GENERALIZED GREEN-KUBO RELATIONS FOR ACTIVE FLUIDS

[preprint arXiv:1907.02560]

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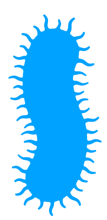
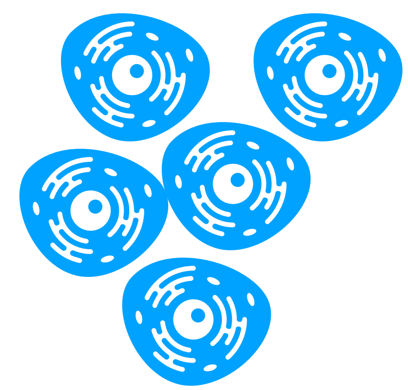
Physics of Active Matter - Statphys 2019

ACTIVE MATTER

Biology has been a long-standing inspiration for physicists…



Bacteria



Cell tissues

Flock of birds

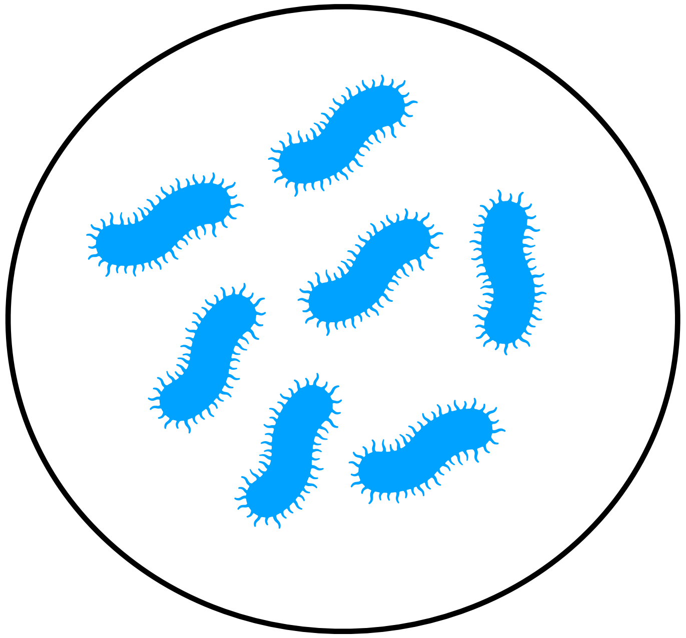
Local conversion of energy into motion **(self-propulsion)**

ACTIVE TRANSPORT

ACTIVE TRANSPORT

Bacterial ratchet motor

[R. Di Leonardo *et al* PNAS (2009)]



"Active bath"

II law of thermodynamics: we can't extract work from a passive bath

PROMISING NEW TRANSPORT PHENOMENA EMERGING FROM SELF-PROPULSION AND COLLECTIVE DYNAMICS

A FUNDAMENTAL CHALLENGE…

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Q: How are transport properties of a fluid affected by self-propulsion?

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Q: How are transport properties of a fluid affected by self-propulsion?

Q: Can we derive closed expressions for the transport coefficients of an active fluid?

SO FAR IN THE LITERATURE…

- **Introducing the concept of effective temperature**
  [LF Cugliandolo *et al*, Fluctuation and Noise Letters, 2019]

- **Taking equilibrium as the initial reference state**
  [A Sharma *et al*, J Chem Phys, 2016]

- **Response from weighted averages over unperturbed dynamics**
  [G Szamel, EPL, 2017]

INTEGRATION THROUGH TRANSIENTS

LINEAR RESPONSE

Green-Kubo relations: transport coefficients from equilibrium ensemble averages

$$\mathcal{T} = \frac{1}{k_B T} \int_0^\infty dt \langle \dot{A}(t) \dot{A}(0) \rangle_{eq}$$

e.g. **Mobility** $\mu \equiv \lim_{t \to \infty} \frac{\mathbf{v}}{\mathbf{f}_{ext}}$ $\mu = \frac{1}{k_B T} \int_0^\infty dt \langle \dot{x}(t) \dot{x}(0) \rangle_{eq}$

Diffusion coefficient

$$\boxed{\mu = \beta D}$$

Einstein relation

Need to generalize for active systems at their steady state!

INTEGRATION THROUGH TRANSIENTS

[M. Fuchs *et al* J. Phys.: Condens. Matter (2005)]

Generic evolution dynamics:

$$\frac{\partial \Psi}{\partial t} = \Omega \Psi = (\Omega_0 + \Omega_{ext})\Psi$$

Condition for steady state (here equilibrium): $\Omega_0 \Psi_0 = 0$

+

Operator identity $e^{\Omega t} = 1 + \int_0^t dt' e^{\Omega t} \Omega$

INTEGRATION THROUGH TRANSIENT

Ensemble average:

$$\langle A \rangle_t = \int d\Gamma A(\Gamma)\Psi(t)$$

Non linear response (ITT expression):

$$\langle A \rangle_t - \langle A \rangle_0 = \int_0^t dt' \langle \frac{\Omega_{ext}\Psi_0}{\Psi_0} A(t') \rangle_0$$

INTEGRATION THROUGH TRANSIENT

Ensemble average:

$$\langle A \rangle_t = \int d\Gamma A(\Gamma) \Psi(t)$$

Non linear response (ITT expression):

Conjugated observable

$$\langle A \rangle_t - \langle A \rangle_0 = \int_0^t dt' \langle \boxed{\frac{\Omega_{ext} \Psi_0}{\Psi_0}} \boxed{A(t') \rangle_0}$$

Evolved with the full operator: $e^{(\Omega_0 + \Omega_{ext})^\dagger} A_0$

INTEGRATION THROUGH TRANSIENT

For equilibrium systems:

Using Zero probability current solution: $\partial_i \Psi_0 = \beta \mathbf{F}_i \Psi_0$

$$\boxed{\Omega_0^\dagger = \bar{\Omega}_0^\dagger}$$

Footprint of equilibrium
DETAILED BALANCE

Time reversal operator

INTEGRATION THROUGH TRANSIENT

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DETAILED BALANCE

Time reversal operator

Together with Stationarity $\langle F_i^x(0) F_i^x(t) \rangle = \dfrac{1}{\mu_0^2} \left[k_B T \mu_0 \delta(t) - \langle \dot{x}_i(t) \dot{x}_i(0) \rangle_o \right]$

[R. Klein *et al* J. Phys A (1981)]

$$\mu_i = \beta \int_0^t dt' \langle \dot{x}_i(t') \dot{x}_i(0) \rangle_0 = \beta D$$

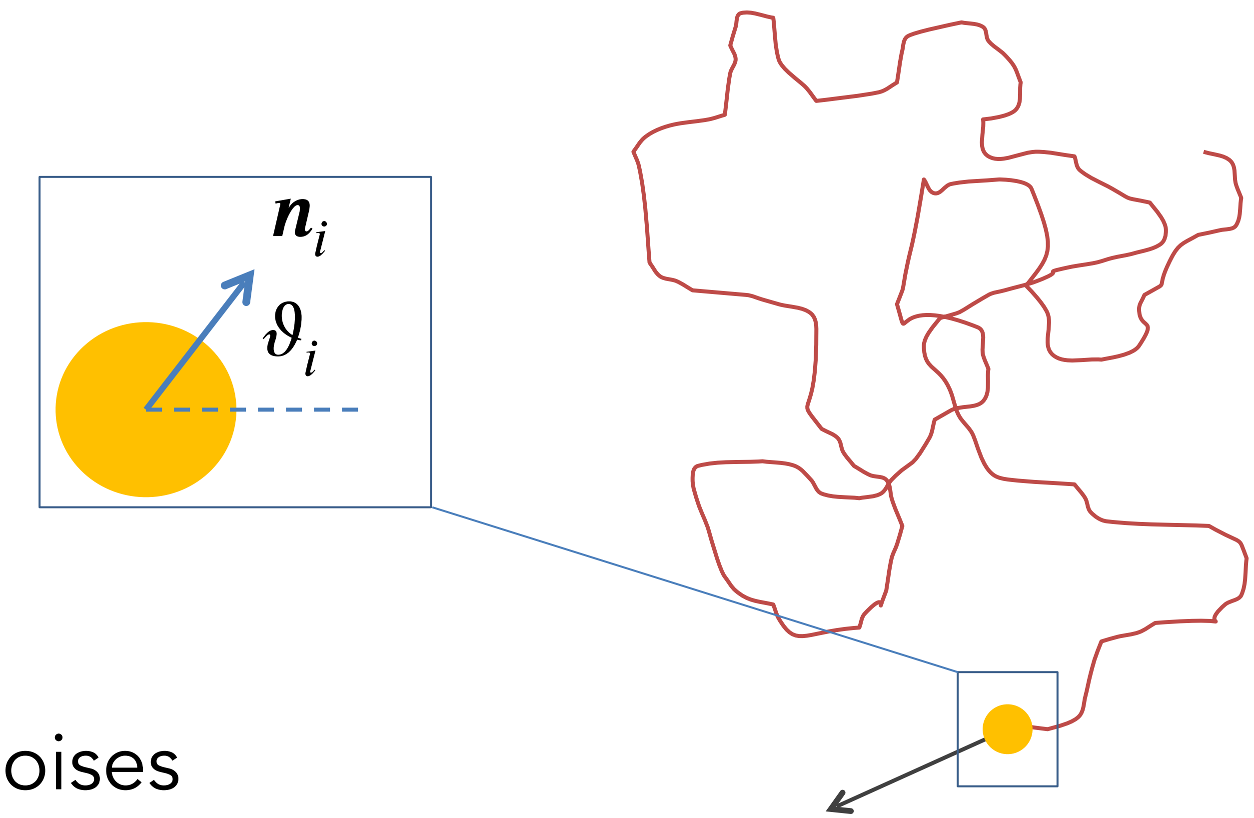
Einstein relation obtained from stationarity and the dynamics only

(no equipartition, no TRI, no Boltzmann…)

ACTIVE BROWNIAN PARTICLES

A MINIMAL STATISTICAL MODEL

$$\begin{cases} \dot{\boldsymbol{r}}_i(t) = \mu_0 \boldsymbol{F}_i + v_0 \boldsymbol{n}_i(t) + \boldsymbol{\xi}_i(t) \\ \dot{\theta}_i(t) = \nu_i(t) \end{cases}$$



$\boldsymbol{\xi}_i(t)$ and $\boldsymbol{\nu}_i(t)$ Gaussian white noises

Different origins!

Orientational vector

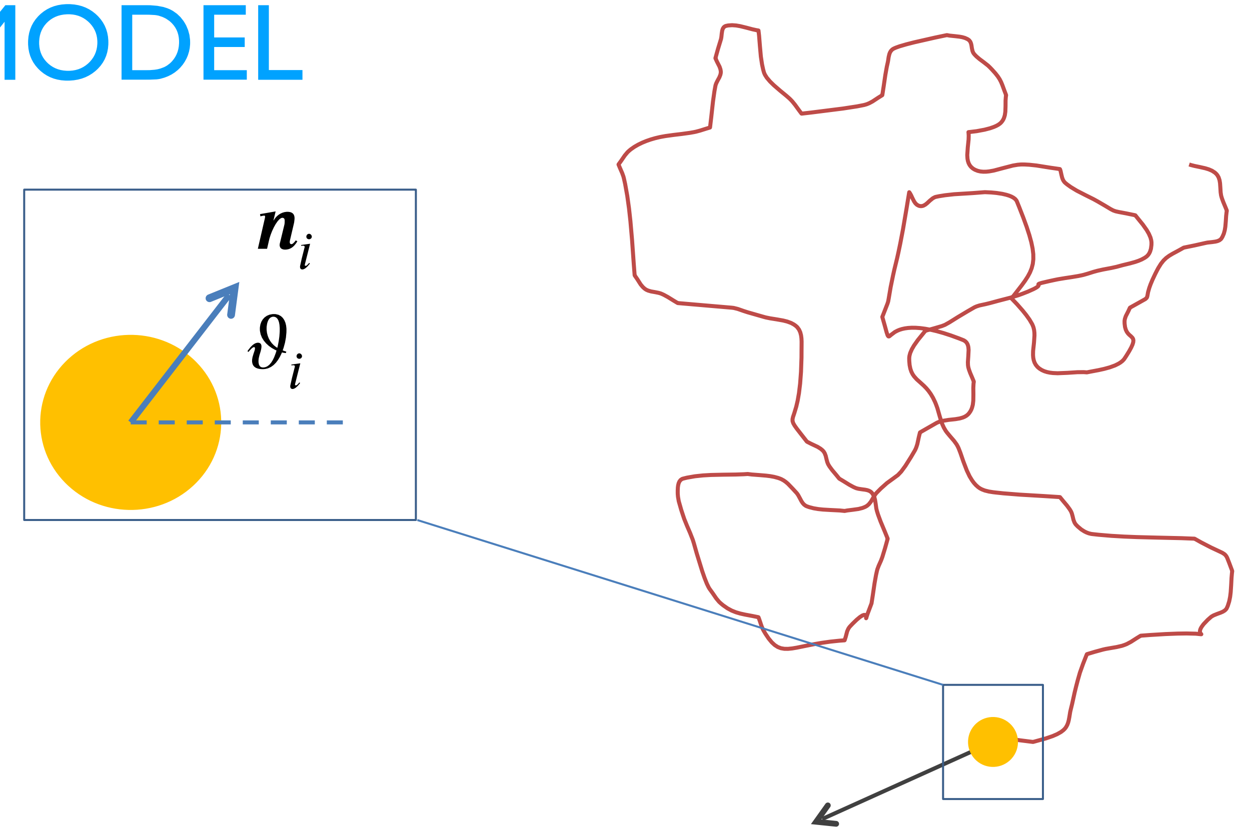
$$\mathbf{n}_i = (\cos\theta_i, \sin\theta_i)$$

Persistent brownian motion

Peclet number: $Pe = \dfrac{v_0 \tau}{R}$

Measure of activity

A MINIMAL STATISTICAL MODEL



Initial probability distribution:

$$\Psi_0 \sim e^{-\beta(U(\{\mathbf{r}_i\})+\boxed{v_0\tau\, \Xi(\{\mathbf{r}_i,\theta_i\}))}}$$

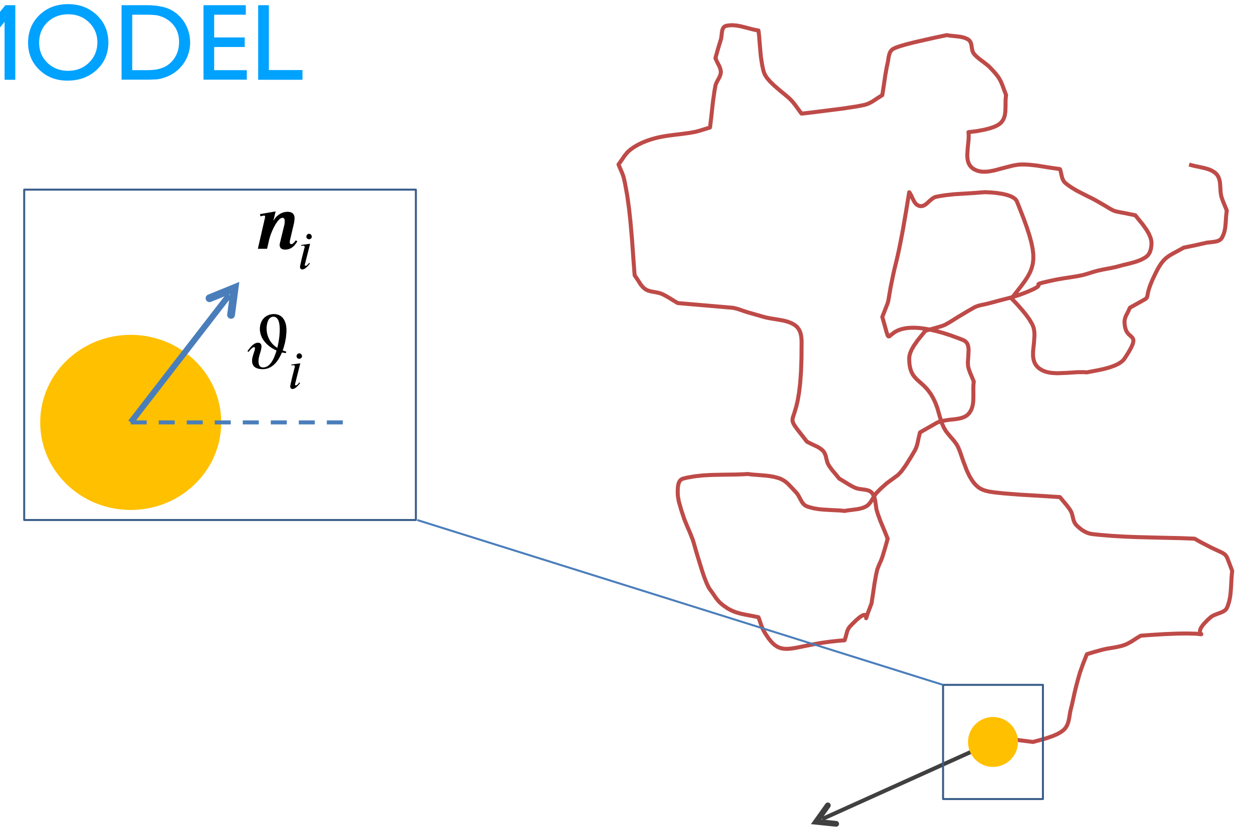
Active information potential

Violation of detailed balance!

$$\boxed{\Omega_0^\dagger - \overline{\Omega}_0^\dagger = 2\mathscr{V}\cdot\nabla}$$

Phase space velocity $\mathscr{V}_i \equiv \boldsymbol{j}_i/\Psi_0 = v_0\left[(\boldsymbol{n}_i + \mu_0\tau\,\partial_i\Xi), \beta\partial_{\theta_i}\Xi\right]$

A MINIMAL STATISTICAL MODEL



Active ITT:

$$\langle A \rangle_t - \langle A \rangle_0 = \beta \int_0^t ds \langle \dot{V}(0) A(s) \rangle_0 - \boxed{\int_0^t ds \langle \mathscr{V} \cdot \nabla V(0) A(s) \rangle_0}$$

Implicit term

Fully general expression

Lack of universality in the nonequilibrium response:
we need to know something on the initial distribution state

The quest for a (markovian) approximation to derive closed expressions for the transport coefficients

REDUCED DYNAMICS

$$\dot{\boldsymbol{r}}_i(t) = \mu_0 \boldsymbol{F}_i + \mu_0 \mathbf{f}_{ext} + \boldsymbol{\chi}_i(t)$$

$$\langle \boldsymbol{\chi}_i(t) \boldsymbol{\chi}_j(t') \rangle = 2\mu_0 k_B T \mathbf{1} \delta(t - t') \delta_{ij} + \frac{v_0^2}{2} e^{-|t-t'|/\tau} \delta_{ij} \mathbf{1}$$

$$\Downarrow$$

Generation of **memory** by coarse-graining

MARKOVIAN APPROXIMATION

***Fox method**: perturbative expansion in the correlation time*

[R. Fox PRA (1986), Farage et *Al*. PRE (2015), UMB Marconi et Al. Soft Matter (2015)]

$$\frac{\partial \Psi(\Gamma, t)}{\partial t} = \Omega^{eff}(\Gamma)\Psi(\Gamma, t) + \Omega^{ext}(\Gamma)\Psi(\Gamma, t)$$

$$\Omega_N^{eff} = \sum_{i=1}^{N} \partial_i \cdot D_i(\Gamma)\left[\partial_i - \beta \boldsymbol{F}_i^{eff}(\Gamma)\right]$$

Valid for:

$$1 - \mu_0 \tau \partial_i \cdot \mathbf{F}_i > 0$$

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$$D_i(\Gamma) = \mu_0 k_B T + \frac{v_0^2 \tau}{2}\left(1 + \frac{\mu_0 \tau \partial_i \cdot \boldsymbol{F}_i}{1 - \mu_0 \tau \partial_i \cdot \boldsymbol{F}_i}\right)$$

Effective diffusion matrix

$$\boldsymbol{F}_i^{eff}(\Gamma) = \frac{\boldsymbol{F}_i}{\mathscr{D}_i} - k_B T \frac{\partial_i \mathscr{D}_i}{\mathscr{D}_i} \qquad \mathscr{D}_i = \frac{D_i}{\mu_0 k_B T}$$

Effective many body force

MOBILITY

Detailed balance restored!

Zero-probability flux solution: $\partial_i \Psi_0(\Gamma) = \beta \boldsymbol{F}_i^{eff}(\Gamma)\Psi_0(\Gamma)$

$$\mu_i = \mu_0 \left(1 - \mu_0 \beta \int_0^t dt' \langle F_i^{eff,x}(0) F_i^x(t') \rangle \right)$$

Force-force correlator

Green-Kubo like relation

From Green-Kubo to Stokes-Einstein

CORRECTIONS TO STOKES-EINSTEIN

$$\mu_0^2 \langle F_n^x(0) \cdot F_n^x(t) \rangle = \langle \mathscr{D}_n \rangle_0 \, \delta(t) - \langle \dot{x}_n(0) \dot{x}_n(t) \rangle_0$$

+ Expansion to first order in $\tau \mu_0 \partial_i \cdot \mathbf{F}_i$

$$D = D_a + \frac{\mu_0 v_0^2 \tau^2}{2} \langle \partial_i \cdot \boldsymbol{F}_i \rangle - \mu_0^2 \int_0^\infty ds \langle F_i^x(0) F_i^x(s) \rangle$$

$$\frac{\mu}{\mu_0} = \frac{D}{D_a} - \frac{\mu_0 v_0^2 \tau^2}{2 D_a} \langle \partial_i \cdot \boldsymbol{F}_i \rangle + \frac{v_0^2 \tau^2 \mu_0^3}{2 D_a^2} \int_0^\infty ds \langle F_n^x(0) \partial_n \cdot \boldsymbol{F}_n(0) F_n^x(s) \rangle_0 + \frac{v_0^2 \tau^2 \mu_0^2}{2 D_a} \int_0^\infty ds \langle \partial_n^x \partial_n \cdot \boldsymbol{F}_n(0) F_n^x(s) \rangle_0$$

Comparison with brownian dynamics simulations

MOBILITY AND DIFFUSIVITY

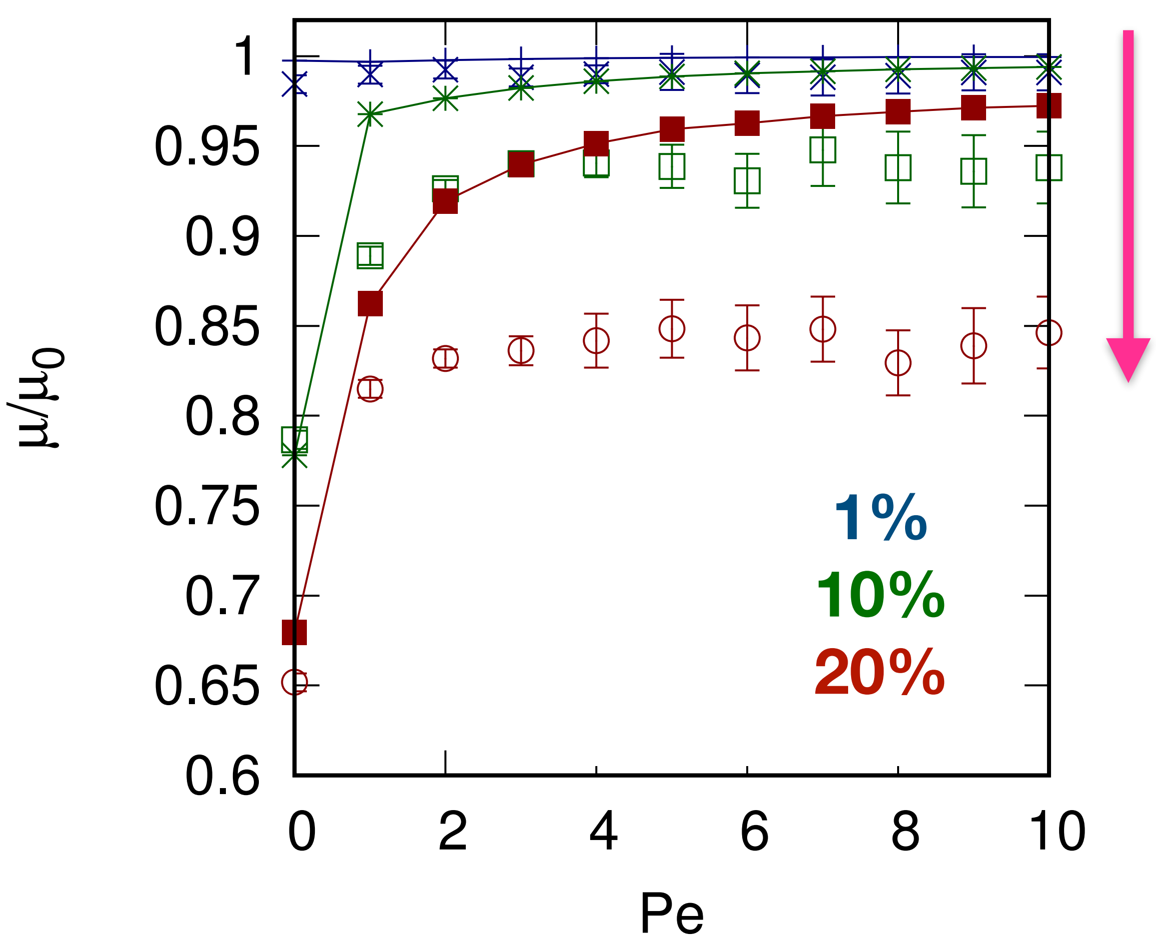
Malliavin weights: transport coefficients from unperturbed dynamics

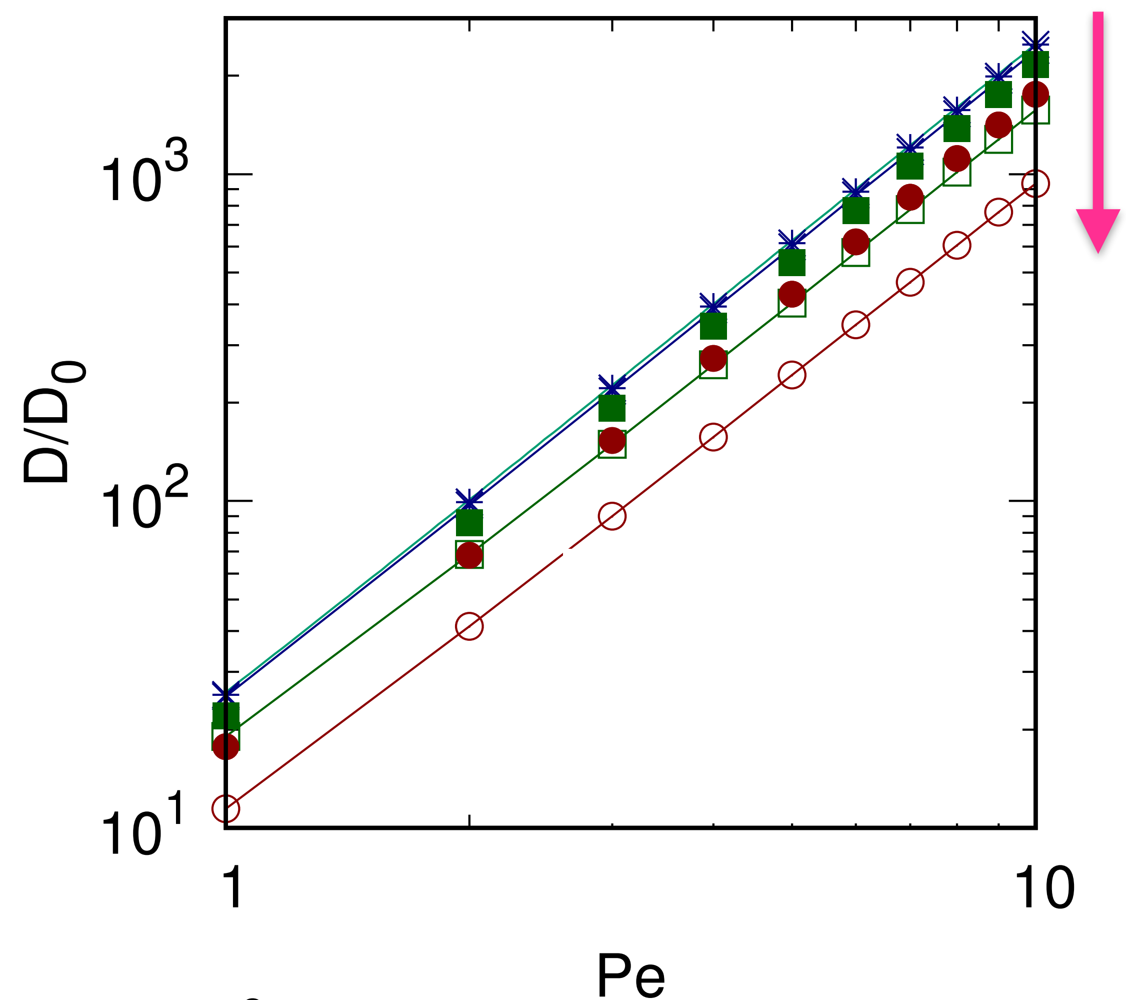
[Warren and Allen, PRL, 2012] [G Szamel, EPL, 2017]

MOBILITY AND DIFFUSIVITY

Malliavin weights: transport coefficients from unperturbed dynamics

Role of interactions





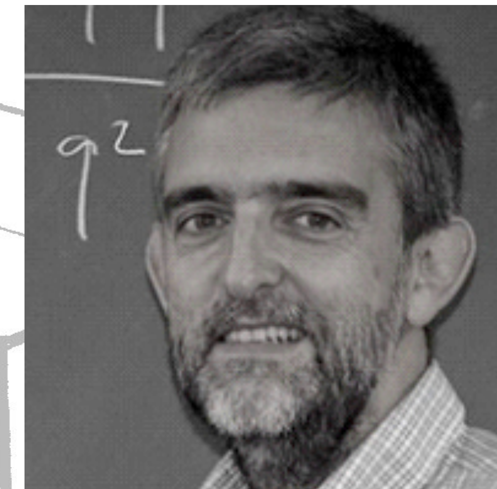
Ideal active gas: $D_a = \mu_0 k_B T + \dfrac{v_0^2 \tau}{2}$

CONCLUSIONS

- General ITT expression for active response
- Closed Green-Kubo expressions for mobility and diffusivity of interacting active particles
- Good qualitative agreement with brownian dynamics simulations
- Unveiling the interplay of interactions and self-propulsion

ACKNOWLEDGEMENT

Demian Levis



Ignacio Pagonabarraga

[preprint arXiv:1907.02560]

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NANOTRANS
Transport of Soft Matter at the Nanoscale

MARIE CURIE ACTIONS

$$e^{\Omega^{\dagger} t} A(\Gamma) \equiv A(T^{t}\,\Gamma) = A(t)$$ **Forward operator**

$$e^{\bar{\Omega}^{\dagger} t} A(\Gamma) \equiv A(T^{-t}\,\Gamma) = A(-t)$$ **Time-reversal operator**

Detailed Balance

$$\Omega^{\dagger} = \bar{\Omega}^{\dagger}$$

MALLIAVIN WEIGHTS

Stochastic technique to compute response function from weighted averages over unperturbed dynamics

Extended phase space

$$\Gamma' \to (\Gamma, q_\lambda)$$

Malliavin weight

$$\langle q_\lambda \rangle \equiv \frac{\partial}{\partial \lambda} \log P(\Gamma, t)$$

$$\Downarrow$$

Response function

$$\frac{\partial \langle A \rangle}{\partial \lambda}(t) = \langle A q_\lambda \rangle (t)$$

Updating rule

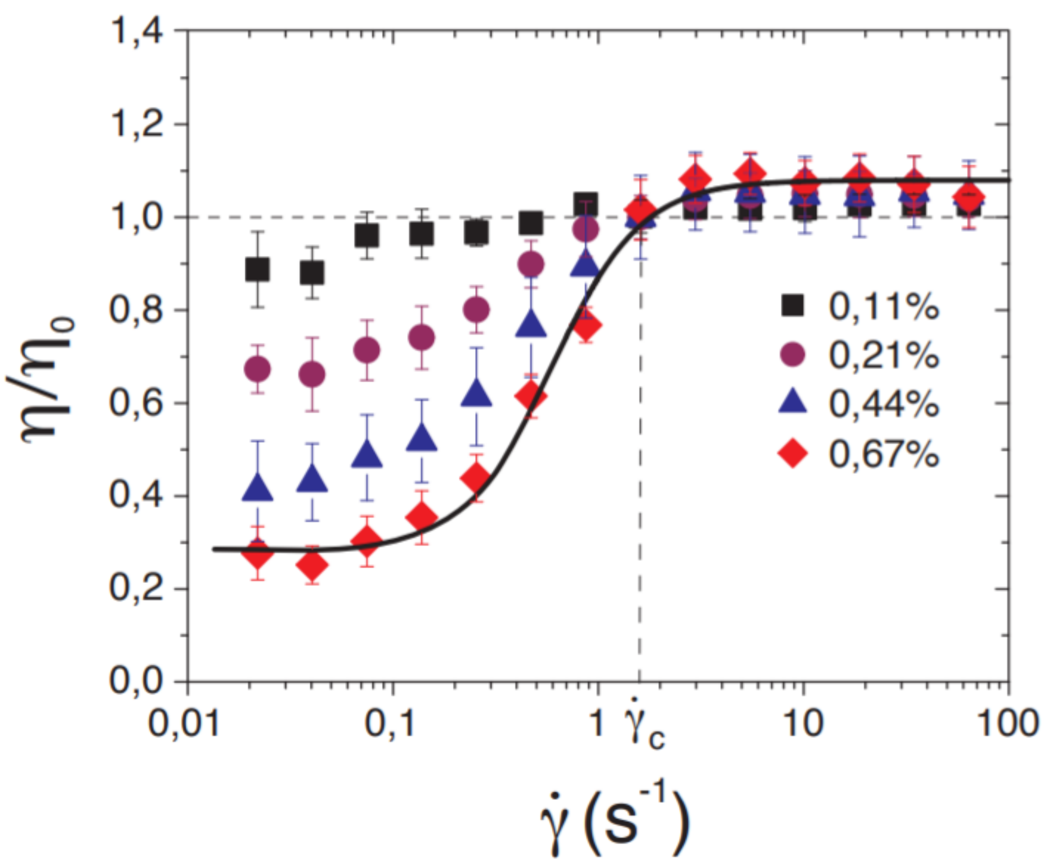
$$q'_\lambda = q_\lambda + \frac{\partial \log \mathbf{W}}{\partial \lambda}$$

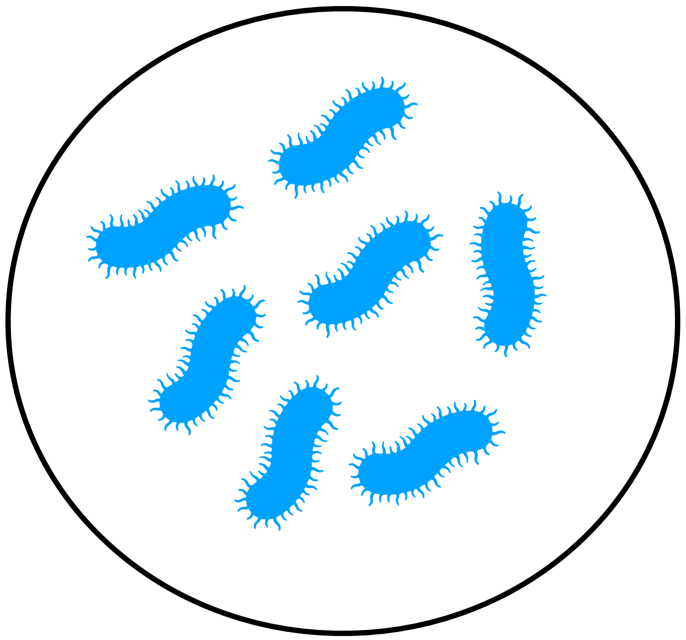
Propagator

ACTIVE TRANSPORT

Turning bacteria into a superfluid

[H. Matias Lopez *et al* PRL (2015)]





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