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Non-equilibrium dynamics of active Brownian particles (ABP) – a paradigm in soft matter/biological physics

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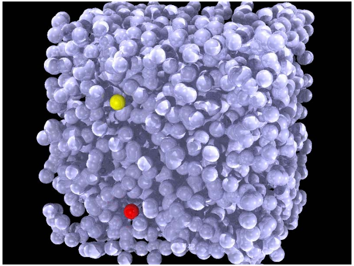
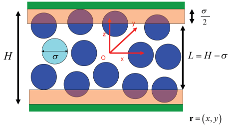
Further Interests: Beyond Brownian Motion

Cellular Crowding & Porous Media



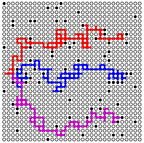
Mandal *et al* PRL (2019)
Spanner *et al* PRL (2016)
Schirmacher *et al* PRL (2015)
Höfling *et al* PRL (2007)
Höfling *et al* PRL (2006)
Rep. Prog. Phys. (2013)

Glass Transition



Mandal *et al* PRL (2017)
Mandal *et al* Nature Comm. (2014)
Lang *et al* PRL (2010)
Franosch *et al* PRL (2012)

Driven Transport

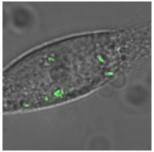


Leitmann *et al* PRF (2018)
Leitmann *et al* PRL (2017)
Leitmann *et al* PRL (2013)

Needles & Biofilaments

S. Mandal *et al* PRL (2020)
Leitmann *et al* PRL (2016)
Höfling *et al* PRL (2006), PRL (2008)

Intracellular Transport



Meier *et al* PNAS (2011)
Witzel *et al* Biophysical Journal (2019)

Discovery of Brownian Motion

Robert Brown

Scottish botanist

1773-1858

Clarkia pilchella

Observation of pollen in water

- micron-sized pollen 5µm
- single-lens light microscope
- agitated and erratic motion
- never to rest
- not connected to life !

wikipedia.org/wiki/Brownsche_Bewegung

earlier: Jan Ingenhousz 1785, coal dust on liquid surfaces

Lucretius: De rerum natura

Statistical Interpretation

Molecular kinetic interpretation

- collisions with solvent molecules, increments as independent
- Gaussian propagator

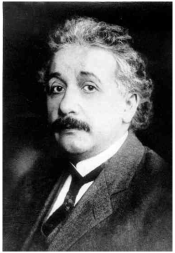
$$P(\mathbf{r}, t) = \frac{1}{(4\pi D t)^{3/2}} \exp\left(-\mathbf{r}^2/4Dt\right)$$

- mean-square displacement

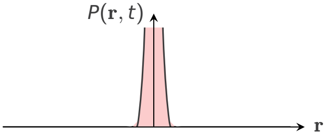
$$\langle [\mathbf{r}(t) - \mathbf{r}(0)]^2 \rangle = 6Dt$$

- solves diffusion equation

$$\partial_t P = D\nabla^2 P$$



Albert Einstein (1905)



statistical interpretation of diffusion

central limit theorem

same time: Marian Smoluchowski

Langevin description

Force balance $m\ddot{x}(t) = -\zeta\dot{x}(t) + f(t)$

deterministic friction force

random force

- Stokes drag $\zeta = 6\pi\eta a$
- short-time correlated noise

$$\langle f(t)f(t')\rangle = 2k_BT\zeta\delta(t-t')$$

- Power spectral density (PSD)

$$\lim_{\mathrm{T}\to\infty}\frac{1}{\mathrm{T}}\langle|\hat{f}_{\mathrm{T}}(\omega)|^2\rangle = 2k_{\mathrm{B}}T\zeta, \qquad \hat{f}_{\mathrm{T}}(\omega) := \int_{-\mathrm{T}/2}^{\mathrm{T}/2} f(t)\exp(\mathrm{i}\omega t)\mathrm{d}t$$

white noise

Paul Langevin

Leonard S. Ornstein

theoretical framework by Ornstein

Fluctuation-Dissipation Theorem

- momentum relaxation time $\tau_p = m/\zeta$
- long times: friction dominates $\zeta \dot{x}(t) \approx f(t) \rightarrow$
  $x(t) - x(0) = \zeta^{-1} \int_0^t f(t') \mathrm{d}t'$
- mean-square displacement

$$\langle [x(t) - x(0)]^2 \rangle = 2 \, k_\mathrm{B} T \zeta^{-1} \, t$$

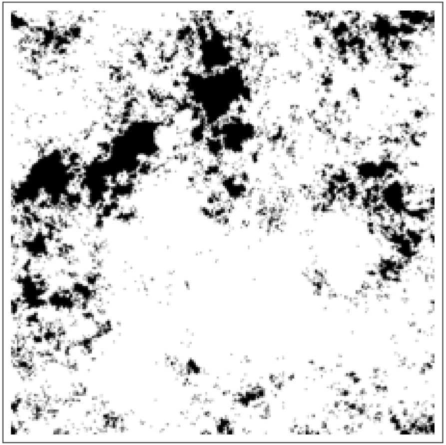
diffusion coefficient D

Einstein relation:
thermal scale $k_B T$ connects two worlds

$$D = k_\mathrm{B}T / \zeta$$

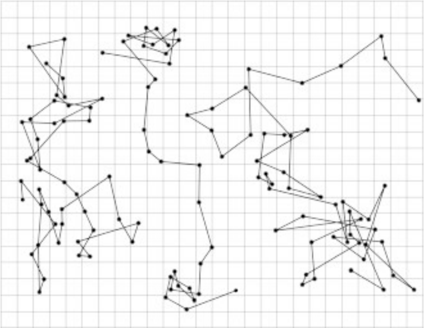
fluctuations friction

Random walk

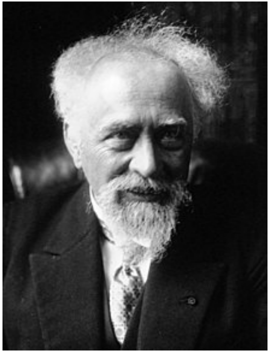


wikipedia.org/wiki/Brownian_motion

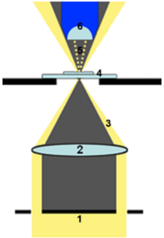
First Experimental Observation



tracings of colloidal particles (1909)



Jean-Baptiste Perrin



dark field (1903)

Nobel Prize 1926:

Physical reality of molecules
Determination of Avogadro constant

Optical trapping

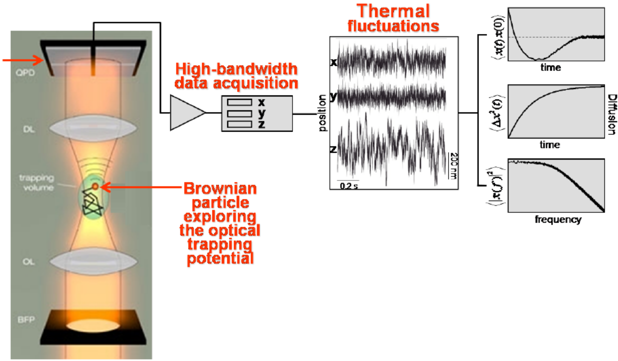
- optical trapping interferometry
  Nobel Prize 2018: Arthur Ashkin
- temporal resolution 1µs
- subnanometer spatial resolution
- trapping forces 1 fN-1pN

Ultrasensitive biophysical tool

- local reporter
- viscoelastic properties

hydrodynamics important

Sylvia Jeney (EPFL)



Harmonic Trapping

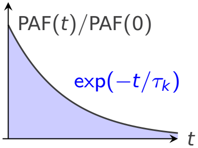
- harmonic restoring forces $F(t) = -kx(t)$
- overdamped limit, friction dominated
- Force balance

$$\zeta \dot{x}(t) + kx(t) = f(t)$$

friction force — optical trap — random force

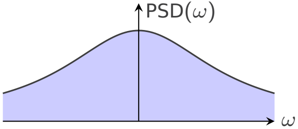
- trap relaxation time $\tau_k \equiv \zeta / k$
- uncorrelated $\langle f(t) x(0) \rangle = 0$ for $t > 0$
- positional autocorrelation function PAF

$$\langle x(t)x(0) \rangle = \frac{k_B T}{k} \exp(-t/\tau_k)$$



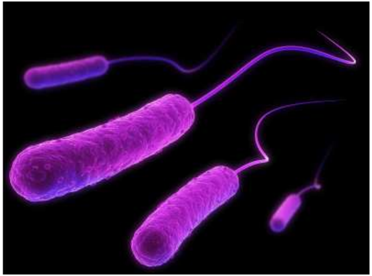
positional autocorrelation

power spectral density
$\mathrm{PSD}(\omega) = \frac{1}{T} \langle |\hat{x}_T(\omega)|^2 \rangle$

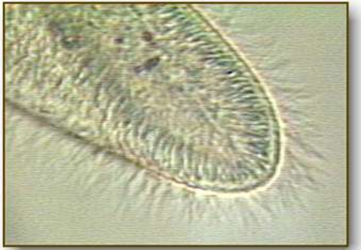


Lorentzian
$\mathrm{PSD}(\omega) = \frac{2k_B T/\zeta}{1+\omega^2 \tau_k^2}$

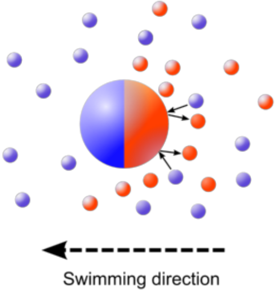
Self-propelled agents



Bacteria using flagella to swim



Paramecium uses cilia



self-propelled Janus particle

- Swimming mechanism by shape deformations or induced gradients in the fluid
- intrinsically far from equilibrium
- recent experimental progress to build artificial self-propelled particles
- plethora of collective phenomena (flocking, swarms, phase separation, trapping,...)
- mostly simulational studies
- lacking: complete characterization of single particle motion

Self-propelled particles

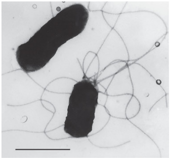
biological microswimmers

- self-propulsion by e.g. flagella, cilia

artificial active agents

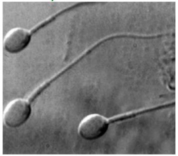
- biomimetic mechanisms
- phoretic motion

E. Coli



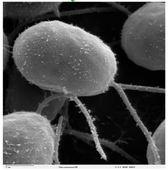
H. Eisenbach, eLS (2001)

Spermatozoa

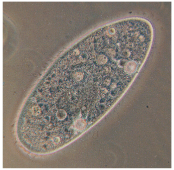


L.D. Akl et al., RBE (2011)

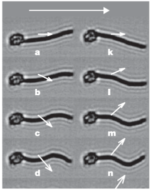
Chlamydomonas



Paramecium

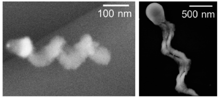


artificial flagellum



R. Dreyfus et al., Nature **437** (2005)

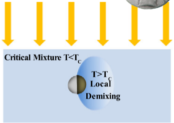
actuated by magnetic field



D. Schamel et al., ACSNano **8** (2014)

Janus particles

T.-C. Lee et al., Nano Lett. **14** (2014)

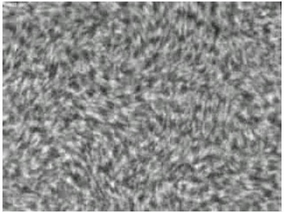


I. Buttinoni et al., JP Condens. Mat. **24** (2012)

Dynamical behavior of active agents

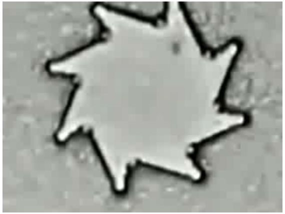
Collective phenomena

’bacterial’ turbulence



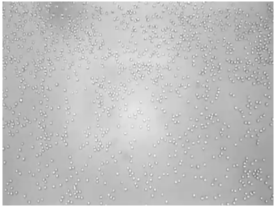
H. Berg’s lab

bacteria ratchet



R. di Leonardo et al., PNAS **21** (2010)

phase separation



J. Palacci et al., Science **339** (2013)

Warm-up: an unrealistic toy model

Active Ornstein-Uhlenbeck particle (AOU)

- activity is mimicked by colored noise

$$\dot{x}(t) = \mu F(x(t)) + \eta(t) + \eta_a(t)$$

deterministic drift — white noise — colored noise

independent centered Gaussian noises $\eta(t)$, $\eta_a(t)$ with

$$\langle \eta(t)\eta(t')\rangle = 2D_0\delta(t-t'), \qquad \langle \eta_a(t)\eta_a(t')\rangle = \frac{D_a}{\tau} e^{-|t-t'|/\tau}$$

correlation time τ, characteristic length $L = \sqrt{2D_a\tau}$
dimensionless Péclet number $\mathrm{Pe} := D_a/D_0$

- drift due to forces F, mobility μ
- colored noise turns it into an non-equilibrium process

AOU particle

Solution for free AOU process

- free particle, no force $F = 0$

$$\text{increment} \quad \Delta x(t) := x(t) - x(0) = \int_0^t [\eta(t') + \eta_a(t')]\mathrm{d}t'$$

→ increment $\Delta x(t)$ is a centered **Gaussian** variable, $\langle \Delta x(t) \rangle = 0$

- Mean-square displacement $\langle |\Delta \mathbf{r}(t)|^2 \rangle = \langle \Delta x(t)^2 \rangle + \langle \Delta y(t)^2 \rangle$

$$\langle \Delta x(t)^2 \rangle = \int_0^t \mathrm{d}t' \int_0^t \mathrm{d}t'' [\langle \eta(t')\eta(t'') \rangle + \langle \eta_a(t')\eta_a(t'') \rangle] = \int_0^t \mathrm{d}t' \int_0^t \mathrm{d}t'' [2D_0 \delta(t' - t'') + \frac{D_a}{\tau} e^{-|t'-t''|/\tau}]$$

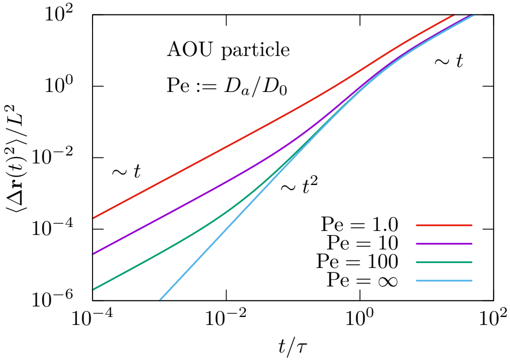
same calculation for $\langle \Delta y(t)^2 \rangle$,

$$\langle |\Delta \mathbf{r}(t)|^2 \rangle = 4D_0 t + 4D_a \left[t + \tau(e^{-t/\tau} - 1)\right] \quad \xrightarrow{t\to\infty} \quad 4D_{\mathrm{eff}} t$$

AOU particle

Mean-square displacement

- **effective** (long-time) diffusion coefficient
  $D_{\text{eff}} = D_0 + D_a$
  crossover from short-time diffusion $2D_0t$ to long-time diffustion $2D_at$ at crossover time τ
- characteristic length of active propulsion $L = \sqrt{2D_a\tau}$
- persistent motion $\sim t^2$ at intermediate times $t \lesssim \tau$
- passive diffusion dominates at short times $t \ll \tau/\text{Pe}$
- functional form identical to ballistic particle and ....



Propagator

- **propagator**

$$\mathbb{P}(\mathbf{r},t) := \langle \delta(\mathbf{r} - \Delta\mathbf{r}(t)) \rangle$$

$\widehat{=}$ pdf for **displacement** $\mathbf{r}$ in **lag time** t

- **Gaussian propagator**

$$\mathbb{P}(\mathbf{r},t) = \frac{1}{\pi \langle |\Delta\mathbf{r}(t)|^2 \rangle} \exp\left(\frac{-\mathbf{r}^2}{\langle |\Delta\mathbf{r}(t)|^2 \rangle} \right)$$

- higher cumulants vanish
e.g. **non-Gaussian parameter** (2D)

$$\alpha_2(t) := \frac{\langle |\Delta\mathbf{r}(t)|^4 \rangle}{2 \langle |\Delta\mathbf{r}(t)|^2 \rangle^2} - 1 = 0$$

AOU particle

Intermediate scattering function

- **intermediate scattering function**

$$F(k,t) := \langle \exp(-i\mathbf{k}\cdot\Delta\mathbf{r}(t))\rangle = \int \mathrm{d}\mathbf{r}\,\exp(-i\mathbf{k}\cdot\mathbf{r})\mathbb{P}(\mathbf{r},t)$$

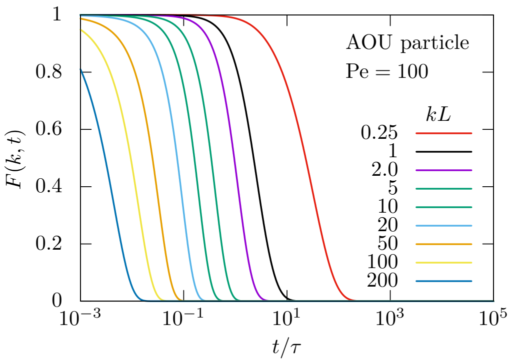
$\mathbf{k}$ wave vector, $\triangleq$ characteristic function of displacement

- isotropic systems → average over directions of $\mathbf{k}$

$$F(k,t) = \langle J_0(k|\Delta\mathbf{r}(t)|)\rangle \qquad \text{in } 2D$$

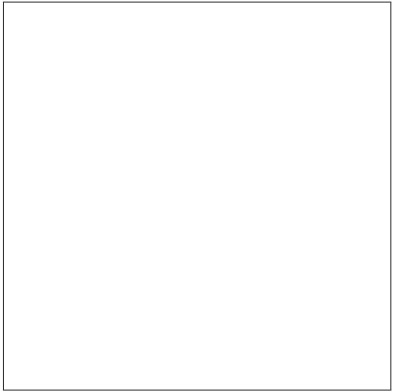
$$= 1 - \frac{k^2}{4}\langle|\Delta\mathbf{r}(t)|^2\rangle + \frac{k^4}{64}\langle|\Delta\mathbf{r}(t)|^4\rangle + O(k^6)$$

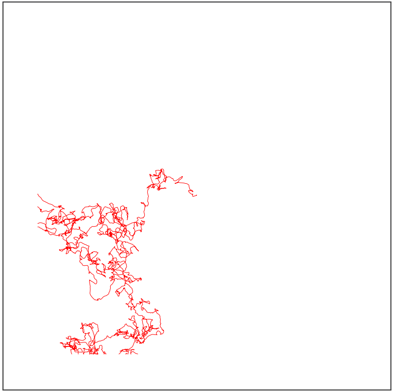
generates low-order moments



Gaussian model (2D) $F(k,t) = \exp(-k^2\langle|\Delta\mathbf{r}(t)|^2\rangle/4)$

Trajectories



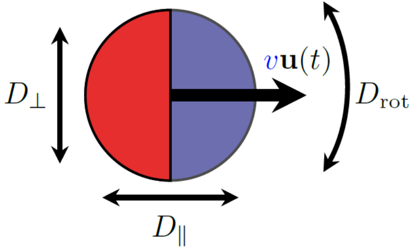


Random walker

Active Ornstein-Uhlenbeck particle

Model set-up

Active Brownian Particle (ABP)



- Active propulsion with constant velocity v along the long axis $\mathbf{u}$, $|\mathbf{u}| = 1$
- Rotational diffusion D_{rot}
- Anisotropic translational diffusion $D_{||}, D_\perp$
- ignores microscopic origin of propulsion, effective description
- simplistic model encoding persistent random walk
  persistence time $\tau_{\text{rot}} = 1/D_{\text{rot}}$
  persistence length $L = v/D_{\text{rot}}$

Active Brownian particle

Propagator

Trajectory

Stochastic equations

Active Brownian particle (2D)

$$\frac{\mathrm{d}}{\mathrm{d}t}\vartheta(t) = \zeta(t)$$

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{r}(t) = v\,\mathbf{u}(t) + \boldsymbol{\eta}(t) = v\begin{pmatrix}\cos\vartheta(t)\\ \sin\vartheta(t)\end{pmatrix} + \boldsymbol{\eta}(t)$$

position — fixed velocity — orientation

$$\langle\zeta(t)\zeta(t')\rangle = 2D_{\mathrm{rot}}\delta(t-t'), \quad \langle\eta_i(t)\eta_j(t')\rangle = [2D_\perp\delta_{ij} + 2(D_\| - D_\perp)u_i(t)u_j(t)]\delta(t-t')$$

independent Gaussian white noise

- translational anisotropy $\Delta D = D_\| - D_\perp$
  mean diffusion coefficient $\bar{D} = (D_\| + D_\perp)/2$
- characteristic length $a = \sqrt{3\bar{D}/D_{\mathrm{rot}}}/2$ replaces radius of the particle
- dimensionless parameters

anisotropy $\Delta D/\bar{D}$ Péclet number $\mathrm{Pe} = va/\bar{D}$

ABP – angular motion

angular diffusion

- conditional probability $\mathbb{P}(\vartheta, t|\vartheta_0), t > 0$

$$\partial_t \mathbb{P} = D_{\rm rot} \partial_\vartheta^2 \mathbb{P}$$

- 2π-periodic solution: **wrapped normal distribution**

$$\mathbb{P}(\vartheta, t|\vartheta_0) = \frac{1}{\sqrt{4\pi D_{\rm rot} t}} \sum_{n=-\infty}^{\infty} \exp\left\{ \frac{-(\vartheta - \vartheta_0 + 2\pi n)^2}{4 D_{\rm rot} t} \right\} = \sum_{\nu=-\infty}^{\infty} e^{-\nu^2 D_{\rm rot} t} e^{i\nu(\vartheta - \vartheta_0)}$$

- angular correlation functions, $\nu, \mu \in \mathbb{Z}, t > 0$

$$\langle e^{i\nu\vartheta(t)} e^{-i\mu\vartheta(0)} \rangle = \int_0^{2\pi} \mathrm{d}\vartheta \int_0^{2\pi} \frac{\mathrm{d}\vartheta_0}{2\pi} \; e^{i\nu\vartheta} e^{-i\mu\vartheta_0} \mathbb{P}(\vartheta t|\vartheta_0)$$

$$\langle e^{i\nu\vartheta(t)} e^{-i\mu\vartheta(0)} \rangle = \delta_{\mu\nu} \exp(-\nu^2 D_{\rm rot} t)$$

ABP - mean-square displacement

pedestrian calculation

- increment

$$\Delta x(t) := x(t) - x(0) = \int_0^t [v \cos\vartheta(t') + \eta_x(t')]\mathrm{d}t' \qquad \Delta y(t) := y(t) - y(0) = \int_0^t [v \sin\vartheta(t') + \eta_y(t')]\mathrm{d}t'$$

- mean-square displacement

$$\langle \Delta x(t)^2 \rangle + \langle \Delta y(t)^2 \rangle = \int_0^t \mathrm{d}t' \int_0^t \mathrm{d}t'' [v^2 \underbrace{\langle \cos\vartheta(t')\cos\vartheta(t'') + \sin\vartheta(t')\sin\vartheta(t'')\rangle}_{\langle \cos[\vartheta(t') - \vartheta(t'')]\rangle = \exp(-D_{\mathrm{rot}}|t'-t''|)} + \underbrace{\langle \eta_x(t')\eta_x(t'') + \eta_y(t')\eta_y(t'')\rangle}_{4\bar{D}\delta(t'-t'')}]$$

$$\langle |\Delta \mathbf{r}(t)|^2 \rangle = 4\bar{D}t + 2v^2\tau_{\mathrm{rot}}\left[t + \tau_{\mathrm{rot}}(e^{-t/\tau_{\mathrm{rot}}} - 1)\right] \quad \xrightarrow{t\to\infty} \quad 4D_{\mathrm{eff}}t$$

rotational time $\tau_{\mathrm{rot}} = 1/D_{\mathrm{rot}}$, **effective diffusion coefficient** $D_{\mathrm{eff}} = \bar{D} + v^2\tau_{\mathrm{rot}}/2$ ✓
same functional form as for AOU particle/underdamped Langevin equation

- higher moments $\langle |\Delta \mathbf{r}(t)|^n \rangle$ become tedious

Fokker-Planck equation

conditional probability density $\mathbb{P}(\mathbf{r}, \vartheta, t|\vartheta_0)$ (Green function)

Perrin equation (Markov process)

$$\partial_t \mathbb{P} = D_{\text{rot}} \partial_\vartheta^2 \mathbb{P} - v\mathbf{u} \cdot (\partial_\mathbf{r} \mathbb{P}) + \partial_\mathbf{r} \cdot \left[D_{\|} (\partial_\mathbf{r} \mathbb{P}) - \Delta D (\mathbb{I} - \mathbf{u}\mathbf{u}) \cdot (\partial_\mathbf{r} \mathbb{P}) \right]$$

orientational diffusion   active propulsion   anisotropic diffusion $\Delta D = D_{\|} - D_{\perp}$

- Fokker-Planck equation for non-equilibrium dynamics
- coupling between orientation, active propulsion, and translation
- spatial Fourier transform $\tilde{\mathbb{P}}(\mathbf{k}, \vartheta, t|\vartheta_0) = \int \mathrm{d}^2 r \exp(-i\mathbf{k} \cdot \mathbf{r}) \mathbb{P}(\mathbf{r}, \vartheta, t|\vartheta_0)$

$$\partial_t \tilde{\mathbb{P}} = D_{\text{rot}} \partial_\vartheta^2 \tilde{\mathbb{P}} - iv\mathbf{u} \cdot \mathbf{k} \tilde{\mathbb{P}} - [D_\perp k^2 + \Delta D (\mathbf{u} \cdot \mathbf{k})^2] \tilde{\mathbb{P}}$$

  reminiscent of quantum pendudulum

- solution for intermediate scattering function

$$F(\mathbf{k}, t) = \langle \exp\left(-i\mathbf{k} \cdot \Delta \mathbf{r}(t)\right) \rangle = \int_0^{2\pi} \mathrm{d}\vartheta \int_0^{2\pi} \frac{\mathrm{d}\vartheta_0}{2\pi} \, \tilde{\mathbb{P}}(\mathbf{k}, \vartheta, t|\vartheta_0)$$

Christina Kurzthaler *et al*, Soft Matter (2017)

Mathieu functions

Separation ansatz

- choose coordinates $\mathbf{k}$ in x-direction

$$\partial_t \tilde{\mathbb{P}} = \left[D_{\text{rot}} \partial_\vartheta^2 - i v k \cos\vartheta - \left(D_\perp k^2 + \Delta D \, k^2 \cos^2\vartheta \right) \right] \tilde{\mathbb{P}}$$

$$= \left[D_{\text{rot}} \partial_\vartheta^2 - i v k \cos\vartheta - \left(\bar{D} k^2 + \frac{\Delta D}{2} k^2 \cos(2\vartheta) \right) \right] \tilde{\mathbb{P}}$$

- separation ansatz $\exp(-\lambda t) z(\vartheta)$

eigenvalue problem $$\left(\frac{\mathrm{d}^2}{\mathrm{d}\vartheta^2} - \frac{i v k}{D_{\text{rot}}} \cos\vartheta - \frac{\Delta D \, k^2}{2 D_{\text{rot}}} \cos(2\vartheta) - \frac{\bar{D} k^2}{D_{\text{rot}}} + \frac{\lambda}{D_{\text{rot}}} \right) z(\vartheta) = 0$$

- ignore self-propulsion for the moment $v = 0$

Mathieu equation $$\left[\frac{\mathrm{d}^2}{\mathrm{d}x^2} + \left(a - 2q \cos(2x) \right) \right] z(x) = 0$$

deformation parameter $q = k^2 \Delta D / 4 D_{\text{rot}}$, **eigenvalue** $a = (\lambda - \bar{D} k^2) / D_{\text{rot}}$

Mathieu functions

Summary of properties

- eigenfunctions called **Mathieu functions** are deformed cosine and sine functions, 2π-periodic (real q)

$$\begin{array}{lll} \text{even } \mathrm{ce}_n(q,x) & \text{eigenvalue } a_n(q) & n = 0,1,2,\dots \\ \text{odd } \ \mathrm{se}_n(q,x) & \text{eigenvalue } b_n(q) & n = 1,2,\dots \end{array}$$

  ascending eigenvalues $a_0(q) < b_1(q) < a_1(q) < b_2(q) < a_2(q)$ for $q > 0$
- symmetries

Eigenvalues	Eigenfunctions	Periodicity	Parity
$a_{2n}(q)$	$\mathrm{ce}_{2n}(q,x)$	Period π	Even
$a_{2n+1}(q)$	$\mathrm{ce}_{2n+1}(q,x)$	Antiperiod π	Even
$b_{2n+1}(q)$	$\mathrm{se}_{2n+1}(q,x)$	Antiperiod π	Odd
$b_{2n+2}(q)$	$\mathrm{se}_{2n+2}(q,x)$	Period π	Odd

- without deformation $q = 0$

$$\begin{array}{lll} \mathrm{ce}_0(0,x) = 1/\sqrt{2} & a_0(0) = 0 & \\ \mathrm{ce}_n(0,x) = \cos(nx) & a_n(0) = n^2 & n = 1,2,\dots \\ \mathrm{se}_n(0,x) = \sin(nx) & b_n(0) = n^2 & n = 1,2,\dots \end{array}$$

Mathieu functions – con't

- Fourier series

$$\mathrm{ce}_{2n}(q,x) = \sum_{m=0}^{\infty} A_{2m}^{2n}(q) \cos\, 2mx \qquad \mathrm{ce}_{2n+1}(q,x) = \sum_{m=0}^{\infty} A_{2m+1}^{2n+1}(q) \cos\,(2m+1)x$$

$$\mathrm{se}_{2n+1}(q,x) = \sum_{m=0}^{\infty} B_{2m+1}^{2n+1}(q) \sin\,(2m+1)x \qquad \mathrm{se}_{2n+2}(q,x) = \sum_{m=0}^{\infty} B_{2m+2}^{2n+2}(q) \sin\,(2m+2)x$$

- **orthogonality and normalization**

$$\int_0^{2\pi} \mathrm{ce}_m(q,x)\mathrm{ce}_n(q,x)\mathrm{d}x = \pi\delta_{mn} \qquad n,m = 0,1,2,\ldots, \int_0^{2\pi} \mathrm{se}_m(q,x)\mathrm{se}_n(q,x)\mathrm{d}x = \pi\delta_{mn} \qquad n,m = 1,2,\ldots$$

$$\int_0^{2\pi} \mathrm{se}_m(q,x)\mathrm{ce}_n(q,x)\mathrm{d}x = 0$$

- **completeness**

2π-periodic $\qquad f(x) = \alpha_0 \mathrm{ce}_0(q,x) + \sum_{n=1}^{\infty} [\alpha_n \mathrm{ce}_n(q,x) + \beta_n \mathrm{se}_n(q,x)]$

Fourier coefficients $\qquad \alpha_n = \frac{1}{\pi}\int_0^{2\pi} f(x)\, \mathrm{ce}_n(q,x)\, \mathrm{d}x \qquad \beta_n = \frac{1}{\pi}\int_0^{2\pi} f(x)\, \mathrm{se}_n(q,x)\, \mathrm{d}x$

Passive Anisotropic Diffusion

- Solution Fokker-Planck equation (define $\mathrm{se}_0(q,\vartheta)=0$)

$$\tilde{\mathbb{P}}(k,\vartheta,t|\vartheta_0)=\frac{e^{-\bar{D}k^2t}}{\pi}\sum_{n=0}^{\infty}\left[\mathrm{ce}_n(\vartheta)\mathrm{ce}_n(\vartheta_0)e^{-a_n(q)D_{\mathrm{rot}}t}+\mathrm{se}_n(\vartheta)\mathrm{se}_n(\vartheta_0)e^{-b_n(q)D_{\mathrm{rot}}t}\right]$$

- **intermediate scattering** function (ISF) after averaging and marginalizing

$$F(k,t)=2e^{-\bar{D}k^2t}\sum_{n=0}^{\infty}e^{-a_{2n}(q)D_{\mathrm{rot}}t}[A_0^{2n}(q)]^2$$

completely monotone functions

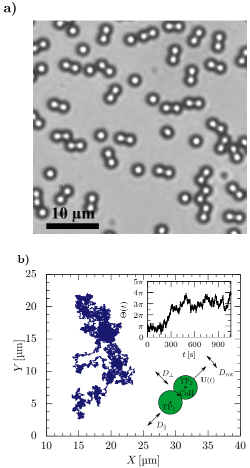
$$(-1)^l\frac{\mathrm{d}^l}{\mathrm{d}t^l}F(k,t)\geq 0 \qquad l=0,1,2,\ldots \qquad \Longleftrightarrow \qquad F(k,t)=\int_{[0,\infty)}e^{-\gamma t}\mathrm{d}m_k(t)$$

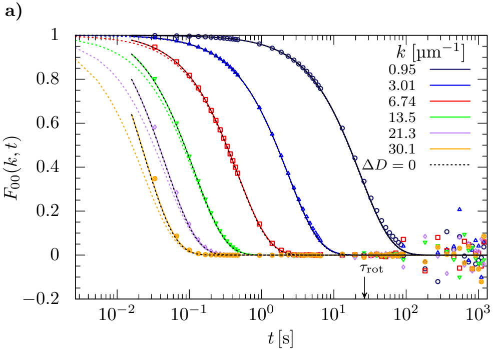
Bernstein's theorem

DB Mayer *et al*, PRE (2021)

Passive Anisotropic Diffusion

Experimental Results





- Small but **measurable** differences from pure exponential at larger k
- Larger differences for more generalized ISF

DB Mayer *et al*, PRE (2021)

Mathieu functions – ABP

Separation ansatz

- choose coordinates $\mathbf{k}$ in x-direction

$$\partial_t \tilde{\mathbb{P}} = D_{\text{rot}} \partial_\vartheta^2 \tilde{\mathbb{P}} - ivk \cos\vartheta \, \tilde{\mathbb{P}} - [D_\perp k^2 + \Delta D \, k^2 \cos^2 \vartheta] \tilde{\mathbb{P}}$$

- separation ansatz $\exp(-\lambda t) z(\vartheta)$

eigenvalue problem

$$\left(\frac{\mathrm{d}^2}{\mathrm{d}\vartheta^2} - \frac{ivk}{D_{\text{rot}}} \cos\vartheta - \frac{\Delta D \, k^2}{D_{\text{rot}}} \cos^2 - \vartheta \frac{D_\perp k^2}{D_{\text{rot}}} + \frac{\lambda}{D_{\text{rot}}} \right) z(\vartheta) = 0$$

- ignore **anisotropy** for the moment $D = D_\perp = D_\parallel$, $\Delta D = 0$, change of variable $x = \vartheta/2$

Mathieu equation

$$\left[\frac{\mathrm{d}^2}{\mathrm{d}\vartheta^2} + (a - 2q \cos(2x)) \right] z(x) = 0$$

imaginary deformation parameter $q = 2ivk/D_{\text{rot}} = 2ikL$, **eigenvalue** $a = 4(\lambda - k^2 D)/D_{\text{rot}}$

Isotropic self-propulsion

- Solution Fokker-Planck equation (define $\mathrm{se}_0(q,\vartheta)=0$)

$$\tilde{\mathbb{P}}(k,\vartheta,t|\vartheta_0)=\frac{e^{-k^2Dt}}{2\pi}\sum_{n=0}^{\infty}\left[\mathrm{ce}_{2n}(\vartheta/2)\mathrm{ce}_{2n}(\vartheta_0/2)e^{-a_{2n}(q)D_{\mathrm{rot}}t/4}+\mathrm{se}_{2n+2}(\vartheta/2)\mathrm{se}_{2n+2}(\vartheta_0/2)e^{-b_{2n+2}(q)D_{\mathrm{rot}}t/4}\right]$$

  2π-periodic → only even Mathieu functions appear

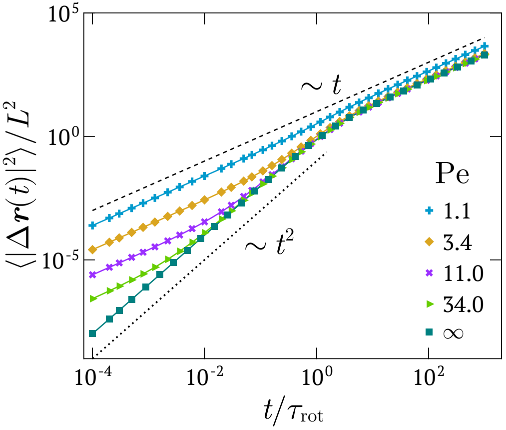
- **intermediate scattering** function (ISF) after averaging and marginalizing

$$F(k,t)=e^{-k^2Dt}\sum_{n=0}^{\infty}e^{-a_{2n}(q)D_{\mathrm{rot}}t/4}[A_0^{2n}(q)]^2$$

C Kurzthaler *et al*, PRL (2018)

Low-order moments (2D)

Mean-square displacement



- Expansion of ISF for **isotropic system**

$$F(k,t) = \langle \exp(-i\mathbf{k}\cdot\Delta\mathbf{r}(t))\rangle = \langle J_0(k|\Delta\mathbf{r}(t)|)\rangle$$

$$F(k,t) = 1 - \frac{k^2}{4}\langle|\Delta\mathbf{r}(t)|^2\rangle + \frac{k^4}{64}\langle|\Delta\mathbf{r}(t)|^4\rangle + \mathcal{O}(k^6)$$

- MSD initially translational diffusion dominates
  persistent swimming

$$\langle|\Delta\mathbf{r}(t)|^2\rangle = 4\bar{D}t + 2\frac{v^2}{D_{\rm rot}^2}\left[D_{\rm rot}t + \left(e^{-D_{\rm rot}t} - 1\right)\right]$$

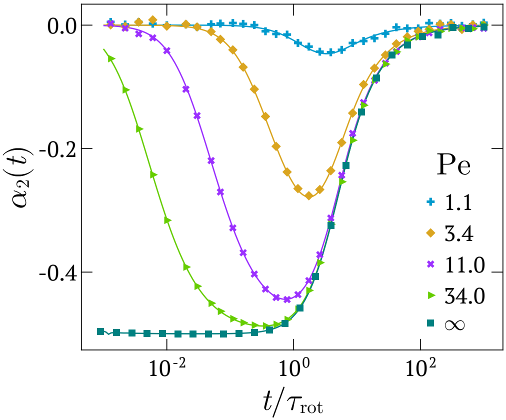
$$\xrightarrow{t\to\infty} 4D_{\rm eff}t$$

effective diffusion coefficient

$D_{\rm eff} = \bar{D} + v^2/2D_{\rm rot}$

Low-order moments (2D)

Non-Gaussian parameter

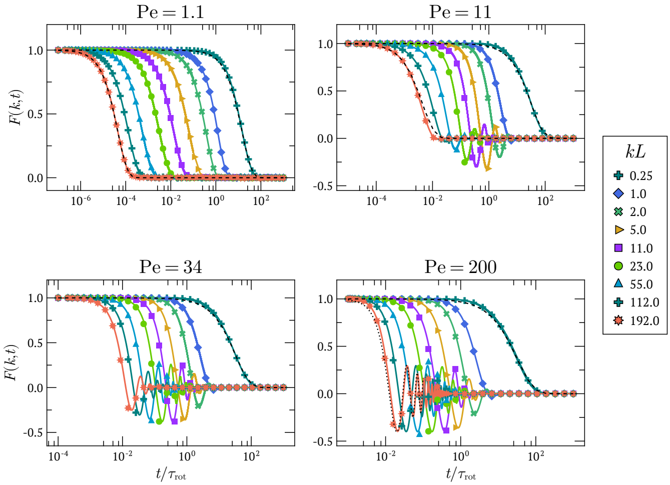


- Non-Gaussian parameter

$$\alpha_2(t) = \frac{\langle |\Delta \mathbf{r}(t)|^4 \rangle}{2\langle |\Delta \mathbf{r}(t)|^2 \rangle^2} - 1$$

- initially non-Gaussian by translational anisotropy characteristic minimum due to persistent swimming eventually again Gaussian

Intermediate scattering function



- characteristic oscillations emerge at intermediate wavenumbers
  fingerprint of persistent swimming

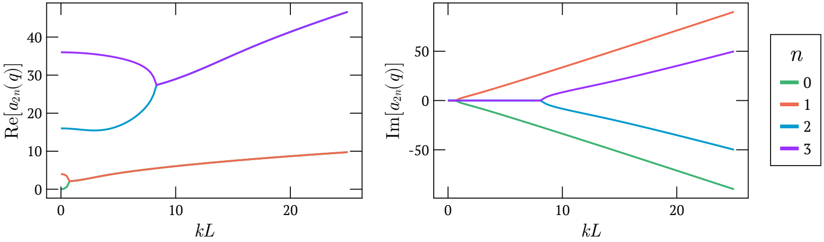
$$F(k,t) = \langle J_0(k|\Delta\mathbf{r}(t)|)\rangle$$

- large wavenumbers
  anisotropic translational diffusion
- small wavenumbers
  effective diffusion

$$D_{\text{eff}} = \bar{D} + v^2/2D_{\text{rot}}$$

figures courtesy Yanis Baouche

How can oscillations emerge?



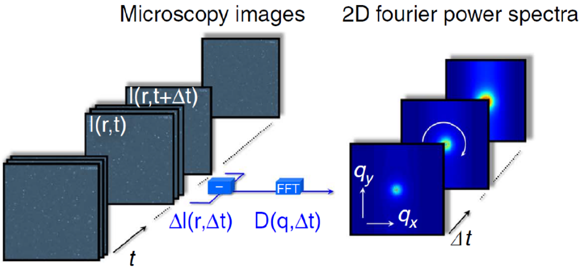
- Intermediate scattering function sum of relaxing exponentials?

$$F(k,t) = e^{-k^2 D t} \sum_{n=0}^{\infty} e^{-a_{2n}(q) D_{\mathrm{rot}} t/4} [A_0^{2n}(q)]^2$$

- Eigenvalue problem is non-Hermitian, eigenvalues become complex
  branching in the eigenvalues → no perturbation theory
  fingerprint of active motion

figures courtesy Yanis Baouche

Dynamic Differential Microscopy (DDM)



Sentjabrskaja *et al*, Nature. Comm. (2016)
Cerbino *et al*, PRL (2008)

- collect images by microscopy, dynamic contrast $\Delta I(\mathbf{r},t) = I(\mathbf{r},t+t_0) - I(\mathbf{r},t_0)$
- 2d Fourier transform $\Delta I(\mathbf{q},t)$ and correlate

$$D(\mathbf{q},t) = \langle |\Delta I(\mathbf{q},t)|^2 \rangle$$

- connects to ISF

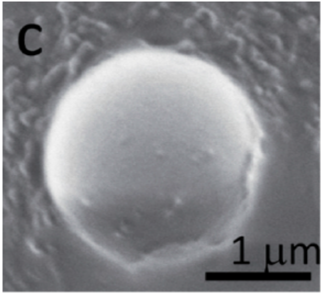
$$D(q,t) = A(q)\left[1 - F(q,t)\right] + B(q)$$

scattering properties — intermediate scattering function — camera noise

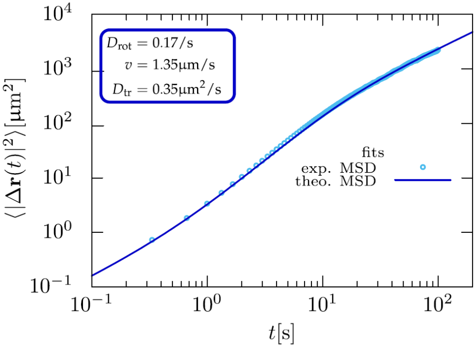
- complementary to single particle tracking (shorter times)

Comparison to Experiments

Janus particles



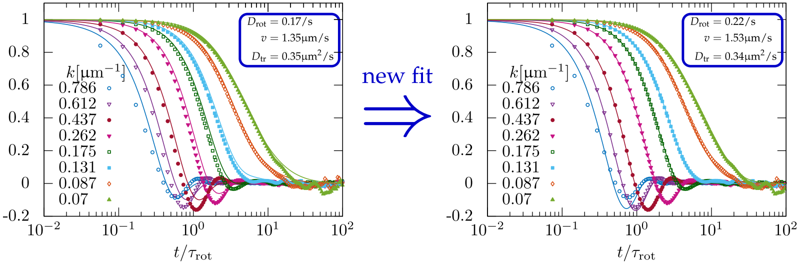
Brown & Poon, Soft Matter (2014)



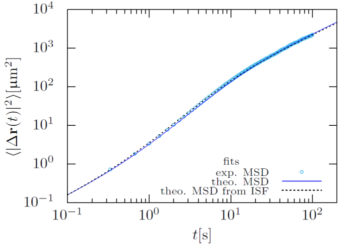
- Collaboration Poon, Martinez (Edinburgh)
- DDM experiments in Janus particles (Pt cover) in 2d
  particles swim to top plate

- so far: extract motility parameters from MSD

Comparison to experiments

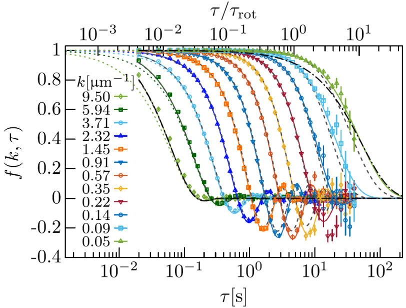


- first try: already good agreement with motility parameters extracted from MSD ✓
- but MSD does not provide spatio-temporal information
  fit not sensitive to all parameters
- use ISF to characterize motility → improved fit ✓

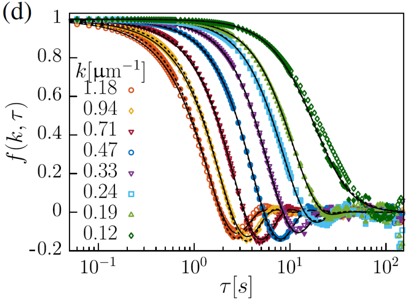


... but experimentalists can do better too

Single Particle Tracking



Differential Dynamic Microscopy



Agreement for complete spatio-temporal dynamics

Kurzthaler *et al* (PRL 2018)

Active Brownian particle model

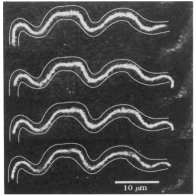
Why is this the model so good?

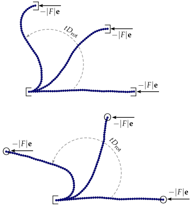
- ABP introduce a single new **mesoscopic** time scale τ_{rot} because reorientation is slow
  → mesoscopic persistence length $L = v\tau_{\text{rot}}$
- Does the magnitude of the velocity fluctuate? Yes, but on much shorter time scales
- Is the distance to the glass plate fixed? No, but fluctuations are much faster
- Is the local heating due to dissipation? Yes, small but solvent temperature quickly.
- What about depletion of fuel? Relevant, but on longer time scales

ABP is the correct coarse-grained
model on relevant time
and length scales

missing feature: **angular drift** for strongly anisotropic particles

Semiflexible polymer





F-actin solution

J. Käs *et al*, Nature (1994)

- persistence length $\ell_p = 2\kappa/k_B T$ (2d)
- Euler buckling force $F_c = \pi^2 \kappa / L^2$ (clamped rod)
- configurations look like trajectories of active Brownian particles

Kurzthaler *et al* PRE (2017)

Worm-like chain model

- idealize polymer configuration to space curve $\mathbf{r} = \mathbf{r}(s)$ s arc length
- Hamiltonian for bending and stretching local tangent $\mathbf{u} = \mathrm{d}\mathbf{r}/\mathrm{d}s$

$$\mathcal{H} = \int_0^L \mathrm{d}s \left[\frac{\kappa}{2} \left(\frac{\mathrm{d}\mathbf{u}(s)}{\mathrm{d}s} \right)^2 - \mathbf{F} \cdot \mathbf{u}(s) \right]$$

κ bending rigidity stetching force

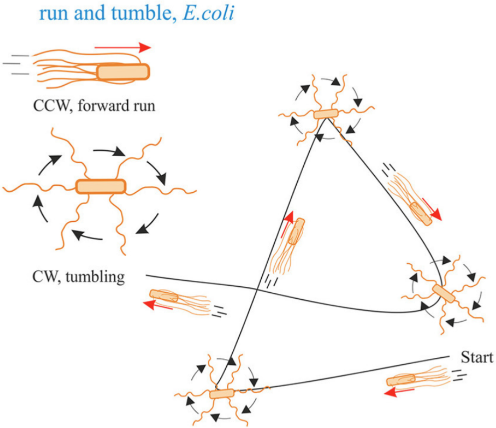
- partition sum path integral

$$Z(\mathbf{u}_L L, \mathbf{u}_0, 0) = \int_{\mathbf{u}(0)=\mathbf{u}_0}^{\mathbf{u}(L)=\mathbf{u}_L} \mathcal{D}[\mathbf{u}(s)] \exp(-\mathcal{H}/k_B T)$$

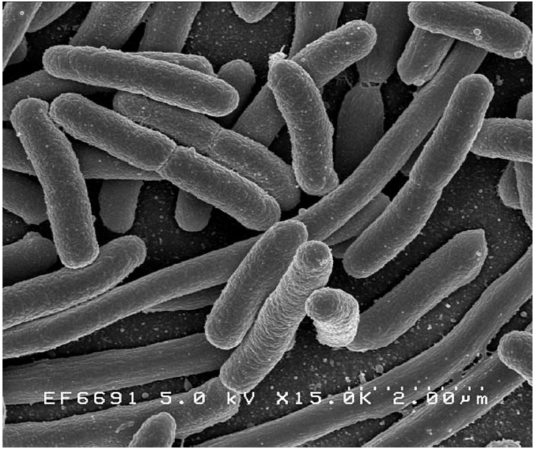
- path integral solved by Fokker-Planck equation

$$\partial_s Z(\mathbf{u}, s|\mathbf{u}_0 0) = \left[\frac{1}{\ell_p} \Delta_{\mathbf{u}} + \mathbf{F} \cdot \mathbf{u} \right] Z(\mathbf{u}, s|\mathbf{u}_0 0)$$

Run-and-tumble motion



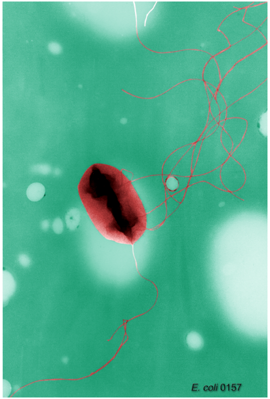
Julio, Bastos-Arrieta *et al.*, Frontiers in Robotics and AI vol. 5, 97 (2018)



Wikipedia

Run-and-tumble motion

Renewal process



Escherichia Coli bacterium

- Run-and-tumble motion
- Markov process for active motion
  propagator $\mathbb{P}(\mathbf{r}, t)$, e.g. persistent run, ABP,..
- random new orientation
  run-time distribution $T(t)$
- Probability for n-th renewal event $Q_n(\mathbf{r}, t)$

$$Q_{n+1}(\mathbf{r}, t) = \int_0^t \mathrm{d}t' \int_{\mathbb{R}^d} \mathrm{d}\mathbf{l}\; Q_n(\mathbf{r} - \mathbf{l}, t - t') T(t') \mathbb{P}(\mathbf{l}, t')$$

- Probability for last renewal event $Q(\mathbf{r}, t) = \sum_{n=1}^{\infty} Q_n(\mathbf{r}, t)$

$$Q(\mathbf{r}, t) - Q_1(\mathbf{r}, t) = \int_0^t \mathrm{d}t' \int_{\mathbb{R}^d} \mathrm{d}\mathbf{l}\; Q(\mathbf{r} - \mathbf{l}, t - t') T(t') \mathbb{P}(\mathbf{l}, t')$$

Renewal process -cont'd

- Probability $Q_1(\mathbf{r},t) = \mathbb{P}(\mathbf{r},t)T_1(t)$ for first renewal
  - Prob' to find interval of length t': $t'T(t')/\tau$
  - Prob' uniformly distributed within such intervals $1/t'$

$$T_1(t) = \int_t^\infty T(t')\mathrm{d}t'/\tau \qquad \tau = \int_0^\infty tT(t)\mathrm{d}t < \infty$$

  stationary process, mean run-time τ
- Prob' $P_n(\mathbf{r},t)$ for n renewal events up to time t:

$$P_n(\mathbf{r},t) = \int_0^t \mathrm{d}t' \int_{\mathbb{R}^d} \mathrm{d}\mathbf{l}\, Q_n(\mathbf{r}-\mathbf{l}, t-t')T_0(t')\mathbb{P}(\mathbf{l},t')$$

  Prob' run-time exceeds t: $T_0(t) = \int_t^\infty T(t')\mathrm{d}t'$
- stationary state propagator $P(\mathbf{r},t) = \sum_{n=0}^\infty P_n(\mathbf{r},t)$

$$P(\mathbf{r},t) - P_0(\mathbf{r},t) = \int_0^t \mathrm{d}t' \int_{\mathbb{R}^d} \mathrm{d}\mathbf{l}\, Q(\mathbf{r}-\mathbf{l}, t-t')T_0(t')\mathbb{P}(\mathbf{l},t')$$

- Prob' for no renewal event $P_0(\mathbf{r},t) = \mathbb{P}(\mathbf{r},t)\int_t^\infty (t'-t)T(t')\mathrm{d}t'/\tau$

Results

- Spatial Fourier transform $\mathbf{r} \mapsto \mathbf{k}$, summary of equations

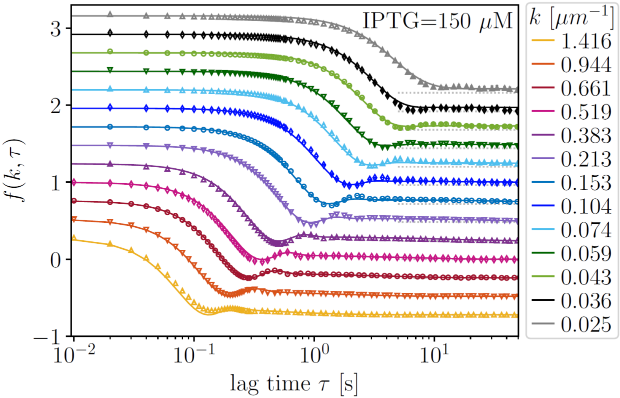
$$Q(\mathbf{k}, t) - Q_1(\mathbf{k}, t) = \int_0^t \mathrm{d}t' Q(\mathbf{k}, t - t') T(t') \mathbb{P}(\mathbf{k}, t')$$

$$P(\mathbf{k}, t) - P_0(\mathbf{k}, t) = \int_0^t \mathrm{d}t' Q(\mathbf{k}, t - t') T_0(t') \mathbb{P}(\mathbf{k}, t')$$

- Solve numerically directly as integral equation
  no Laplace transform needed, arbitrary waiting time distributions
- Upgraded to finite tumble times
  → Combined renewal event consisting of two steps
- account for non-motile cells → diffusion only
- exponentially distributed run τ_R and tumbling times τ_T
- post average over swimming velocity
  Schulz distribution $\propto v^Z \exp\left[-\frac{v}{\langle v \rangle}(Z+1)\right]$
  standard deviation $\sigma_v = \langle v \rangle / \sqrt{Z+1}$

Experimental results

DDM experiments on *E-coli*



C. Kurzthaler *et al*, arXiv:2212.11222

Résumé

Free active Brownian particle

- intermediate scattering functions for **active Brownian particle**
- mapping to the quantum rotor
- characteristic oscillations as **fingerprint** of active motion
- corroborated by experiments on Janus particles

E.-Coli bacteria

- intermediate scattering functions for **run-and-tumble particles**