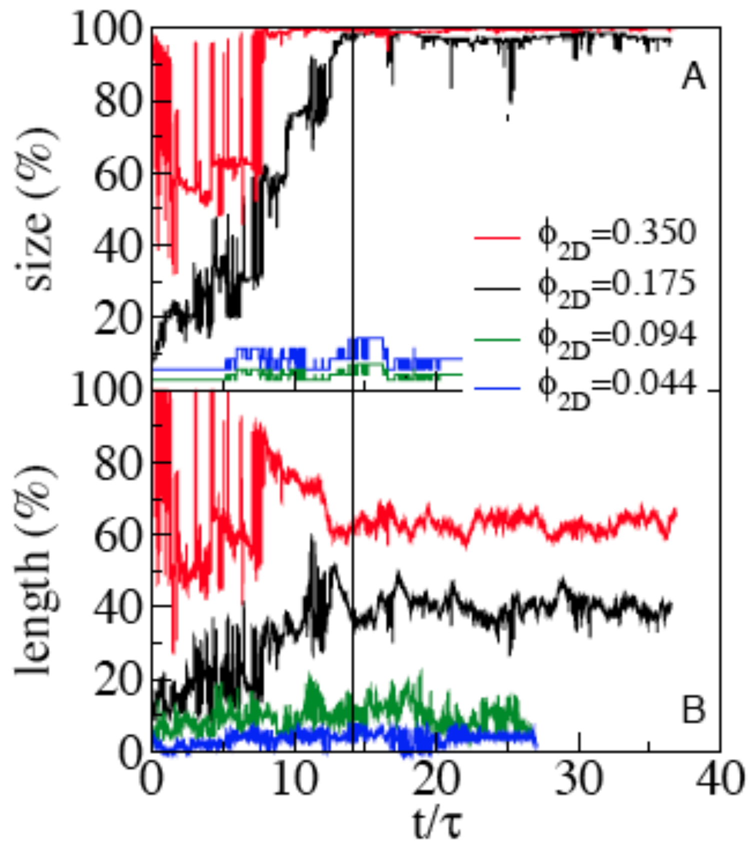


3. Collective transport



Cluster formation: ring stability?

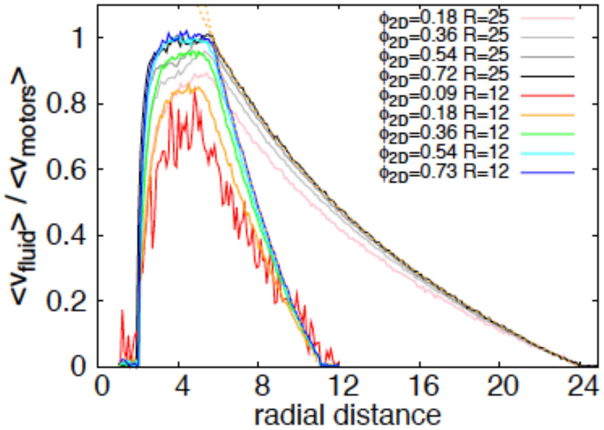
Shock wave?

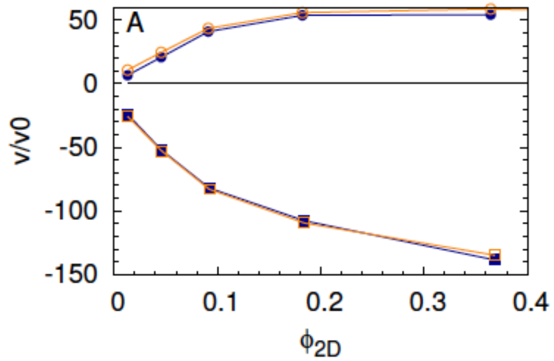


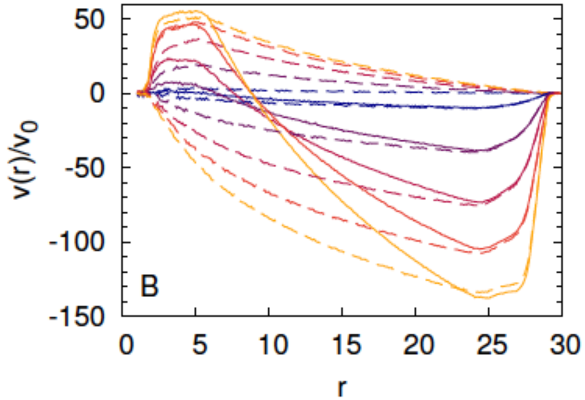
Fraction of motors in largest cluster

3. Collective transport

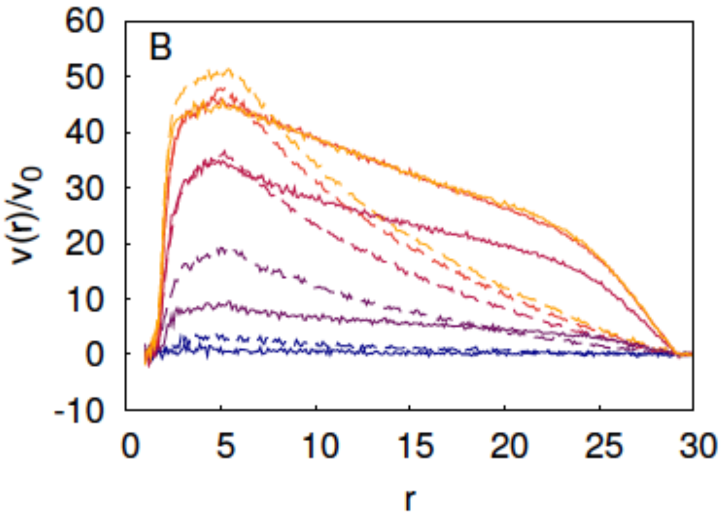
Molecular motors generate cytoplasmatic flow

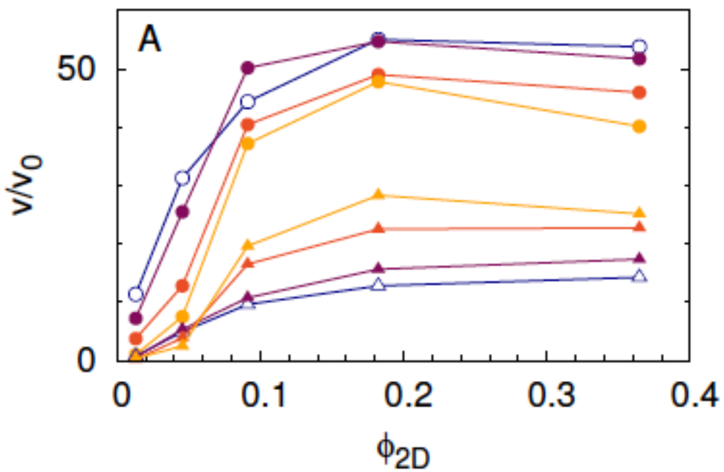






Drives suspended organelles for free

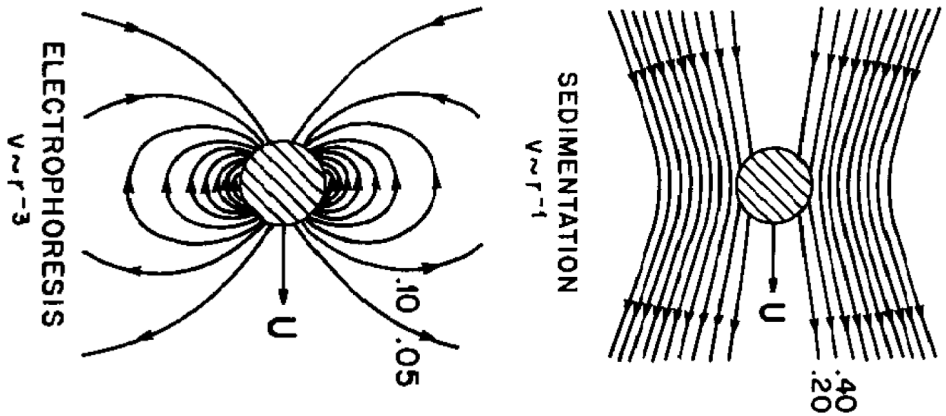




Interference / Coupling with motors

4. Microswimmers

Dynamics of active particles

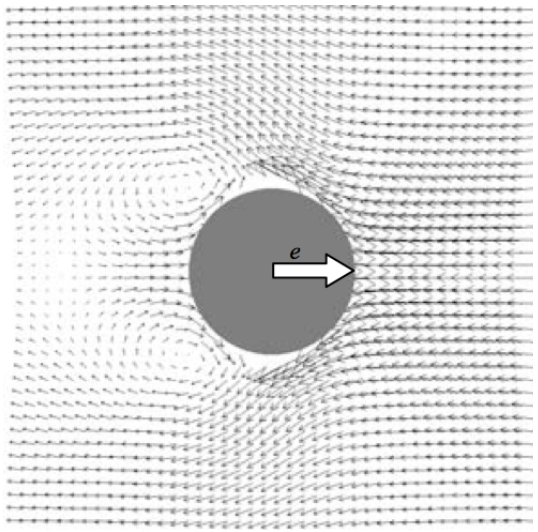


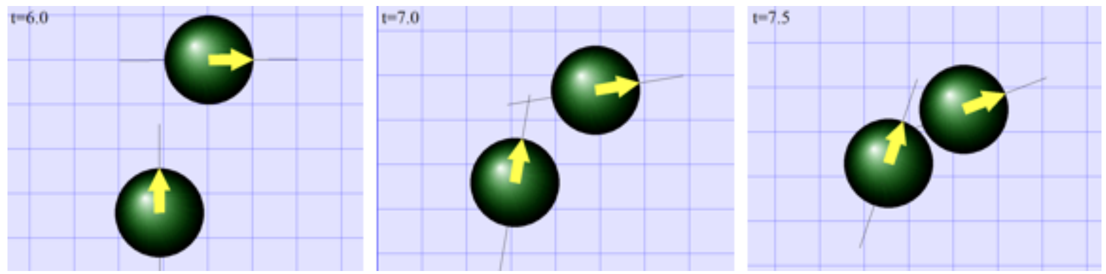
Low Reynolds numbers

Absence of external driving
closer to electrophoresis?

Relevance of swimming mechanism

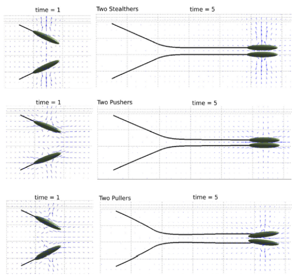
Fluid flows with vorticity





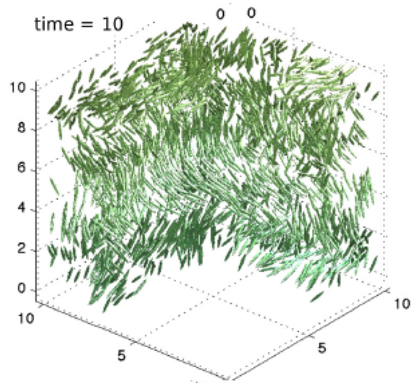
Coupling translation/rotation
relevance of near field interactions

4. Microswimmers



Hydrodynamic coupling - long range

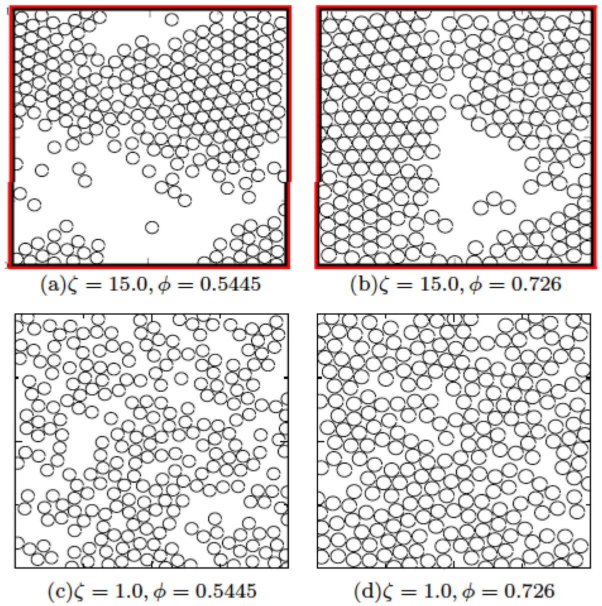
Relevance of shape



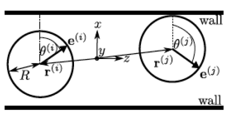
Lushi et al. (2013)

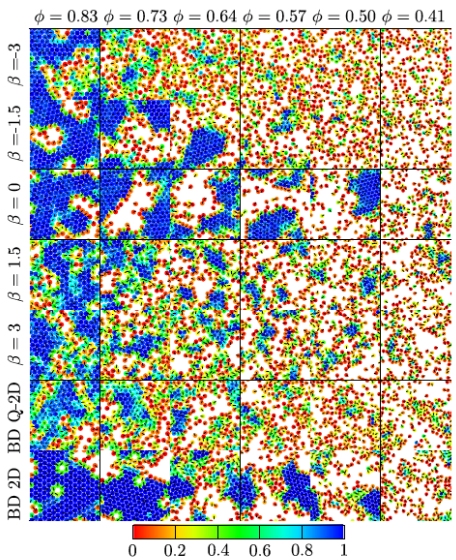
Disks/spheres

prevent crystallization?



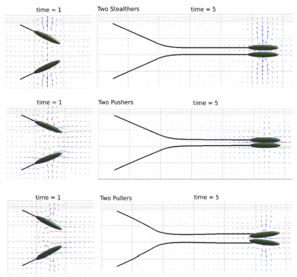
Matas-Navarro et al. (2014)





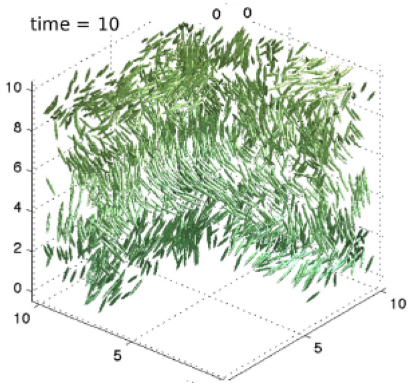
Zottl et al. (2014)

4. Microswimmers



Hydrodynamic coupling - long range

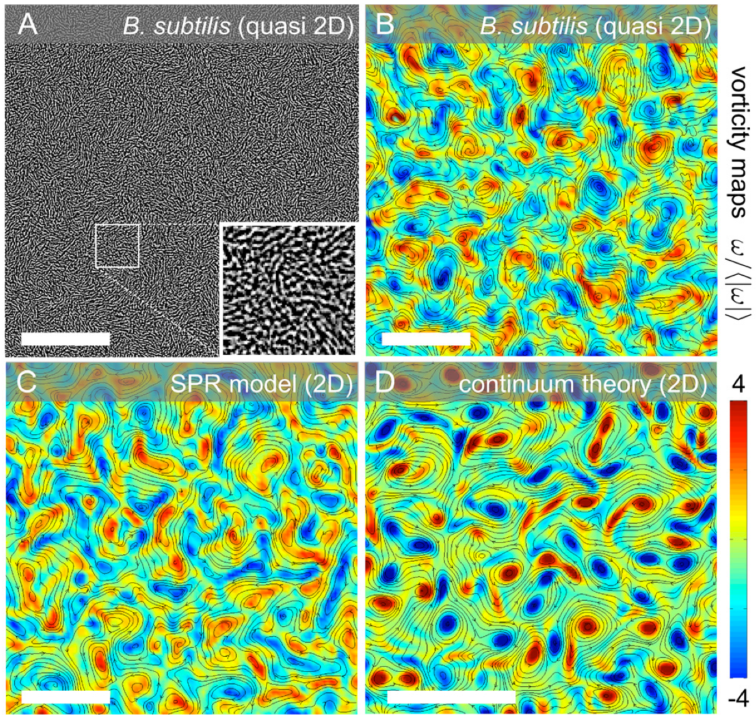
Relevance of shape

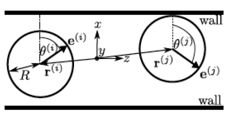


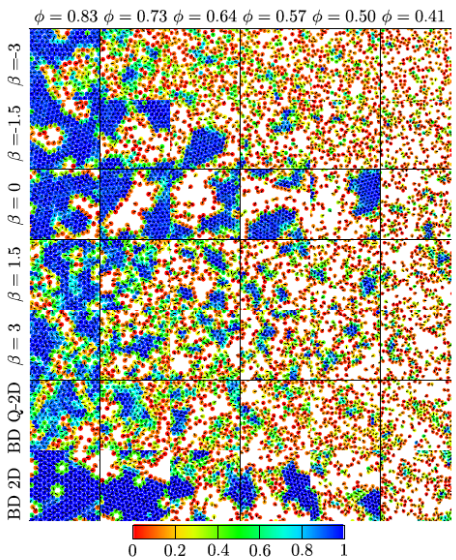
Lushi et al. (2013)

Unsteady flows

Meso-scale turbulence



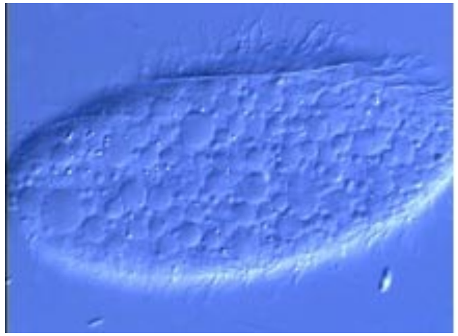


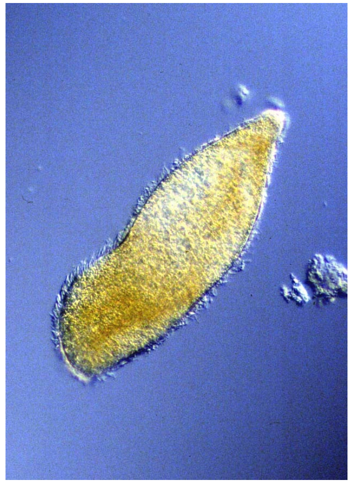


Zottl et al. (2014)

Wensick et al. (2012)

4. Microswimmers





Opalina

Squirmers

Metachronal wave on Opalina, Paramecium.

Fixed tangential velocity profile on the surface (Lighthill, 1952; Blake, 1971)

Surface tangential velocity

$$\mathbf{v}_S = \sum_{n=1}^{\infty} B_n V_n(\cos\theta)\mathbf{t}$$

$$\mathbf{v}_S = (B_1 \sin\theta + B_2 \sin\theta\cos\theta)\mathbf{t}$$

$\beta = B_2/B_1$

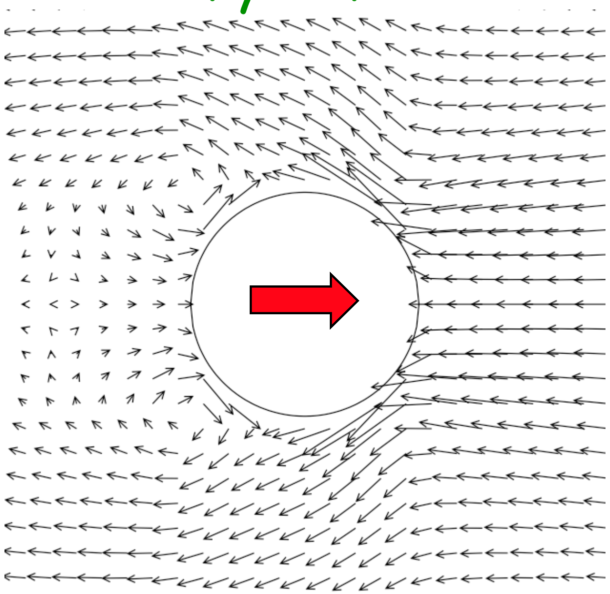
$$u_\infty = \frac{2}{3} B_1$$

Steady squirmer

(Pedley 1986)

Propulsion velocity

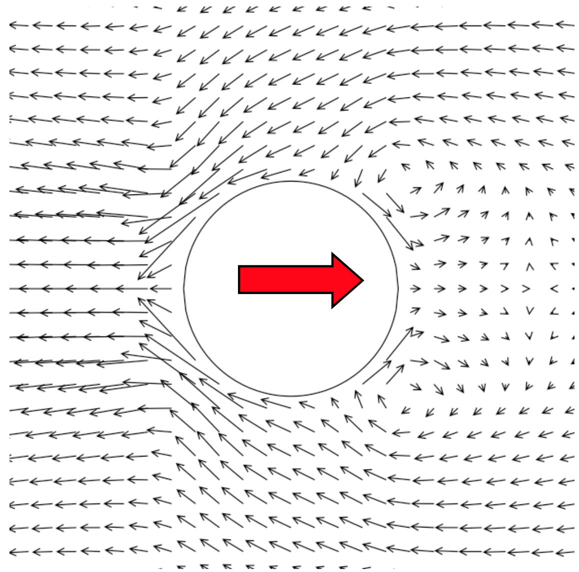
Chlamydomonas



$\beta>0$

puller

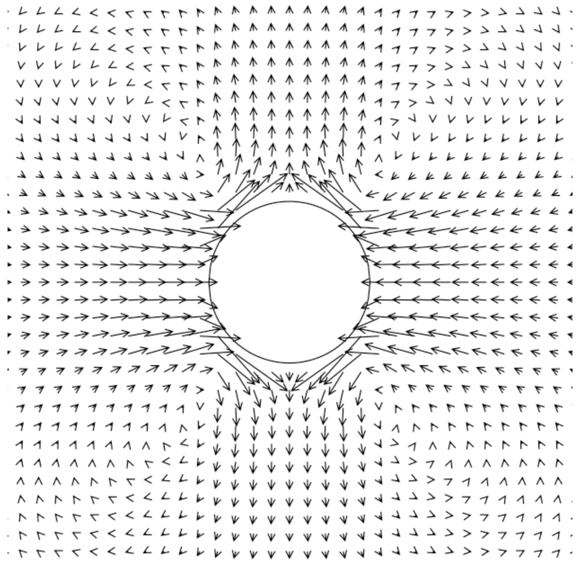
E. coli



$\beta<0$

pusher

$v \sim 1/r^2$



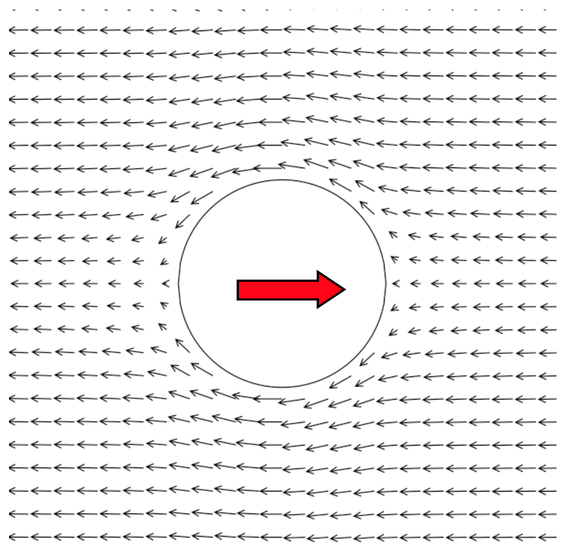
$B_1=0$

$B_2\neq 0$

Apolar

$\beta=0$

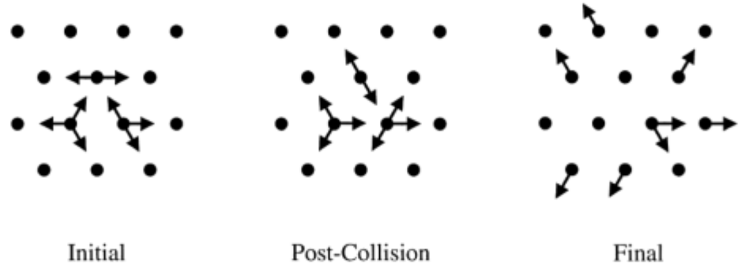
Passive squirmer



$v \sim 1/r^3$

4. Microswimmer suspension: Model

Lattice kinetic model: "microscopic" dynamics



$$f_i(r + c_i, t+1) = f_i(r,t) - \omega[f_i(r,t) - f_i^{eq}(r,t)]$$

$$\sum f_i = \rho$$

$$\sum f_i c_i = \rho v$$

$$\sum f_i c_i c_i = \rho vv + P$$

Conserved variables
Proper symmetries

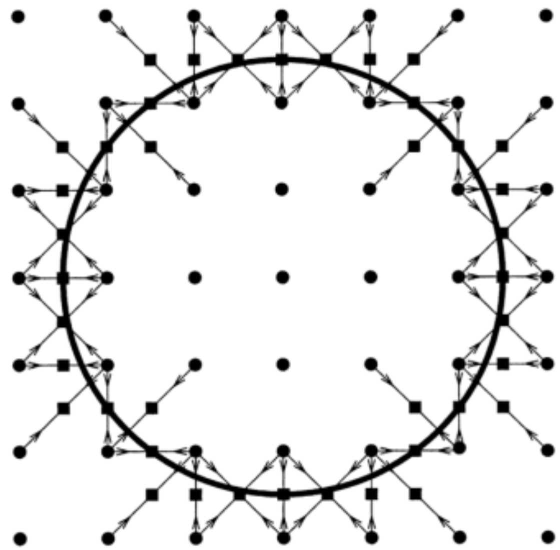
Hydrodynamic equations

Colloid

rigid hollow surface

collision

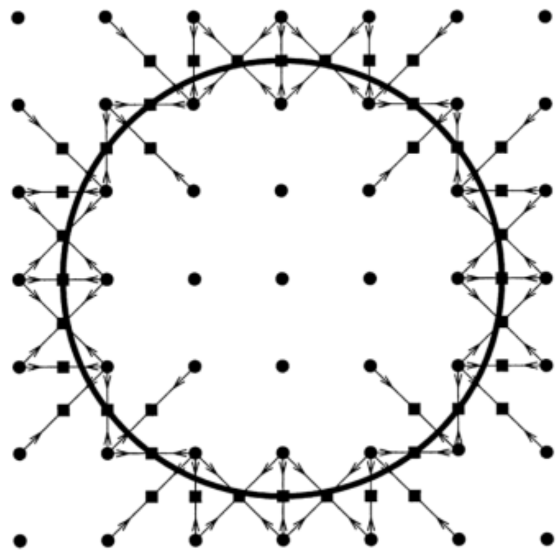
bounce-back



Hybrid scheme: molecular dynamics

Pre-selection of relevant degrees of freedom

4. Microswimmer suspension: Model



Hard core

No temperature

No tumbling

focus on hydrodynamic coupling

$$\mathbf{v}_S = (B_1 \sin\theta + B_2 \sin\theta \cos\theta)\mathbf{t}$$

Slip velocity as a local bounce-back

Additional attraction

competition with activity

Transition to an ordered phase:
LJ interaction strength is reduced and
B2 is not too big.

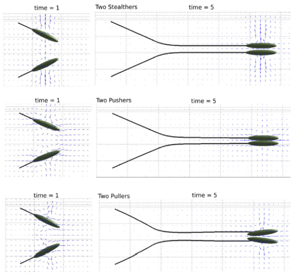
Stokes Law, small Reynolds

$$F_d = 6\pi\eta R_p v_s \qquad v_s = \frac{2}{3} B_1$$

$$\eta = 0.5,\ R_p = 2.3$$

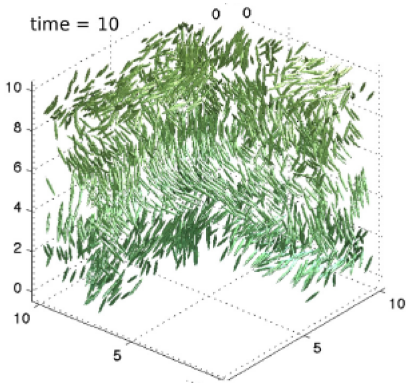
$$\xi = \frac{F_d}{F_{LJ}(r = \sigma_{LJ})}$$

4. Microswimmers



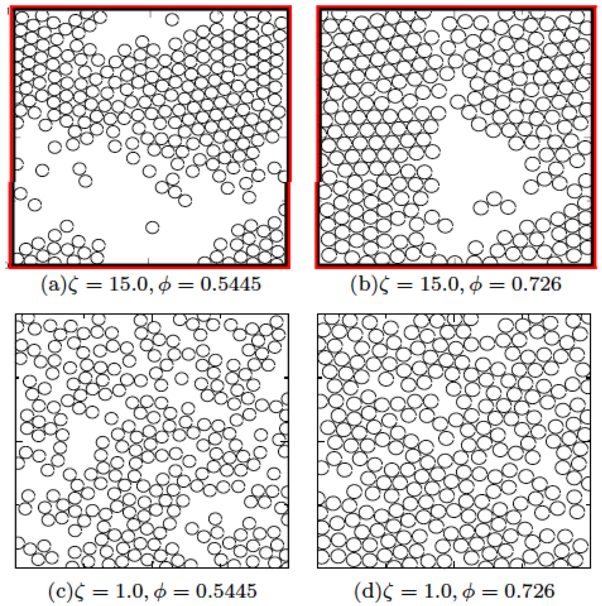
Hydrodynamic coupling - long range

Relevance of shape

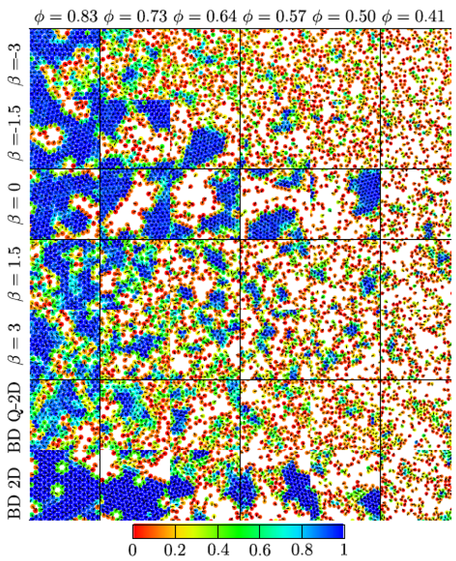


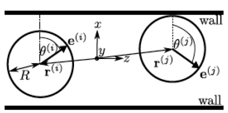
Lushi et al. (2013)

Disks/spheres
prevent crystallization?



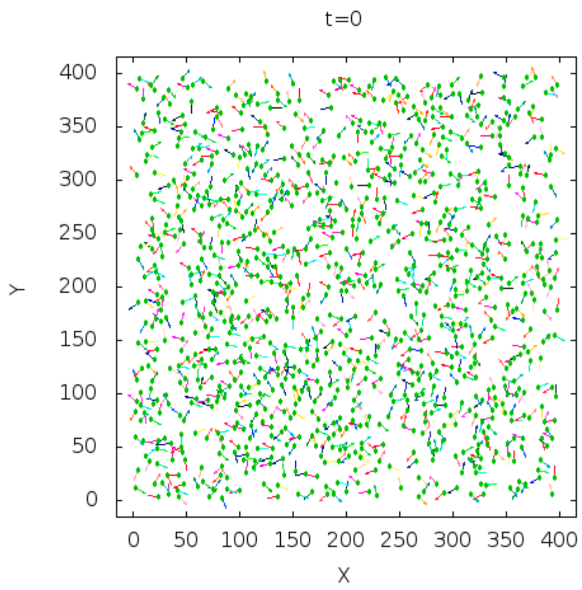
Matas-Navarro et al. (2014)

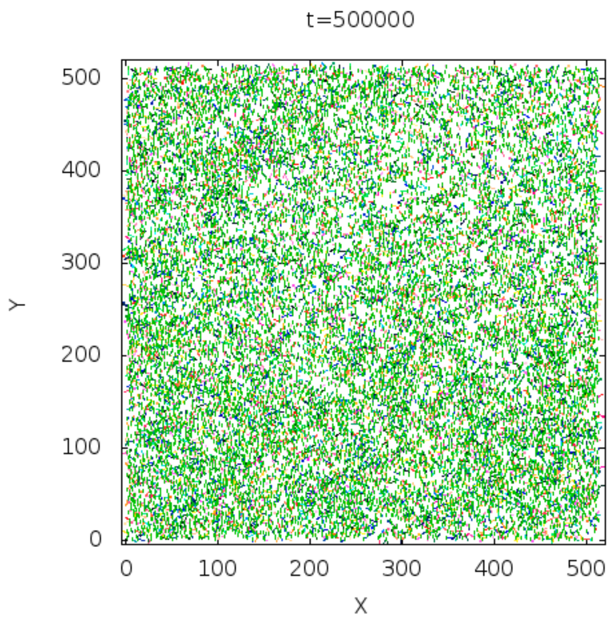




Zottl et al. (2014)

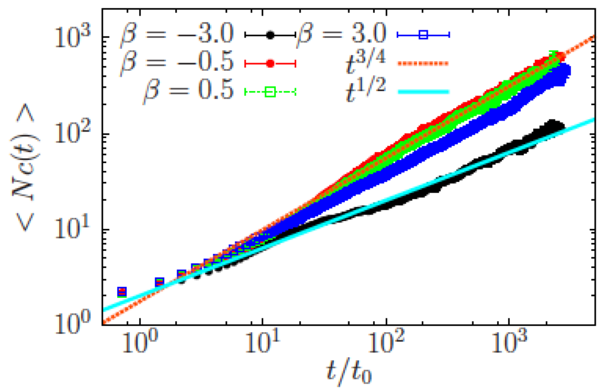
5. Cluster morphologies

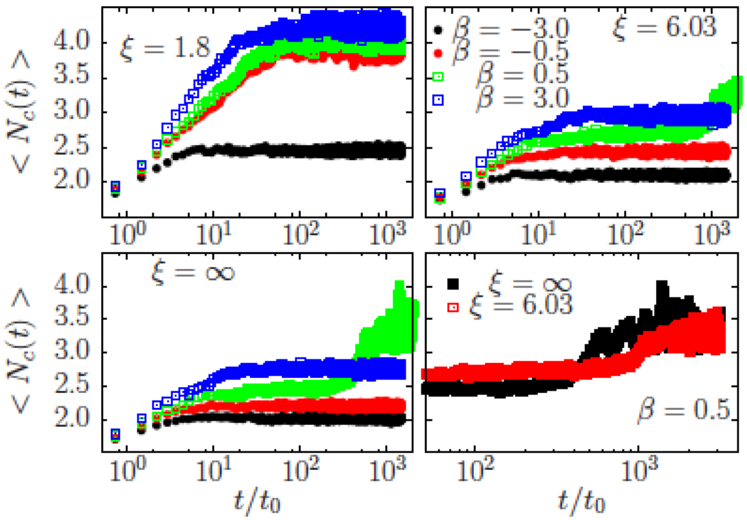




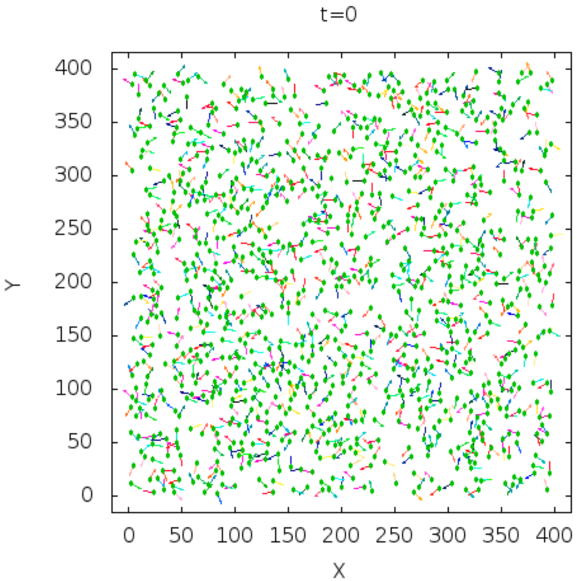
Dynamic structures

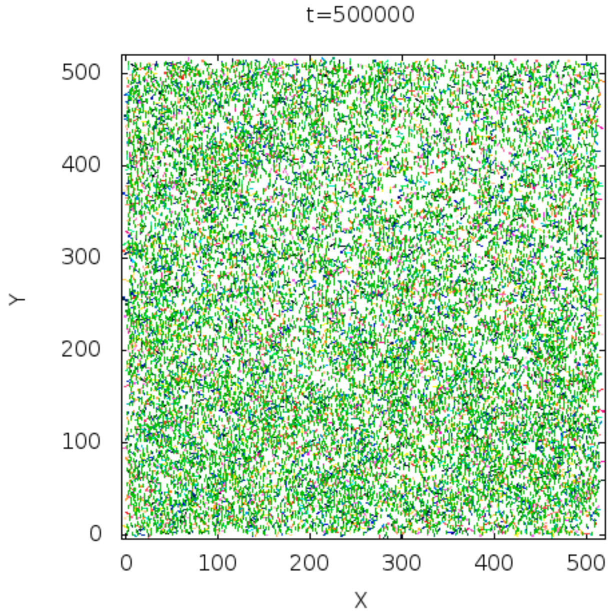
Morphological characterization?





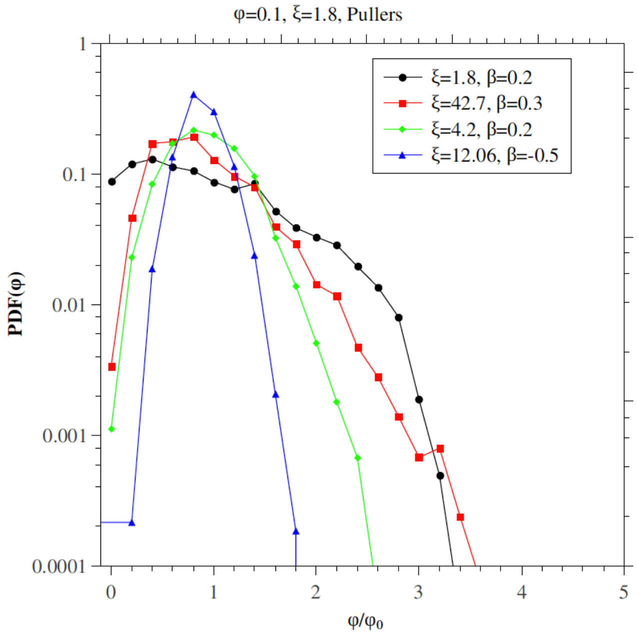
5. Cluster morphologies



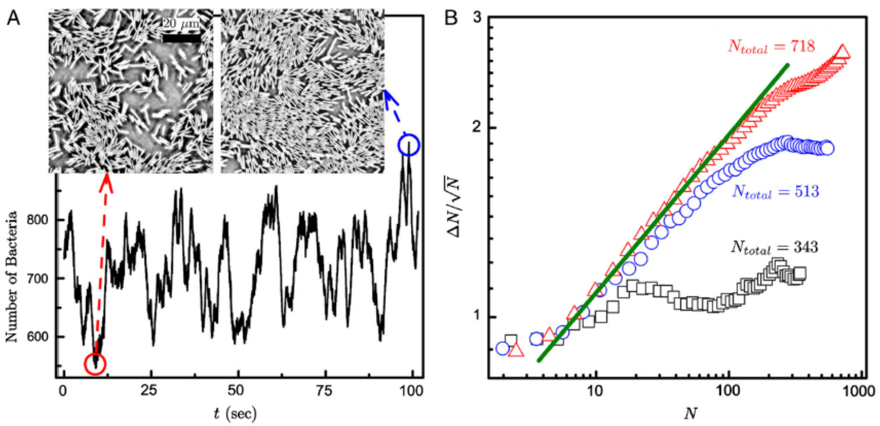


Dynamic structures

Morphological characterization?



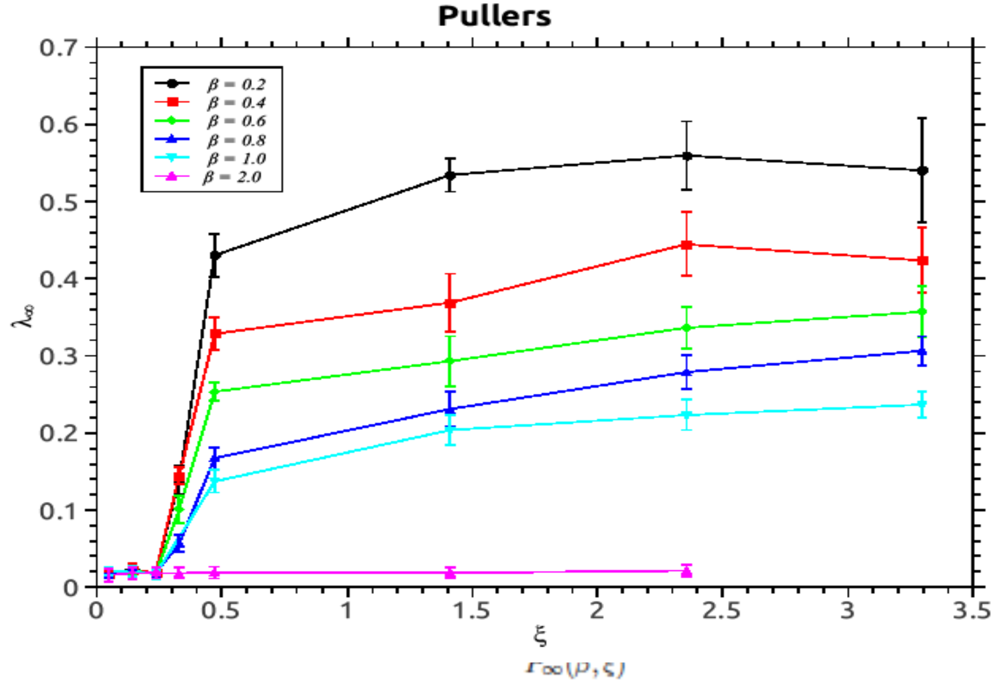
Density fluctuations

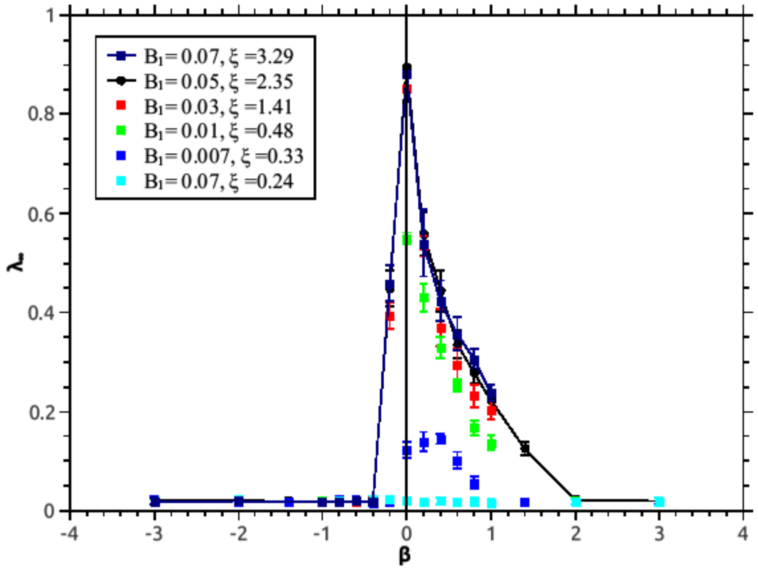


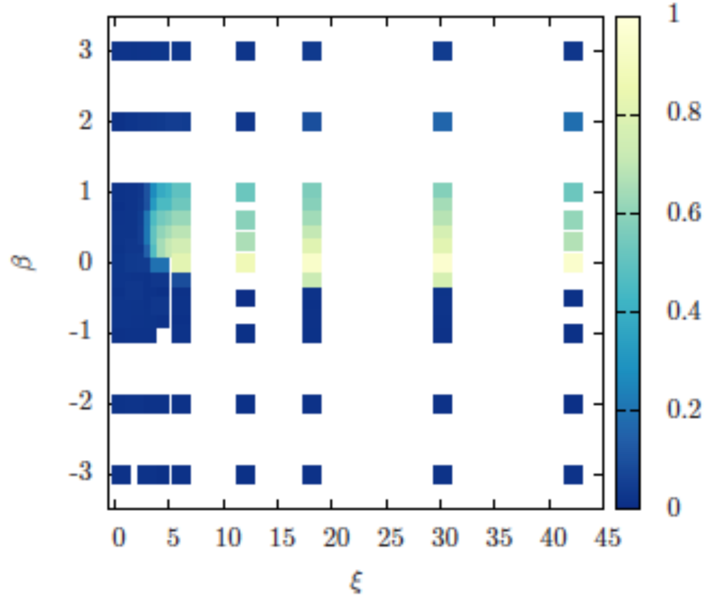
Zhang et al. PNAS (2010)

5. Squirmer suspensions: density fluctuations

Quantify degree of ordering
sensitive to active stresses
distinguish puller/pusher



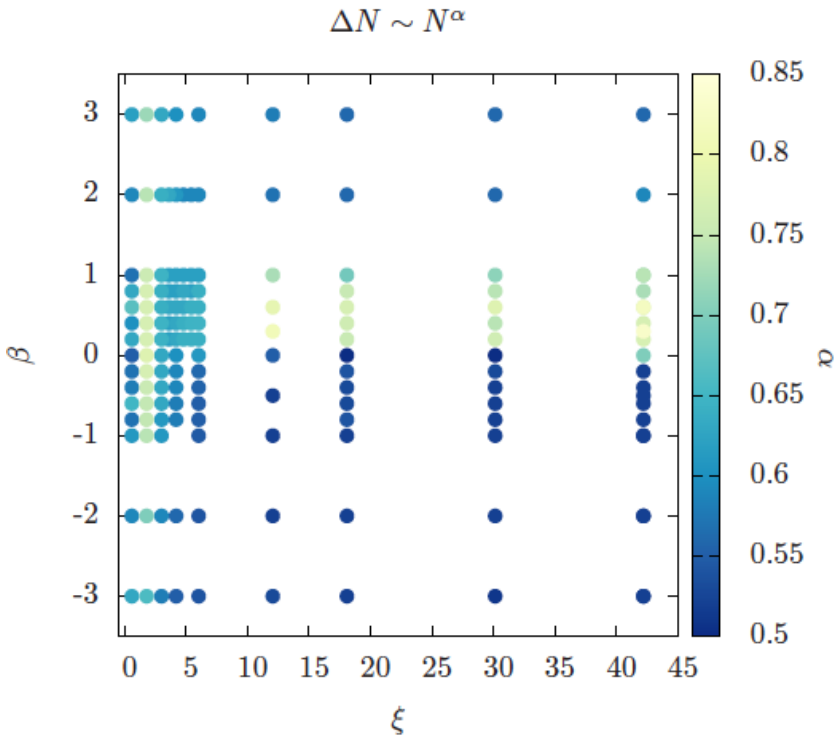


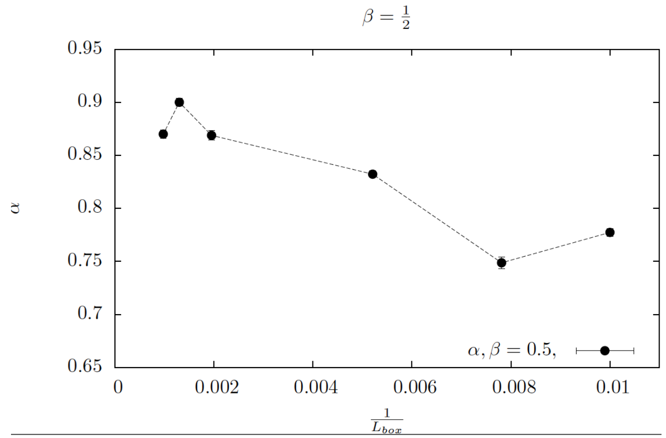


Squirmer attraction
enhances cohesion
destroys ordering

5. Squirmer suspensions: density fluctuations

Effect on density fluctuations
favours large dynamic clusters

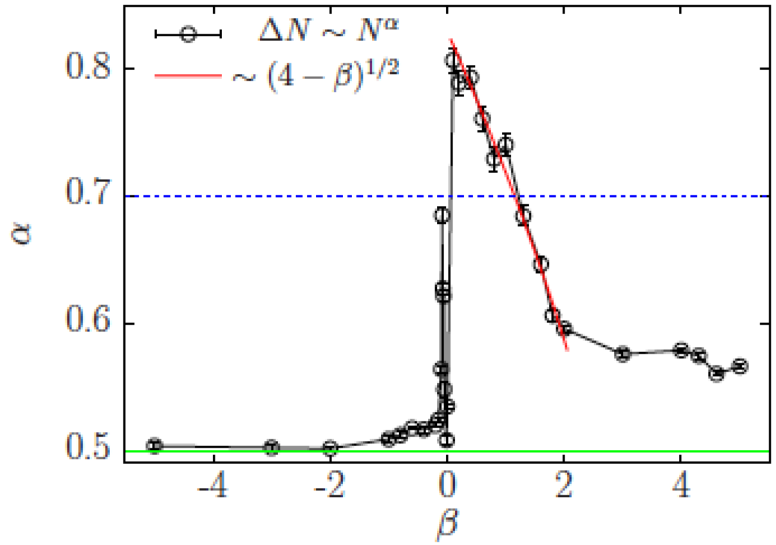




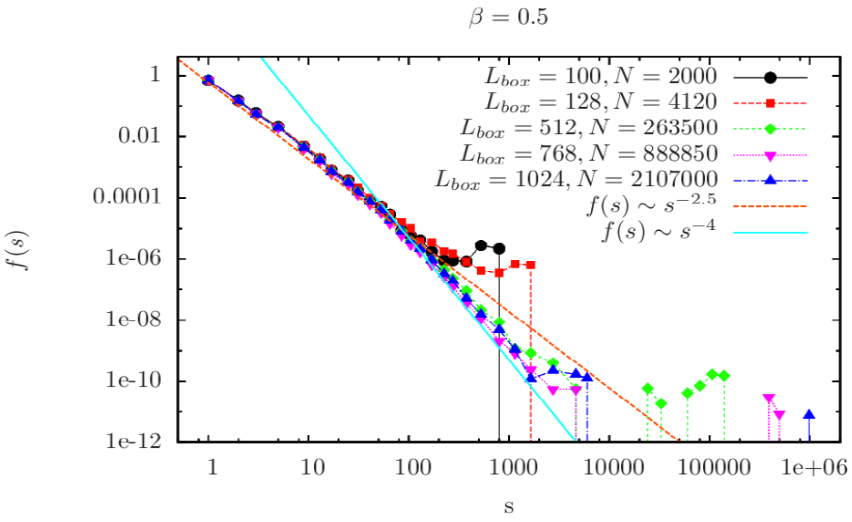
1/L

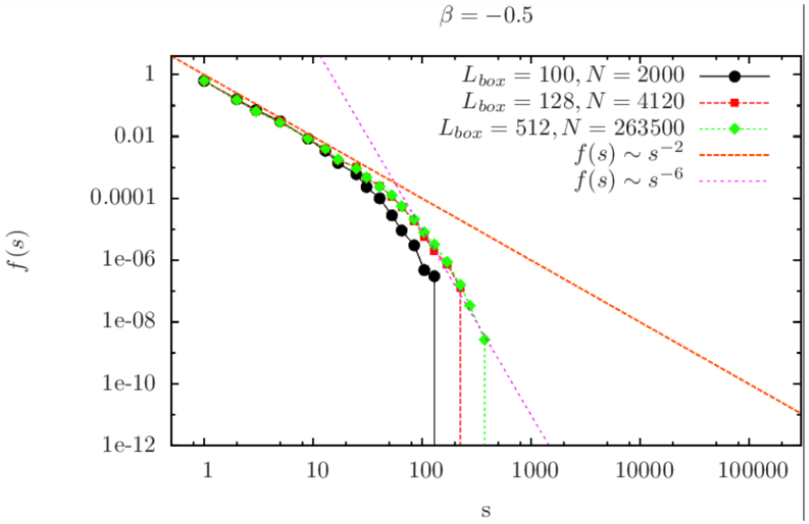
Significant finite size effects

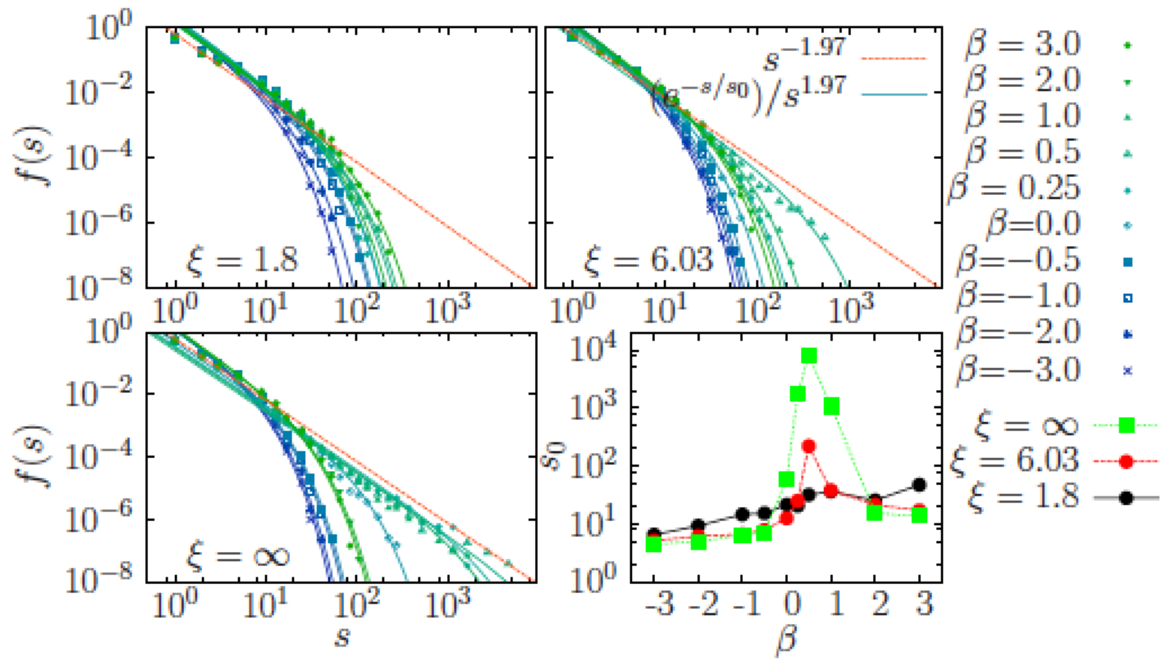
Need to reach large system sizes
Strong correlations



5. Squirmer suspensions: Cluster distributions







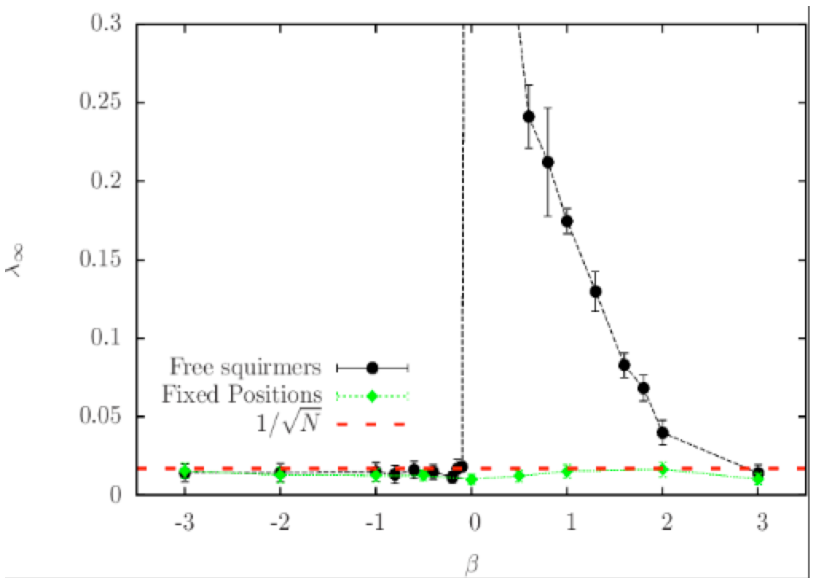
$$f(s) \sim s^{-\gamma_0} \exp(-s/s_0)$$

Power-law decay

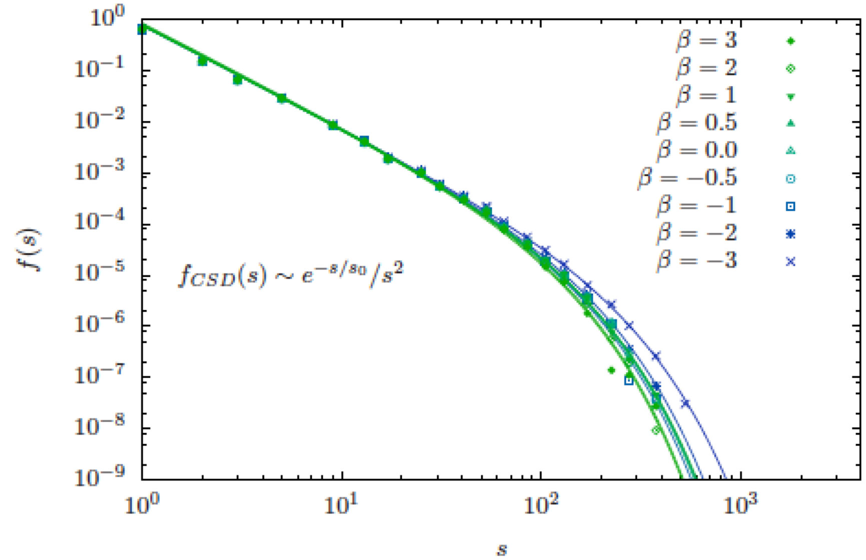
Wide range dynamic structures

5. Squirmer suspensions: Cluster distributions

Relevance of translational motion



.. and reorientation

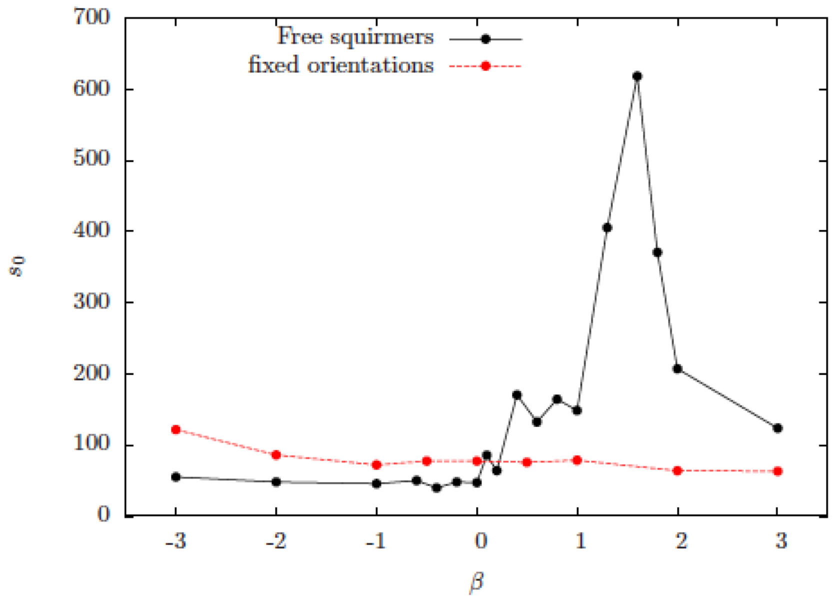


Hydrodynamic coupling

lack of translation/rotation coupling

Nematogenic character lost

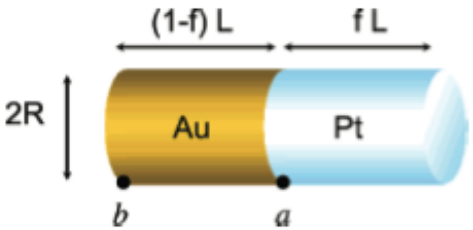
$$f(s) \sim \; s^{-\gamma_0} \; \exp(-s/s_0)$$



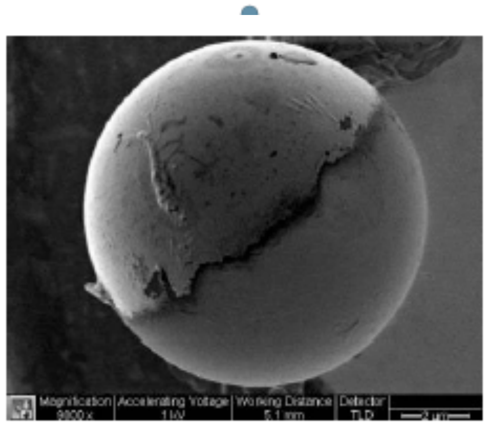
6. Chemical swimmer suspensions

What if particles might generate concentration gradients? → **self-propulsion!!!**

$$2H_2O_2 \rightarrow 2H_2 + O_2$$



(Paxton et al, JACS **126**, 13424 (2004))



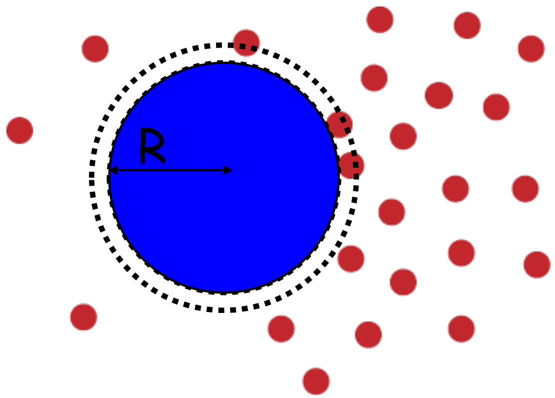
http://www.mems.duke.edu/fds/pratt/MEMS/faculty/benjamin.yellen

activity modelled by simple updating rule → non-conserved dynamics for ϕ!

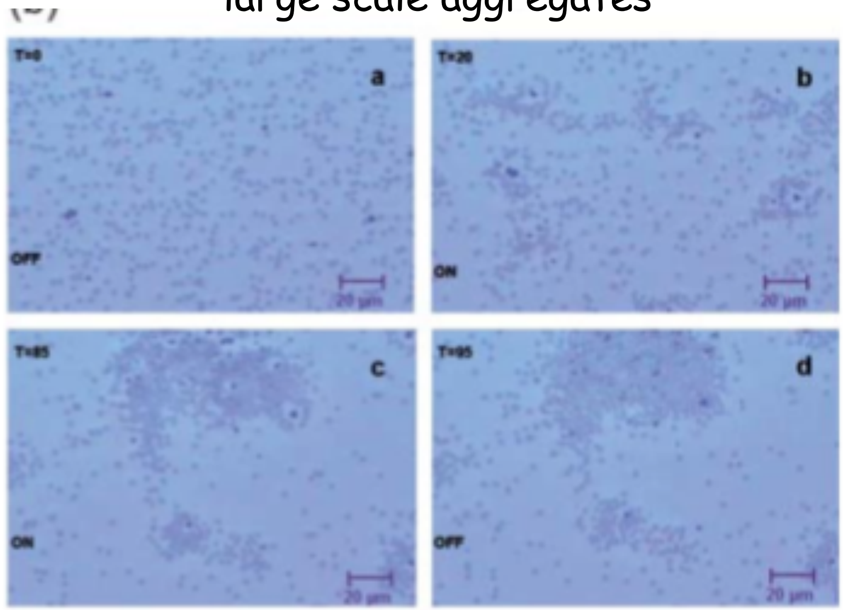
6. Colloidal phoresis: a multiscale "transport phenomenon"

<u>Phoretic transport</u>: motion of colloidal particle under the effect of external field (electric field, concentration/temperature gradients)

(solute molecules size)/(colloidal particle size)



large scale aggregates



(Sen et al, Faraday Discuss. **143**, 15 (2009))

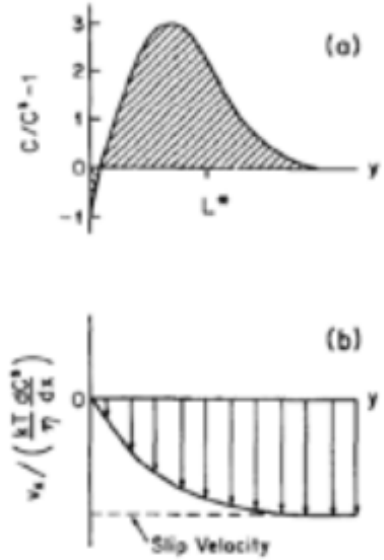
need for coarse-graining

$$\delta/R \to 0$$

Colloidal phoresis

Concentration interacts with a solid surface

delocalized membrane?



$$C = C^s \exp(-\Phi/kT),$$

Concentration gradient parallel to the surface

asymmetric change in chemical potential

Generates pressure gradient

Due to asymmetry induced by wall

$$\frac{\partial p}{\partial y} + C\frac{d\Phi}{dy} = 0,$$

Surface-induced flow

$$\eta\frac{\partial^2 v_x}{\partial y^2} - \frac{\partial p}{\partial x} = 0.$$

$$v^s = -\frac{kT}{\eta}\int_0^\infty y[\exp(-\Phi/kT) - 1]\, dy \frac{dC^s}{dx}.$$

6. Modeling colloidal phoretic transport

The presence of the solute can be taken into account by means of an effective "slip velocity" as boundary condition for the flow on the colloid surface

$$\mathbf{v}(\mathbf{r}_S) = \mu(\mathbf{r}_S)(\mathbf{I} - \hat{\mathbf{n}} \otimes \hat{\mathbf{n}})\nabla\phi(\mathbf{r}_S) \qquad \mathbf{r}_S \in \Sigma$$

$$\mu(\mathbf{r}_S) = \frac{k_B T}{\eta}\int_0^\infty r\,[1 - \exp(-\Psi(\mathbf{r})/k_B T)]\,dr$$

surface phoretic mobility

colloid/solute interaction potential (short-ranged)

→ velocity of a (spherical) particle of radius R

$$\mathbf{V} = -\frac{1}{4\pi R^2}\int\int_\Sigma \mu(\mathbf{r}_S)(\mathbf{I} - \hat{n} \otimes \hat{n})\nabla\phi(\mathbf{r}_S)d\mathbf{r}_S$$

$$\mathbf{v} = \mu\nabla\phi$$

Name	Field variable (Y_∞)	μ	Remarks
Electrophoresis	Electrical potential	$-\frac{\varepsilon\zeta}{4\pi\eta}$	ζ = zeta potential of particle surface
Diffusiophoresis	Concentration of a chemical species (nonionic)	$\frac{kT}{\eta}KL^*$	See (11) for K and L^*
Diffusiophoresis	Concentration of a chemical species (ionic)	$4\frac{kT}{\eta}\kappa^{-2}\left[\frac{\bar{\zeta}}{2}\beta - \ln(1-\xi^2)\right]^b$	$\bar{\zeta} = Ze\zeta/kT$; see (4) for κ^{-1}, (13) for ξ, and (17) for β
Thermophoresis	Temperature	$\frac{2}{\eta T}\int_0^\infty y\bar{h}\,dy$	$\bar{h}$ is the local specific enthalpy increment at distance y from the solid surface: $\bar{h} = h(y) - h(\infty)$

(Anderson, ARFM **21**, 61 (1985); Golestanian et al, New J. Phys. **9**, 126 (2007))

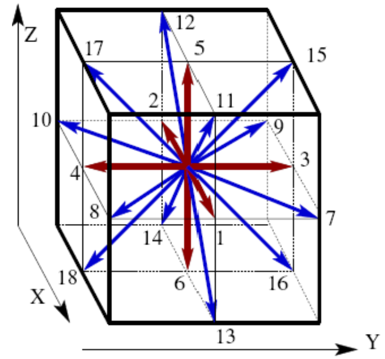
6. Numerical approach

(Hybrid) Lattice Boltzmann algorithm for multiphase fluids + (resolved) particles

Solvent

$$f_l(\mathbf{x}+\mathbf{c}_l\Delta t, t+\Delta t) - f_l(\mathbf{x},t) = -\frac{\Delta t}{\tau}\left(f_l(\mathbf{x},t) - f_l^{(eq)}(\mathbf{x},t)\right)$$

$$\rho(\mathbf{x},t) = \sum_l f_l \qquad \rho(\mathbf{x},t)\mathbf{u}(\mathbf{x},t) = \sum_l \mathbf{c}_l f_l \qquad l = 0,\ldots,18$$



in the limits $Kn = \frac{\lambda_{mfp}}{L} \ll 1 \quad Re_p = \frac{vR}{\nu} \ll 1 \quad Ma = \frac{v}{c_s} \ll 1$

$$\rho[\partial_t \mathbf{v} + \mathbf{v}(\nabla\cdot\mathbf{v})] = -\nabla P - \phi\nabla\mu + \eta\nabla^2\mathbf{v}$$

"Fuel"

$$\partial_t\phi + \mathbf{v}\cdot\nabla\phi = M\nabla^2\mu$$

Advection-diffusion equation via finite differences

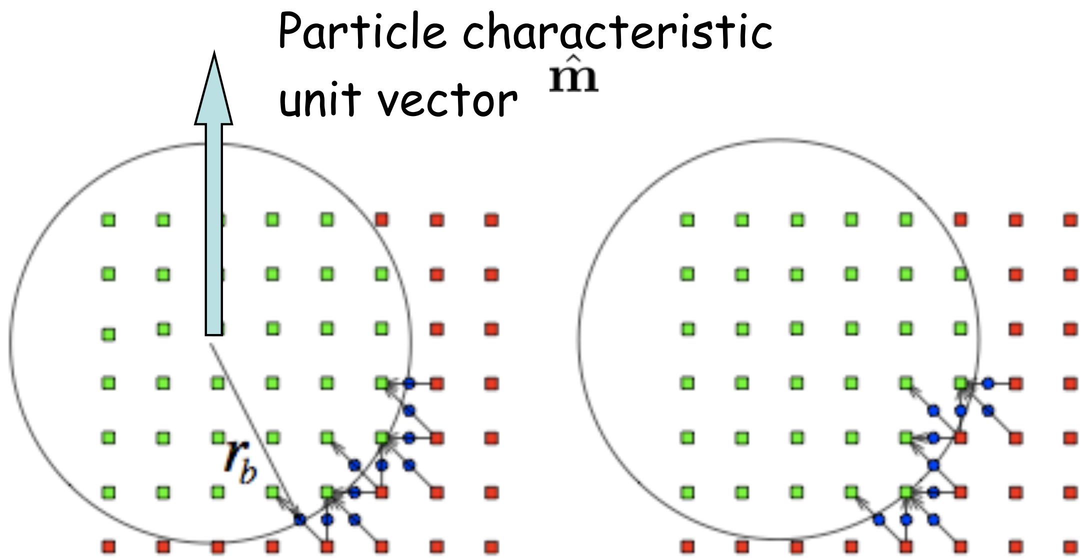
feedback on the fluid via a forcing term in the LB equilibria $\mathbf{F} \propto \phi\nabla\phi$

6. Numerical approach

Lattice Boltzmann algorithm for multiphase fluids + (resolved) particles

Colloidal particles

Suspended particles are defined by a set of of “links” between lattice nodes



(AJC Ladd, J. Fluid Mech. **271**, 285 (1994))

bounce-back-on-links algorithm:
mass/momentum conservation
between particle and fluid

$$f_{l'}(\mathbf{x}, t+\Delta t) = f_l^*(\mathbf{x}, t) - \frac{2a_{c_l}\rho\mathbf{u}_l\cdot\mathbf{c}_l}{c_s^2}$$

$$\mathbf{u}_l = \mathbf{U} + \boldsymbol{\Omega}\wedge(\mathbf{x}_l - \mathbf{X}_{CoM})$$

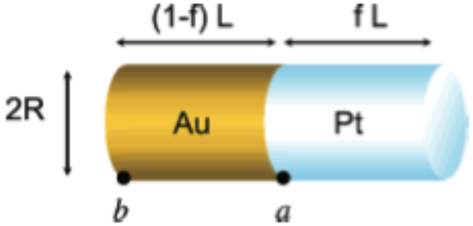
+ position dependent **slip velocity**
at the particle surface

$$\mathbf{v}(\mathbf{r}_S) = \mu(\mathbf{r}_S)(\mathbf{I} - \hat{\mathbf{n}}\otimes\hat{\mathbf{n}})\nabla\phi(\mathbf{r}_S)$$

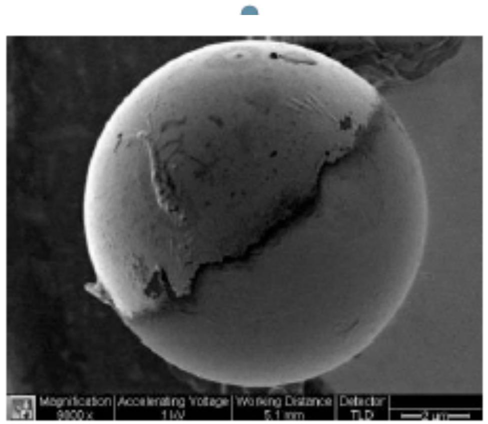
6. Chemical swimmer suspensions

What if particles might generate concentration gradients? → **self-propulsion!!!**

$$2H_2O_2 \rightarrow 2H_2 + O_2$$



(Paxton et al, JACS **126**, 13424 (2004))



http://www.mems.duke.edu/fds/pratt/MEMS/faculty/benjamin.yellen

activity modelled by simple updating rule → non-conserved dynamics for ϕ!

$$\phi(\mathbf{r},t) \rightarrow \phi(\mathbf{r},t) - \alpha(\mathbf{r})\phi(\mathbf{r},t)$$

→ particle surface activity

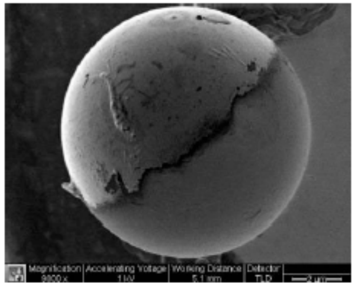
→ Inclusion of a global source term
(mimicks coupling with an external “bath” of concentration field)

$$\phi(\mathbf{r},t) \rightarrow \phi(\mathbf{r},t) + \delta\phi \qquad \delta\phi = \frac{1}{|\Omega|}\int_\Omega \phi(\mathbf{r},t)d\mathbf{r}$$

6. "Janus" particles

In particular we use an asymmetric surface activity of the form

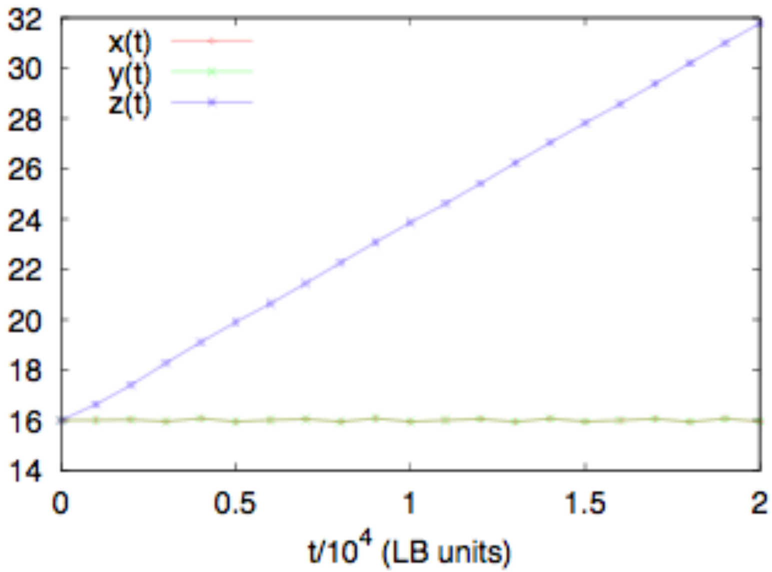
$$\alpha(\mathbf{r}) = \alpha_0 H\left(\theta - \frac{\pi}{2}\right) \rightarrow$$ Janus particle

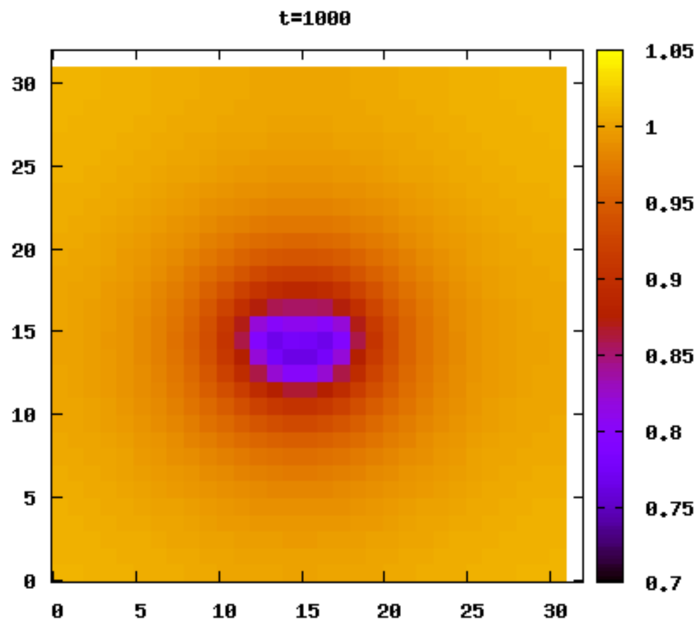


http://www.mems.duke.edu/fds/pratt/MEMS/faculty/benjamin.yellen

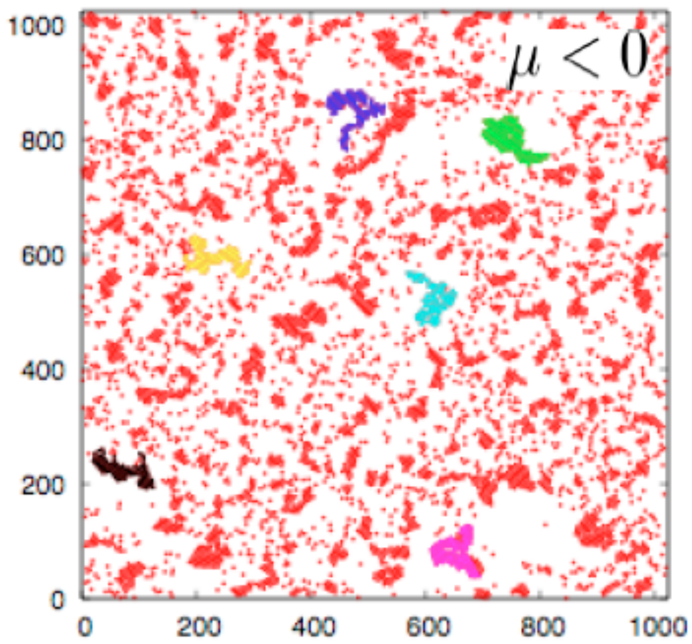
The velocity of an isolated particle of constant mobility μ can be computed exactly

$$\mathbf{V} = \frac{\mu\alpha_0}{4D}\hat{\mathbf{m}}$$

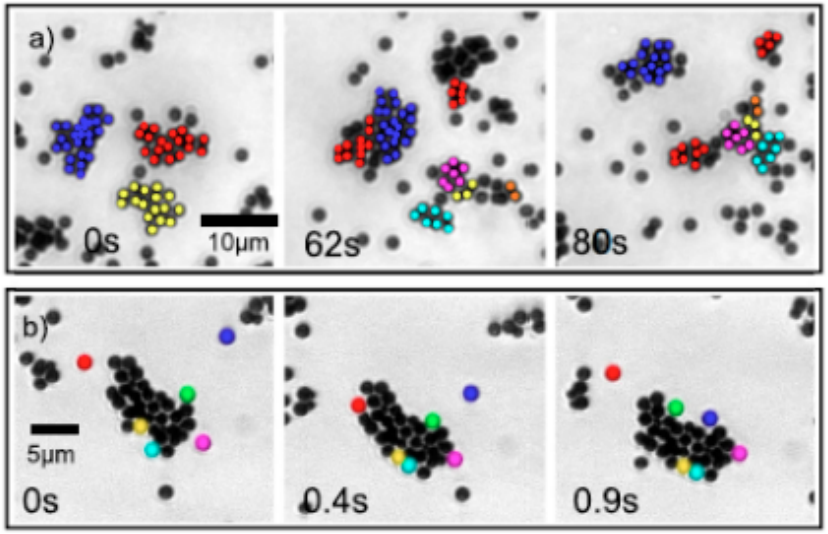


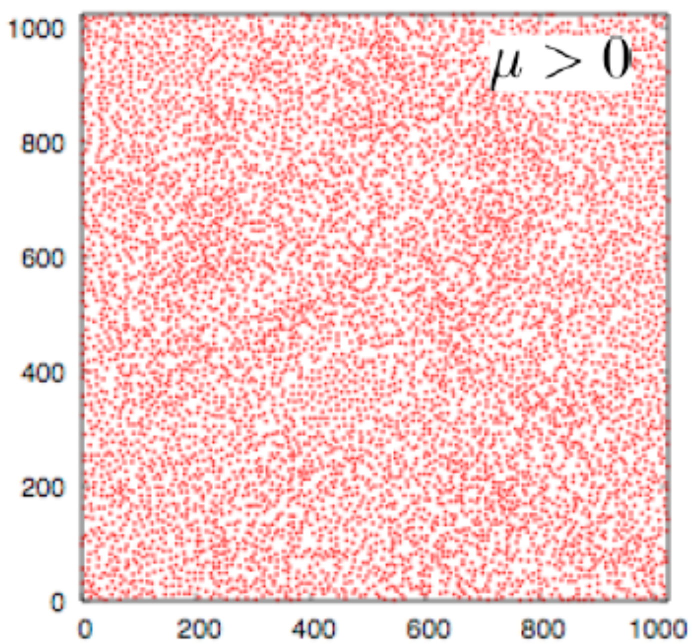


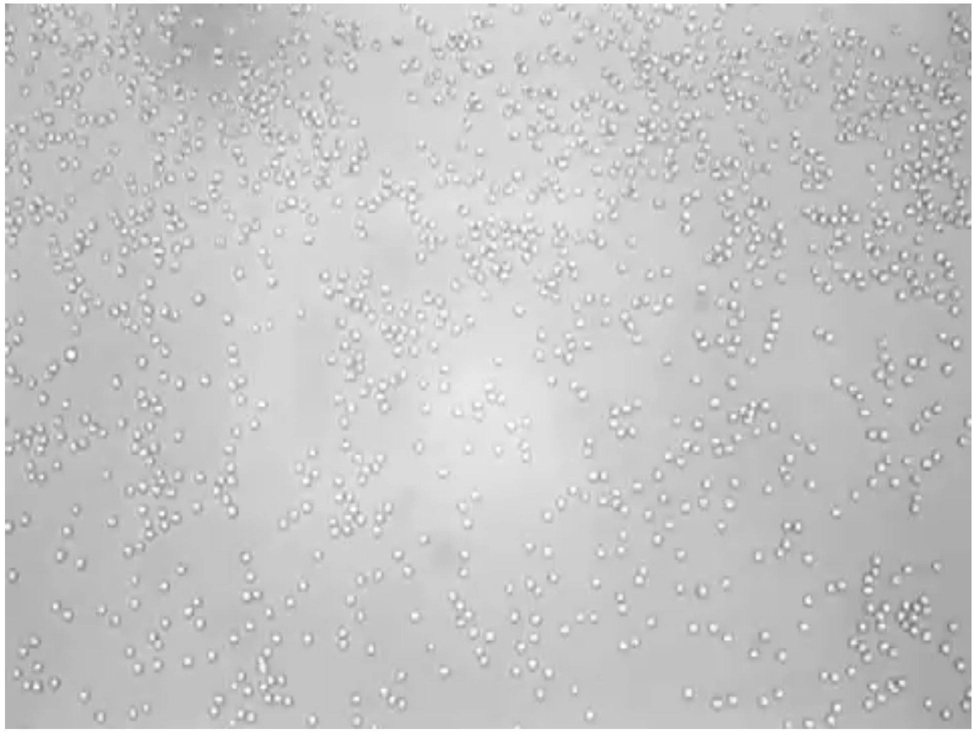
6. Collective dynamics in “2D”: phoretic mobility?



Teurkhauff et al. PRL (2012)



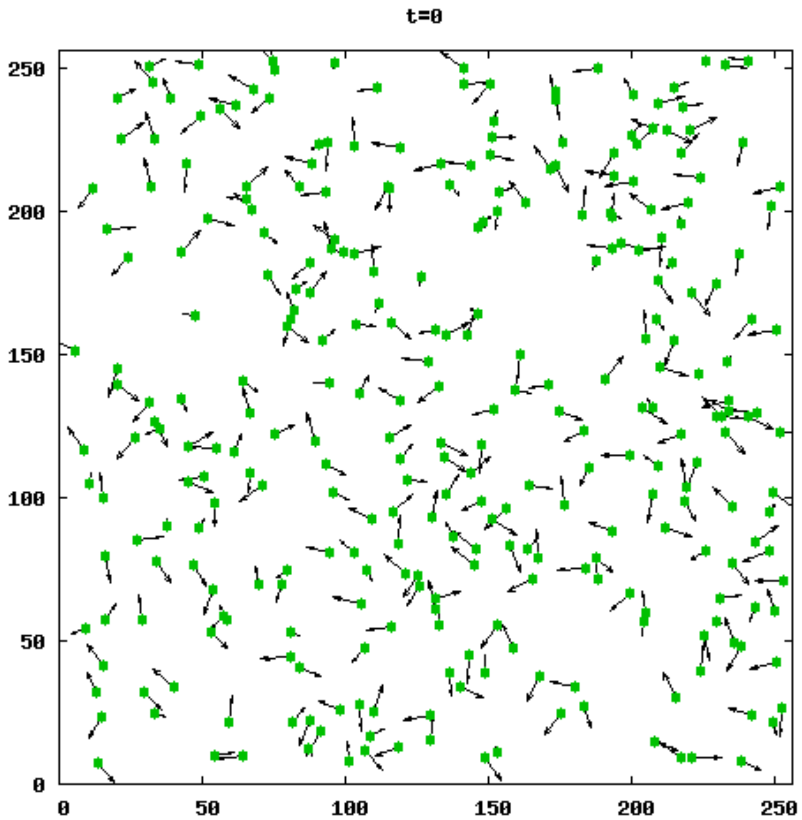


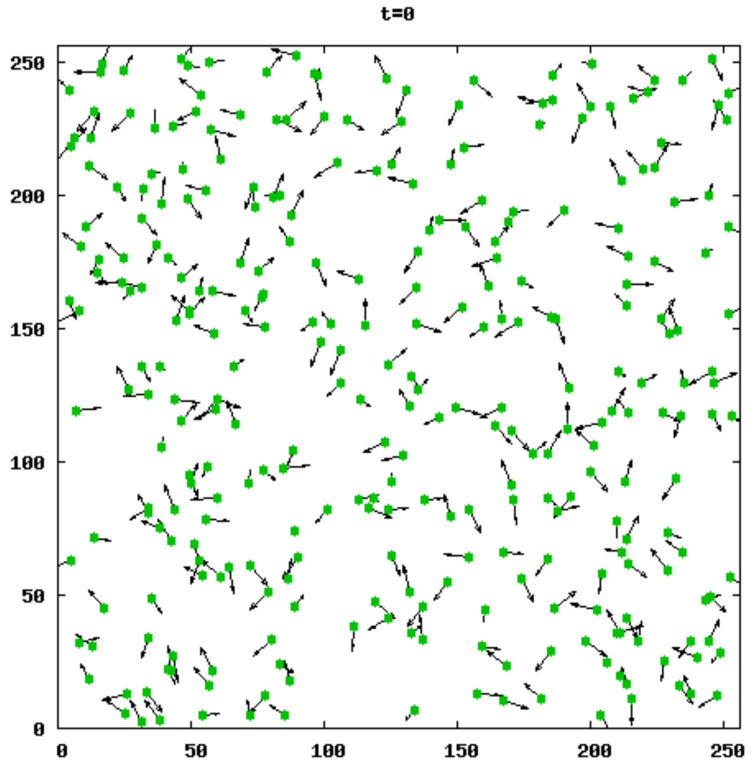


Palacci et al. Science (2013)

6. Repulsive chemical swimmers

No hydrodynamics





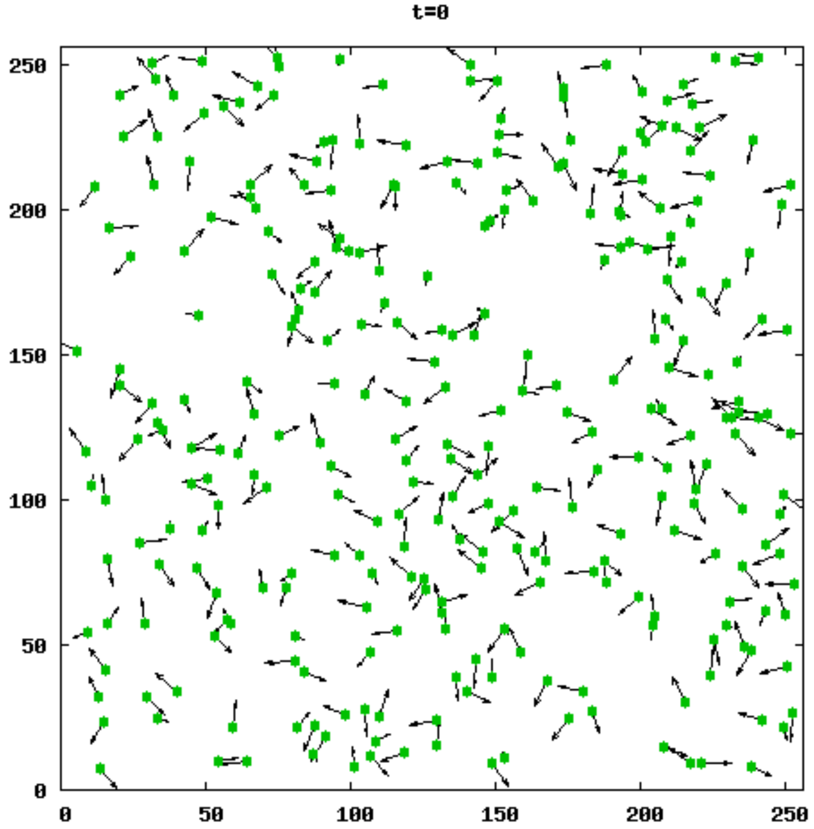
Towards a crystal structure

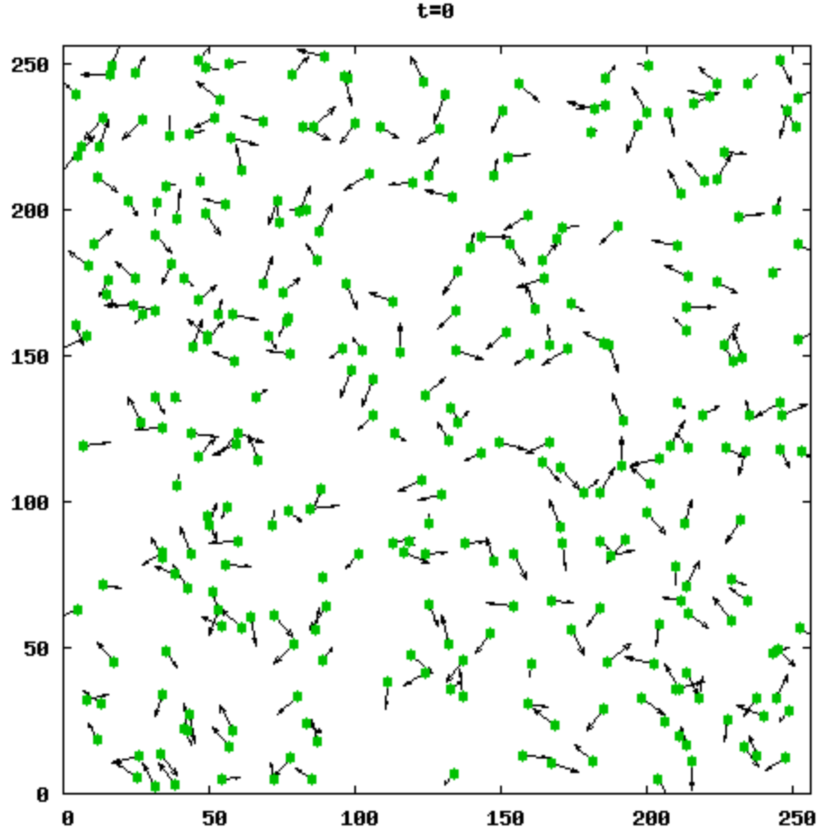
Faster dynamics

larger number of "defects"

6. Attractive chemical swimmers

No hydrodynamics





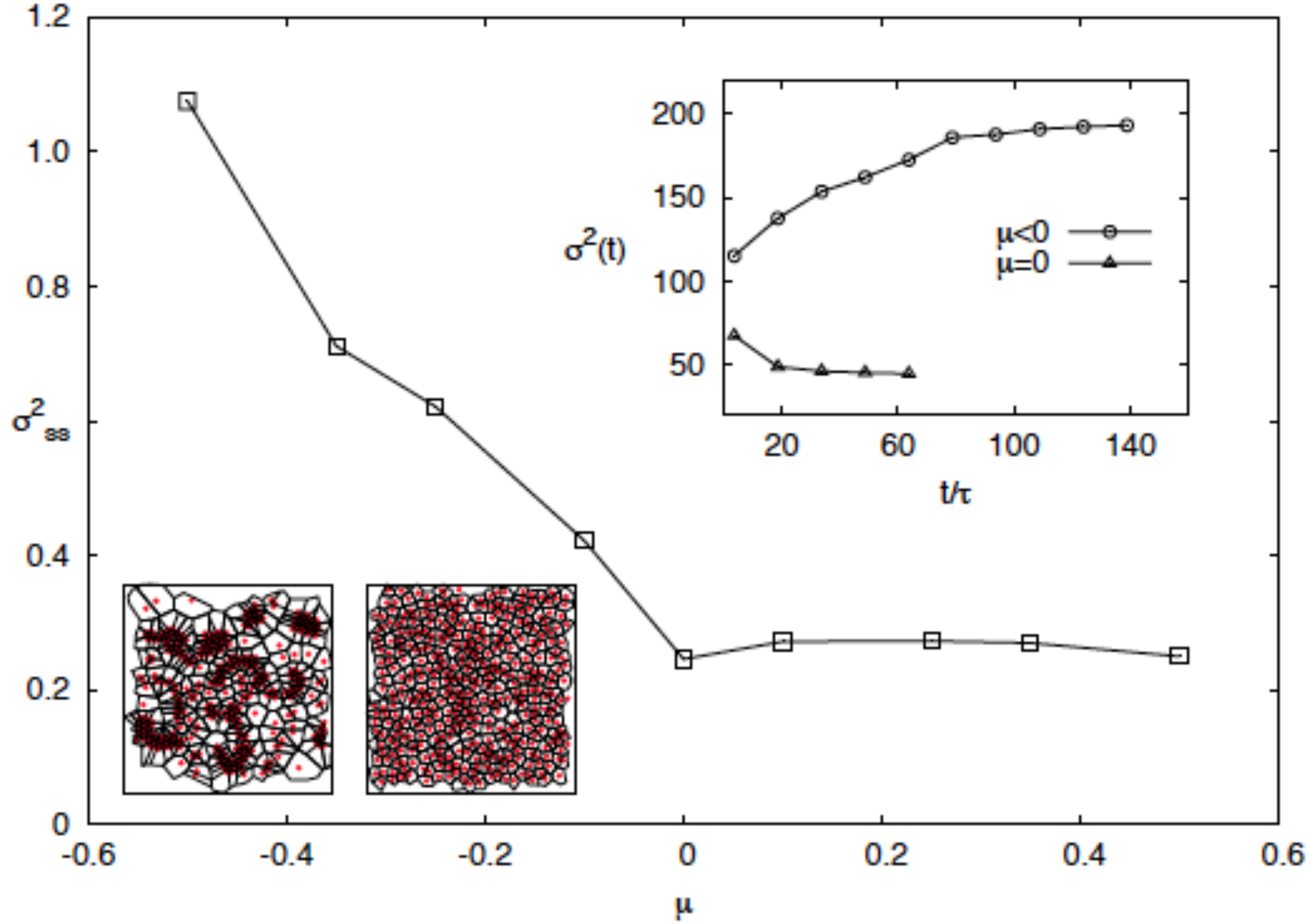
Cluster formation

6. Density fluctuations

A proper indicator to distinguish dynamical regimes?

Use variance of Voronoi tesselation

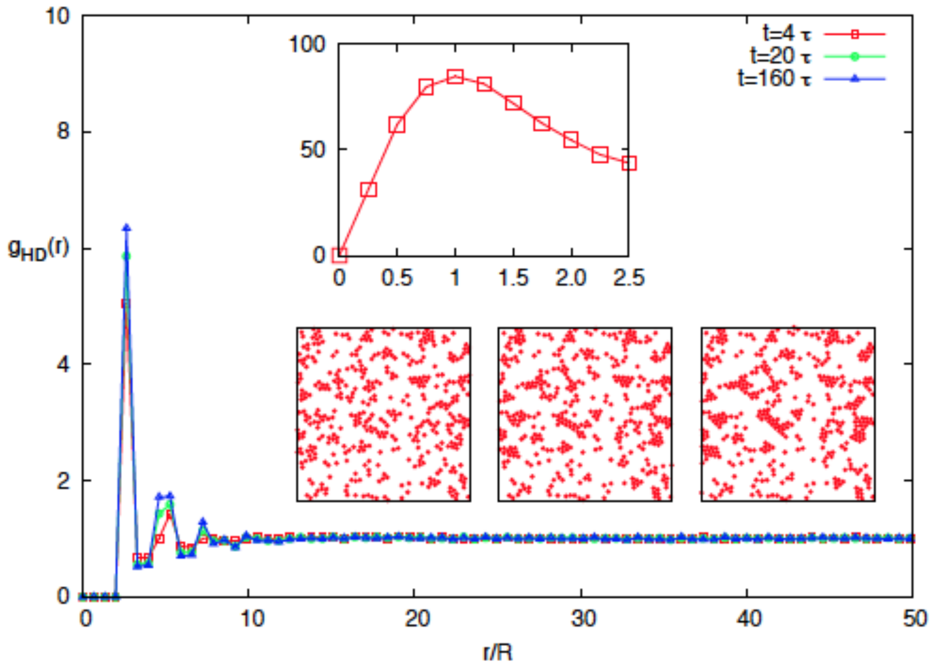
$$\sigma_{\mathcal{S}}^2(t) \equiv (1/N)\sum_{i=1}^{N}(\mathcal{S}_i - \overline{\mathcal{S}})^2$$



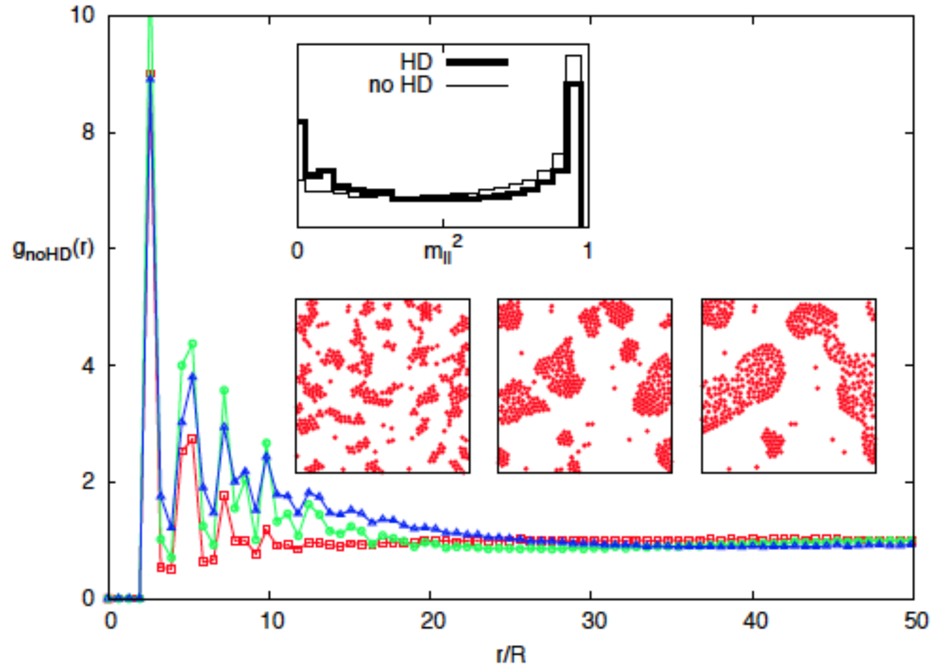
6. Radial distribution functions

Clustering regime $\mu = -0.5$

With hydrodynamics



Without hydrodynamics



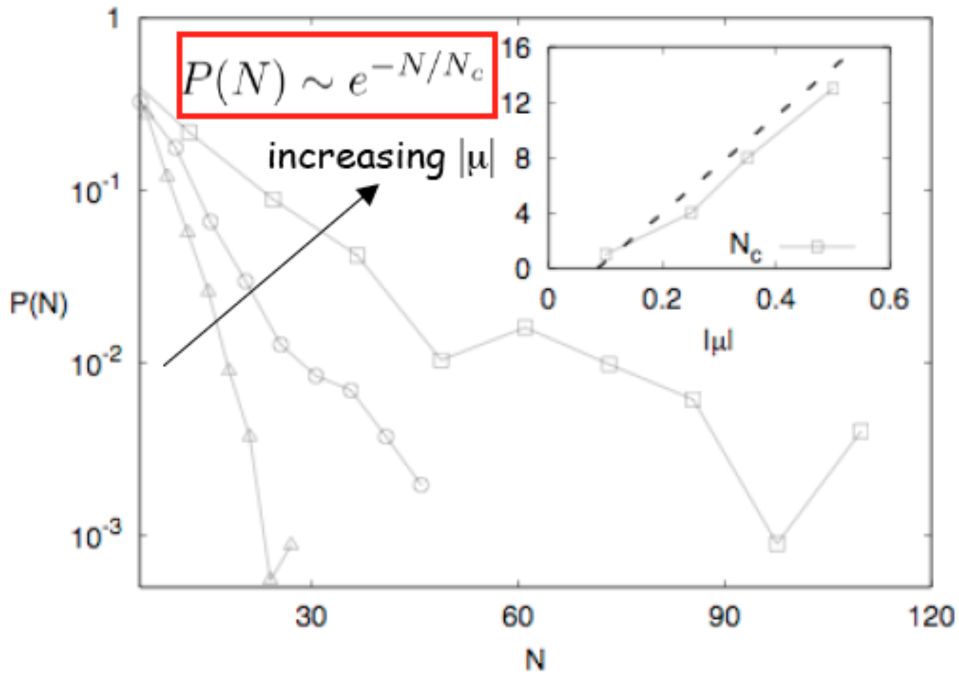
$$\mathcal{D}(\tilde{t}) = \int_0^{\infty} \left(g_{HD}(r,\tilde{t}) - g_{noHD}(r,\tilde{t})\right)^2 dr$$

No Hydro: larger friction

$$\tau_f^{(HD)} \approx 5\tau_f^{(noHD)}$$

6. Statistics and geometry of clusters

PDF and mean particle number
as function of coupling strength



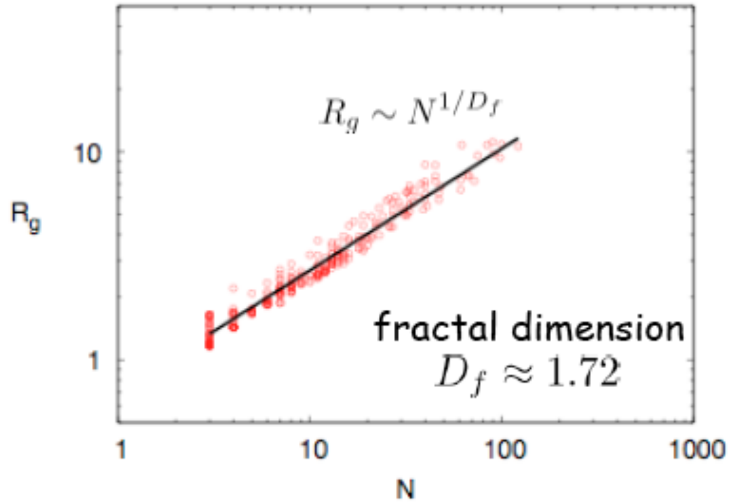
$\langle N \rangle \sim V_p \Rightarrow N_c \sim \mu$

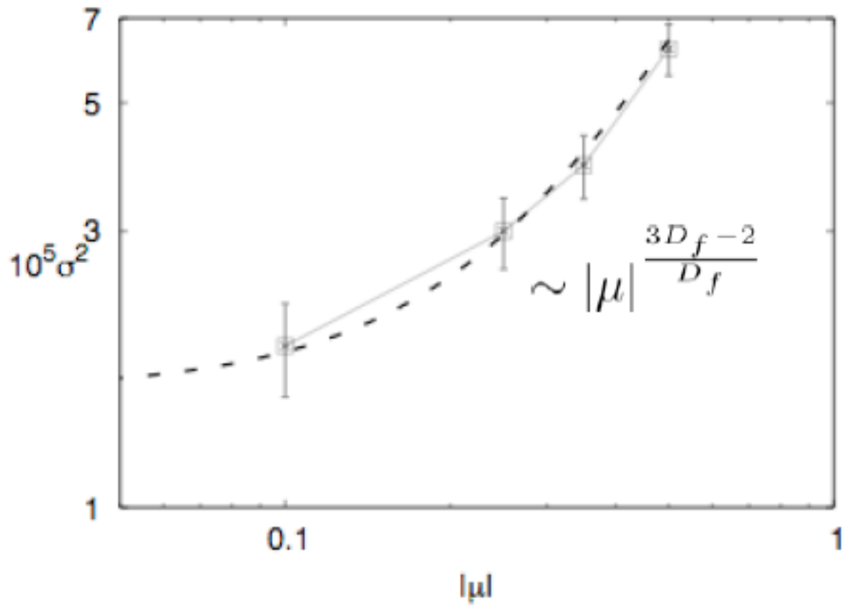
(Theurkauff et al)

colloids live on a set $\mathcal{S} = \bigcup_{i=1}^{N_{clus}} \mathcal{C}_i$ (i-th cluster)

$$\sigma_\rho^2 \sim \langle \rho(x)^2 \rangle \propto \sum_{i=1}^{N_{clus}} \left(\frac{N_i}{\mathcal{A}_i}\right) \mathcal{A}_i P(\mathcal{A}_i) \sim \mu^{\frac{3D_f-2}{D_f}}$$

gyration radius vs number of colloids





7. Conclusions

Active matter

Release energy at small scales (natural/synthetic)

intrinsically out of equilibrium

New mechanisms to develop patterns and structures

Competition between attraction/activity

Interplay hydrodynamics/attraction

Large density fluctuations

Macroscopic cluster

Induce polar ordering: dominant effect of translation/rotation

Dynamic clusters

Interplay hydrodynamics/attraction

Large density fluctuations