Active Brownian Particles at High Densities

Thomas Voigtmann

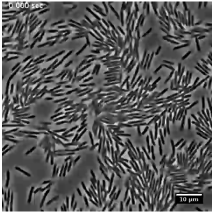
Theory of Soft Matter Group, German Aerospace Cologne / University of Düsseldorf

Jülich, February 2018

Collective Dynamics in Self-Propelled Agents

[Daniel Biber, Wildlife Photography Award 2017]

[Nicholas Hope, `http://www.bubblevision.com/`]

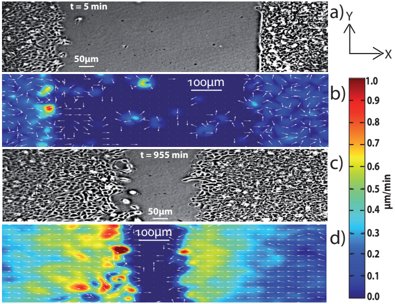


[Matthew Copeland, U Wisonsin, Madison]

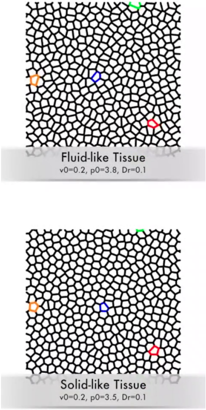
- birds flock, fish school, insects swarm, bacteria move collectively
- emergent phenomena on *many length scales*
- *interaction mechanisms? minimal models for collective behavior?*

Collective Dynamics in Biological Tissue

- cell structures show *collective rearrangements*
- driven by *active processes*
  - *molecular crowding slows down* the motion
  - *self propulsion speeds up*



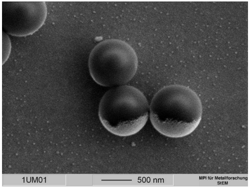
velocities in wound healing [Nnetu *et al.*, New J Phys (2012)]

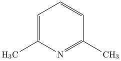


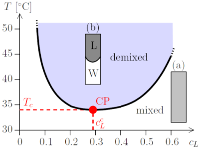
[Bi *et al.*, Phys Rev X (2016)]

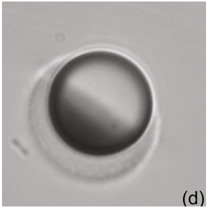
Microswimmers: Janus particles

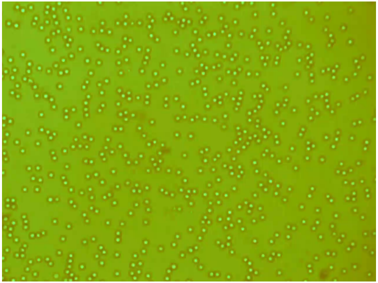
- *active-particle systems* harvest energy
  $\Rightarrow$ *directed motion*
- *model systems for non-equilibrium stat. phys.:*
  Janus-particle colloids in specific solvents





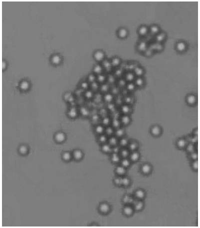


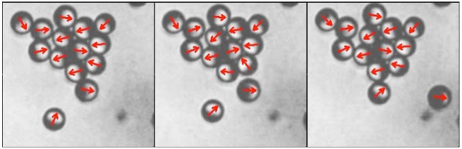




[C. Bechinger lab, U Stuttgart]

Motility-Induced Phase Separation (MIPS)



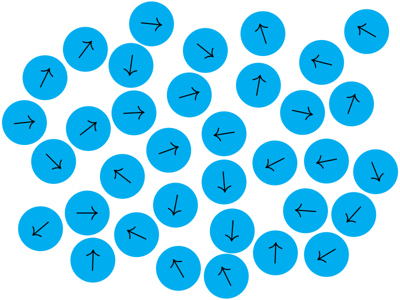


[Buttinoni *et al.*, Phys Rev Lett (2013)]

- active Brownian particles form dynamic clusters
- *non-equilibrium* analogue to liquid–gas phase separation (!?)
- mechanism: *swim lower at high density* (interaction effect)

Active Brownian Particles

Active Brownian Hard Spheres

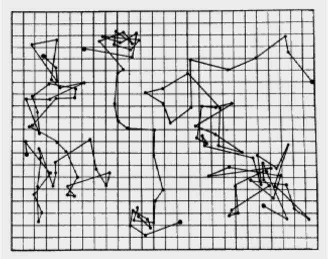


- hard spheres: spherical steric interactions
- activity: individual drift with random (uncoupled) direction

Active Brownian Particles

Brownian Motion

Passive Colloidal Systems: Brownian Motion



[Perrin, Ann Chim Phys VIII (1909)]

- $a \sim 1\ \mu\mathrm{m}$ sized particles in suspension
- perpetual *agitated, erratic* motion
- *not connected to "living force"*
- Jan Ingenhousz (1785): coal dust
- Robert Brown (1827): pollen
- Jean-Baptiste Perrin
  Nobel prize (1926)

scales:

$kT \sim 4\,\mathrm{pN\,nm}$

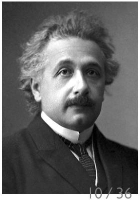
$a \sim\ \mu\mathrm{m}$

$\tau \sim\ \mathrm{ms}$

Brownian Motion: Statistical Interpretation

collisions with solvent molecules $\Rightarrow$ *independent displacements*

$$p(\Delta\vec{r}, t) = \frac{1}{(4\pi D t)^{3/2}} e^{-\Delta\vec{r}^2/4Dt}$$



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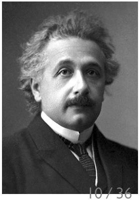
solves diffusion equation – *diffusion coefficient* D

$$\partial_t p = D\,\vec{\nabla}^2 p$$



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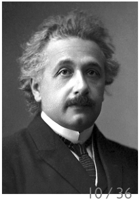
mean-squared displacement (MSD) is *diffusive*

$$\langle \Delta\vec{r}^2 \rangle = (2d)Dt$$



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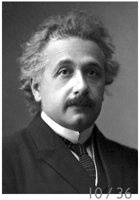
Stokes-Einstein(-Sutherland) relation: consequence of fluctuation-dissipation theorem (FDT)

$$D = k_B T / \zeta$$

fluctuation

dissipation

diffusivity $D \Leftrightarrow$ *mobility* $\mu = 1/\zeta$



Smoluchowski Equation

N-particle configuration-space distribution function $p(\Gamma, t)$,

$$\partial_t p + \underline{\nabla} \cdot (\underline{u} p) = 0$$
$$\underline{u} = \underline{\underline{\mu}} \cdot \underline{F} - \underline{\underline{D}} \cdot \underline{\nabla} \ln p$$

Smoluchowski (1905):

$$\boxed{\partial_t p = \underline{\nabla} \cdot \underline{\underline{D}} \cdot (\underline{\nabla} - \beta \underline{F}) p \qquad \beta = 1/kT}$$

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equivalent stochastic differential equation (Langevin):

$$\boxed{d\vec{r}_j = \mu \vec{F}_j \, dt + \sqrt{2D} d\vec{W}_j}$$

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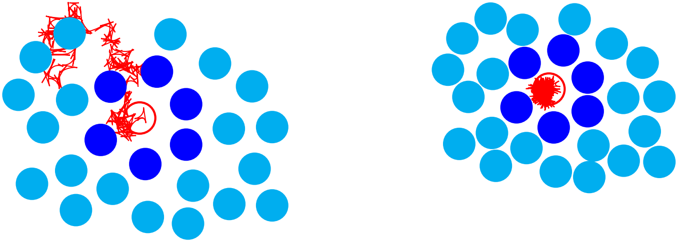
FDT, and $F = -\underline{\nabla} U$
$\Rightarrow$ MSD at most diffusive, $\langle \delta r^2(t) \rangle = o(t)$


Passive High-Density Dynamics: Glass Transition



structural relaxation time $\tau \to \infty$

"cage effect" – MSD subdiffusive

fluid

dynamical response functions

decay to zero

glass

response to perturbation

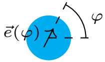
never decays

Active Brownian Particles

Active Motion

Model: Active Brownian Particles (2D)

Brownian motion with "active drift" term
along instantaneous orientation $\vec{e}(\varphi) = (\cos\varphi, \sin\varphi)^T$



$$d\vec{r}_j = \mu \vec{F}_j \, dt + v_0 \vec{e}(\varphi_j) \, dt + \sqrt{2D_t} d\vec{W}_j \,, \qquad D_t = kT/\zeta \,,$$

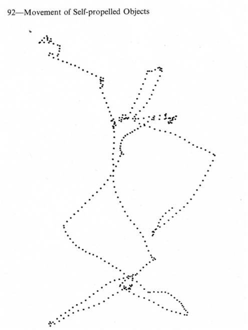
$$d\varphi_j = \sqrt{2D_r} dW_{\varphi,j}$$

Smoluchowski equation

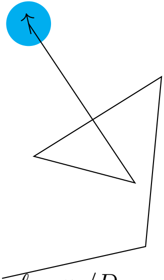
$$\partial_t p(\Gamma, t) = \Omega(\Gamma) \, p(\Gamma, t) \qquad \Gamma \equiv \{\vec{r}_j, \varphi_j\}_{j=1,\ldots N}$$

$$\Omega = D_t \sum\nolimits_j \vec{\nabla}_j \cdot (\vec{\nabla}_j - \beta \vec{F}_j) - v_0 \vec{\nabla}_j \cdot \vec{e} + D_r \partial^2_{\varphi_j} \equiv \Omega_{\mathrm{eq}} + \delta\Omega$$

Low-Density Active Brownian Motion: Effective Diffusion



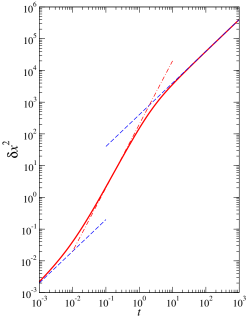
[Berg, Random Walks in Biology (Princeton, 1983)]



$$\ell \sim v_0/D_r$$

$$D_{\text{swim}} \sim \ell^2/\Delta t$$

$$D_{\text{eff}} \sim D_t + v_0^2/D_r$$



- short times *passive Brownian*: $\langle \delta x^2 \rangle \sim D_t t$
- long times *active diffusion*: $\langle \delta x^2 \rangle \sim D_{\text{eff}} t$
- *intermediate ballistic motion*: $\langle \delta x^2 \rangle \sim v_0^2 t^2$

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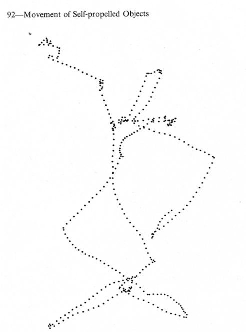

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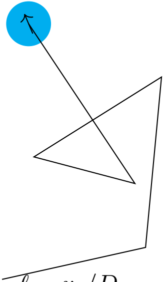

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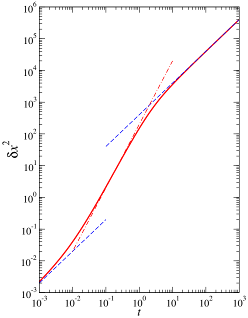

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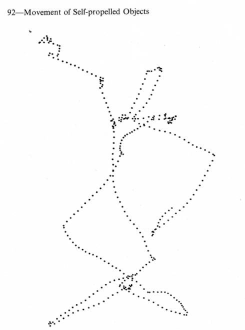

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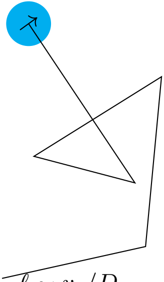

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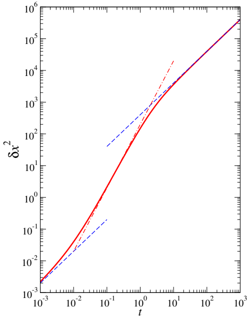

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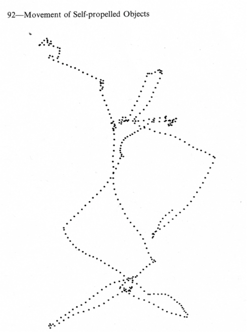

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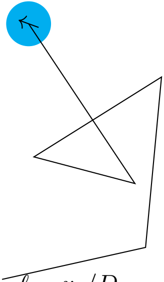

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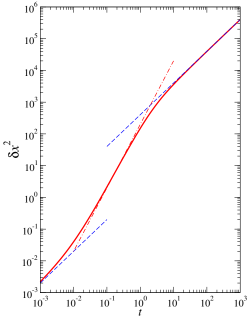

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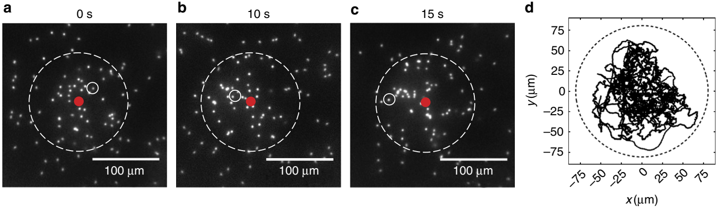
Low-Density Active Brownian Motion: Experiment



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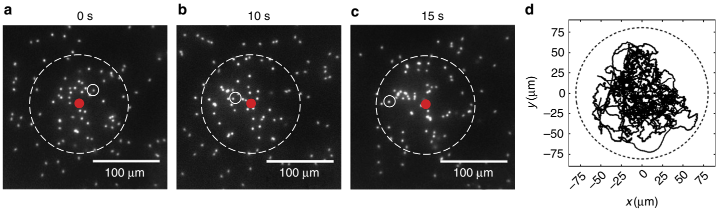

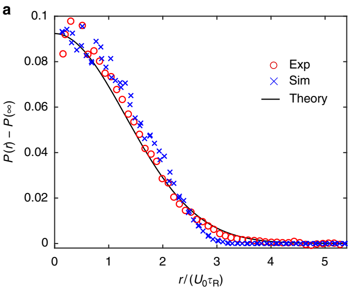

- in confinement $L \gg \ell$: *effective equilibrium*
- "swim pressure"

$$p = \rho\zeta D_{\text{eff}} = \rho k T_{\text{eff}}$$

- "effective temperature" $T_{\text{eff}}/T = 1 + v_0^2/2D_r$

[Takatori *et al.*, Nature Commun (2016)]
[Takatori, Yan, Brady, Phys Rev Lett (2014)]

Activity as Effective Temperature?

low density: *activity* $\mapsto T_{\text{eff}} \sim v_0^2/D_r$

Question:

still true at high density?

$\ell_{\text{interaction}} \lesssim \ell_{\text{p}}$

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Question:

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Answer:

NO!

Active Glasses

Mode-Coupling Theory of the Glass Transition

transient density correlation function

$$S_{ll'}(\vec{q},t) = \langle \varrho_l(\vec{q}) \exp[\Omega^\dagger t] \varrho_{l'}(\vec{q}) \rangle_{\mathsf{eq}} \qquad \varrho_l(\vec{q}) = \sum_j e^{i\vec{q}\cdot\vec{r}_j} e^{il\varphi_j}$$

Mori-Zwanzig formalism, mode-coupling approximation

$$\partial_t \boldsymbol{S}(\vec{q},t) + \boldsymbol{\omega}_{T+R}(\vec{q}) \cdot \boldsymbol{S}^{-1}(q) \cdot \boldsymbol{S}(\vec{q},t) + \int_0^t \boldsymbol{m}(\vec{q},t-t') \cdot \boldsymbol{\omega}_T^{-1}(\vec{q}) \cdot \left[\partial_{t'} \boldsymbol{S}(\vec{q},t') + \boldsymbol{\omega}_R \cdot \boldsymbol{S}(\vec{q},t')\right] dt' = \boldsymbol{0}$$

$$m_{ll'}(\vec{q},t) \approx \frac{1}{2N} \sum_{\vec{p},l_1\ldots l_4} \mathcal{V}_{ll_1l_2}(\vec{q},\vec{k},\vec{p}) \mathcal{V}^{+}_{l'l_3l_4}(\vec{q},\vec{k},\vec{p}) \Phi_{l_1l_3}(\vec{k},t) \Phi_{l_2l_4}(\vec{p},t)$$

$$\omega_{T,ll'}(\vec{q}) = q^2 D_t \delta_{ll'} - \frac{iv_0}{2} q e^{i(l-l')\varphi_q} S_{ll}(q) \delta_{|l-l'|,1} \qquad \omega_{R,ll'} = l^2 D_r \delta_{ll'}$$

Glass Transition

$$\boldsymbol{F}(q) = \lim_{t\to\infty} \boldsymbol{S}(q,t) \begin{cases} \neq \boldsymbol{0} & \textit{glass} \\ = \boldsymbol{0} & \textit{fluid} \end{cases}$$

Numerics

M coupled nonlinear integro-differential equations in $t \in [0, T]$,
typically $M = 10^3$, $T = 10^{6\ldots10}$

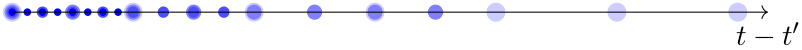
$$\partial_t f(t) = -a\, f(t) - \int_0^t m[f(t-t'), f(t-t')]\, \partial_{t'} f(t')\, dt'$$

straightforward discretization in t-domain, $f(t) \mapsto f_i \in \mathbb{C}^M$
implicit equation for each time step (solved by $O(10^{0\ldots5})$ iterations)

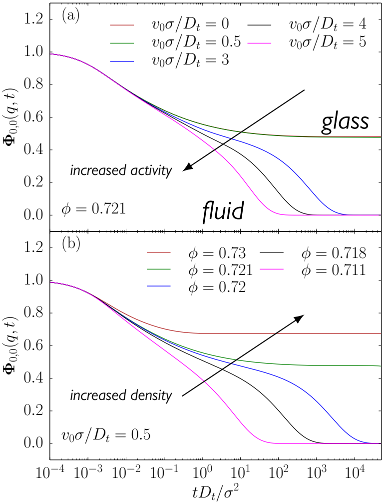
$$\boldsymbol{A} \cdot f_i = \boldsymbol{B} \cdot m[f_i, f_i] + \boldsymbol{C}_i$$

most time spent on evaluating $m[f, f]$

multi-grid decimation: $f_{2i}^{(h)} \mapsto f_i^{(2h)}$ (better for integrals)



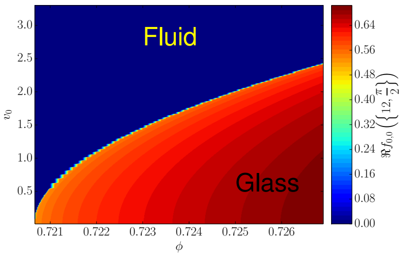
Slow Relaxation in Active Brownian Particle Systems

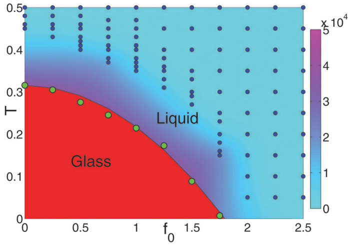


- increasing *activity melts the glass*
- high density causes *structural arrest*

$\Rightarrow$ *active glass* state possible

Glass Transition of Active Brownian Disks





[Mandal *et al.*, Soft Matter (2016)]

- activity softens, then melts the glass
- qualitative agreement with simulations of 3D ABP

[Liluashvili, Ónody, ThV, Phys Rev E **96**, 062608 (2017)]

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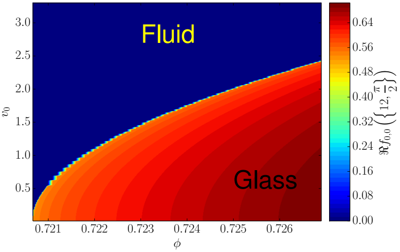

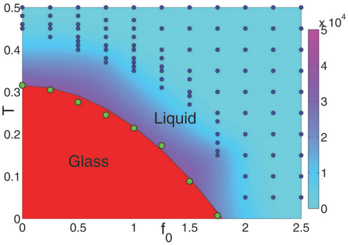

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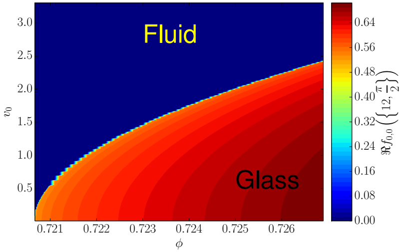
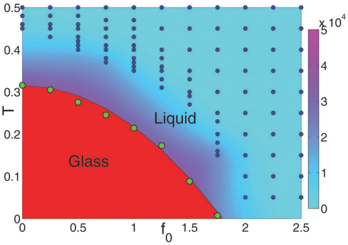
Glass Transition of Active Brownian Disks

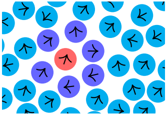




[Mandal *et al.*, Soft Matter (2016)]

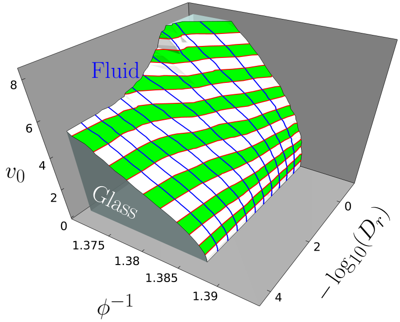
- activity softens, then melts the glass
- qualitative agreement with simulations of 3D ABP

physics: cage effect + fluidization by activity

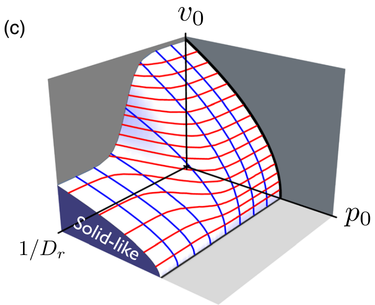


[Liluashvili, Ónody, ThV, Phys Rev E **96**, 062608 (2017)]

Rotational Diffusion changes Glass Transition



mode-coupling theory



simulations of cell-layer model

[Bi *et al.*, PRX (2016)]

- iso-kinetic lines *not functions of single* $T_{\text{eff}} \sim v_0^2/D_r$
- competition of length scales: $\ell_p = v_0/D_r$ and cage size $\ell_c \approx 0.1a$

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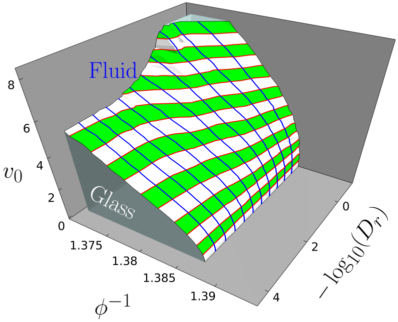

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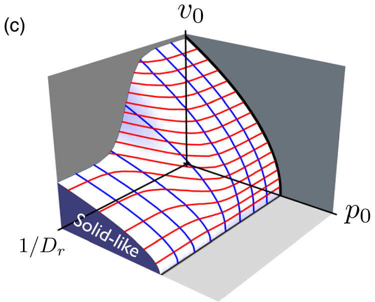

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Active *vs.* Passive Glass

passive-particle glass:

transition & structure *independent* on details of dynamics

"metastable minimum in free-energy landscape"

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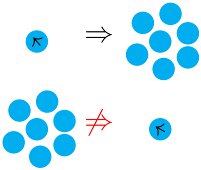
passive-particle glass:
transition & structure *independent* on details of dynamics
"metastable minimum in free-energy landscape"

active-particle glass:
transition depends on full dynamical history
dynamical balance



Nonlinear Response Theory

Nonlinear Response Theory

aim: calculate arbitrary non-eq. averages of observables

nonequilibrium phase-space density $\rho(\Gamma, t)$:

$$\partial_t \rho(\Gamma, t) = \left(\Omega_{\mathsf{eq}}(\Gamma) + \delta\Omega(\Gamma, t) \right) \rho(\Gamma, t)$$

[Brader, ThV, Cates, Fuchs, Phys Rev Lett (2007); Brader *et al.*, Phys Rev Lett (2008)]

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$$\boxed{\langle f \rangle(t) = \langle f \rangle_{\text{eq}} + \int_{-\infty}^{t} dt' \left\langle \left[\frac{\delta\Omega(t')\rho_{\text{eq}}}{\rho_{\text{eq}}}\right] \exp_- \left[\int_{t'}^{t} \Omega^\dagger(\tau)\, d\tau\right] f \right\rangle_{\text{eq}}}$$

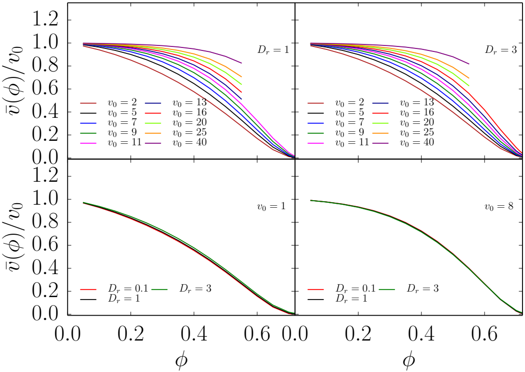
approximate integral using mode-coupling theory

[Brader, ThV, Cates, Fuchs, Phys Rev Lett (2007); Brader *et al.*, Phys Rev Lett (2008)]

Nonequilibrium Swim Velocity

coarse-grained swim speed $v(\rho)$: parameter for field theories

$$v(\rho) = v_0 \left[1 - \int_0^\infty dt \cdots \right]$$

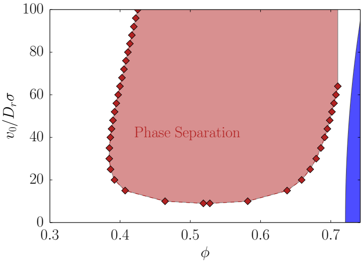


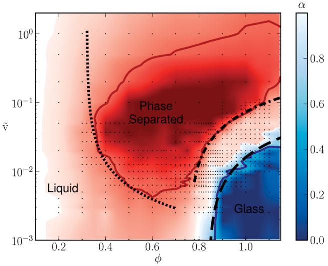
- density dependent $v(\rho)$ from fixed v_0 per particle
- increasing v_0, shape of $v(\rho)$ changes

[Liluashvili, ThV (in preparation, 2018)]

Motility Induced Phase Separation (MIPS) and Glass

$$\mathcal{F}_{\text{ex}} = \int d\vec{r}\, f_{\text{ex}}(\rho(\vec{r})) \qquad f_{\text{ex}}(\rho) = \int^{\rho} d\rho' \ln\left[\frac{v^2(\rho')}{2D_r} + D_t\right]$$





[MCT]

[Fily *et al.*, Soft Matter (2014)]

- microscopic model ⇒ input parameters for mesoscopic model
- systematic coarse graining possible

Conclusion and Outlook

Conclusion

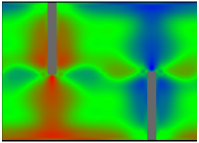
- high-density theory for active Brownian particles
  - *competition of length scales* $\ell_{\text{interaction}}, \ell_{\text{swim-persistence}}$
  - *no single "effective temperature"*
- combine microscopic theory with mesoscopic models
  - *systematic approach to calculate (coarse-grained) transport coefficients*

A. Liluashvili, J. Ónody, and Th. Voigtmann,
Physical Review E **96**, 062608 (2017)

Deutsche Forschungsgemeinschaft (DFG)
Special Priority Programme "Microswimmers"

John von Neumann Institute for Computing (NIC)
Project HKU26 @ JURECA

Multi-Scale Fluid Dynamics?



Multi-Scale Computational Fluid Mechanics

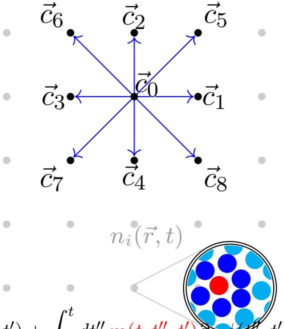
Navier Stokes

constitutive equation

microscopic dynamics

collision and streaming

effective interaction

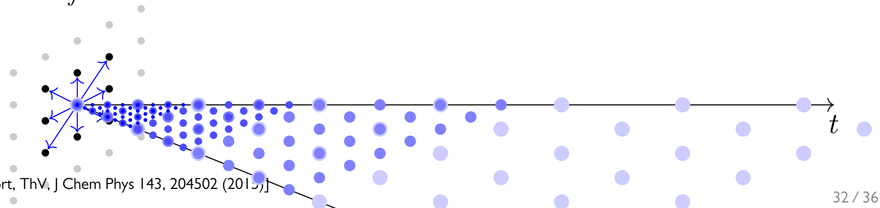


$$\varrho\left[\partial_t\vec{v}+(\vec{v}\cdot\vec{\nabla})\vec{v}\right]=-\vec{\nabla}p+\vec{\nabla}\cdot\boldsymbol{\sigma}$$

$$\boldsymbol{\sigma}(t)=\int^t dt'[-\partial_{t'}\boldsymbol{B}_{tt'}]\phi^2(t,t',[\boldsymbol{B}])$$

$$\tau_0\partial_t\phi(t,t')+\phi(t,t')+\int_{t'}^t dt''\,m(t,t'',t')\partial_{t''}\phi(t'',t')=0$$

$$m(t,t'',t')=h(\boldsymbol{B}_{tt''})h(\boldsymbol{B}_{tt'})\mathcal{F}[\phi(t,t'')]$$



[Papenkort, ThV, J Chem Phys 143, 204502 (2015)]

Multi-Scale Computational Fluid Mechanics

Navier Stokes

cluster ☹ booster ☺

constitutive equation

cluster ☺ booster ☹

microscopic dynamics

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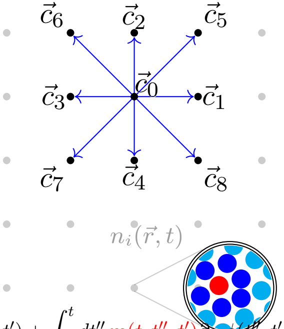

$$\varrho\left[\partial_t\vec v + (\vec v\cdot\vec\nabla)\vec v\right] = -\vec\nabla p + \vec\nabla\cdot\boldsymbol{\sigma}$$

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$$\tau_0\partial_t\phi(t,t') + \phi(t,t') + \int_{t'}^t dt''\, m(t,t'',t')\,\partial_{t''}\phi(t'',t') = 0$$

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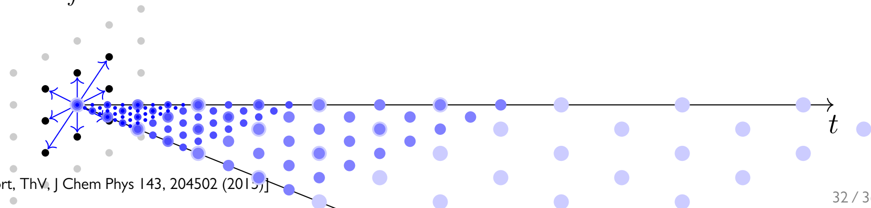


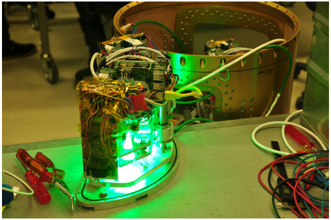
[Papenkort, ThV, J Chem Phys 143, 204502 (2015)]

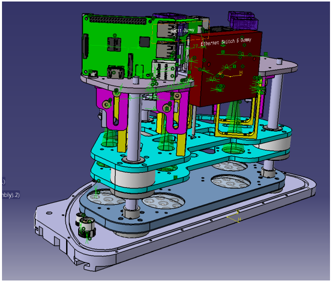
2D *vs.* 3D – Experiments?

- experiments on artificial microswimmers: restricted to quasi-2D (sedimentation!)
- *does MIPS depend on spatial dimensionality??*

$\Rightarrow$ experiments under *microgravity conditions*

RAMSES – RAndom motion of micro-swimmers Experiment in Space

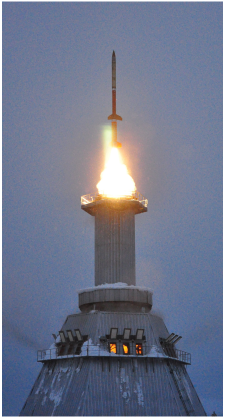




Materials Physics on a Sounding Rocket

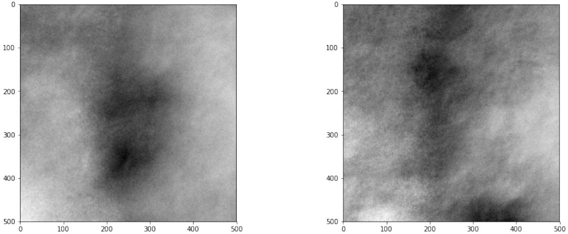
MAPHEUS – MAterialPHysik. Experimente Unter Schwerelosigkeit

- two-stage S31 / improved Malamute aka "Patriot"
- max. acceleration $20g$, apogee $\sim 250\,\mathrm{km}$
- $\sim 400\,\mathrm{kg}$ payload, $\sim 5\,\mathrm{min}$ µg time
- start from Esrange (Sweden) 2018-02-17



Materials Physics on a Sounding Rocket

Preliminary Data



- can observe ABP clustering in 3D
- occurs at much lower densities than expected (!?)

Thank you

- Alexander Liluashvili, Jonathan Ónody, Julian Reichert (theory)
- Raphael Keßler, Christoph Dreißigacker, Jörg Drescher (engineering)

- Celia Lozano, Clemens Bechinger (experiment; U Konstanz)
- Suvendu Mandal (simulation; U Innsbruck)