Non-equilibrium dynamics of active agents

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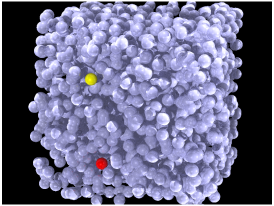
FOR1394 DFG Research Unit
Nonlinear Response
to Probe Vitrification

Further Interests: Beyond Brownian Motion

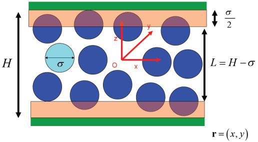
Cellular Crowding & Porous Media



Spanner *et al* PRL (2016)
Schirmacher *et al* PRL (2015)
Schnyder *et al* Soft Matter (2015)
Höfling *et al* PRL (2006), PRL (2007)
JCP (2008), Soft Matter (2011)
Rep. Prog. Phys. (2013)
Kammerer *et al* EPL (2010)
Franosch *et al* Chem. Phys. (2010)

Collaboration:
Höfling (FU Berlin)

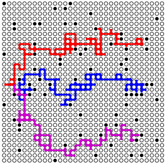
Glass Transition



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

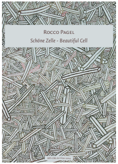
Collaboration:
R. Schilling (Mainz)
F. Varnik (Bochum)

Driven transport



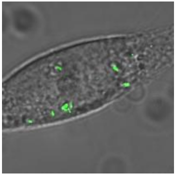
Leitmann *et al* PRL (2017)
Leitmann *et al* PRL (2013)

Needles & Biofilaments



Leitmann *et al* PRL (2016)
Munk *et al* EPL (2009)
Höfling *et al* PRL (2006), PRL (2008)
Höfling *et al* PRE (Rapid) (2008)

Intracellular Transport



Meier *et al* PNAS (2011)

Collaboration:
Heinrich (Leiden)

Discovery of Brownian Motion

Robert Brown

scottish botanist

1773-1858

Clarkia pilchella

wikipedia.org/wiki/Brownsche_Bewegung

Observation of pollen in water

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

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

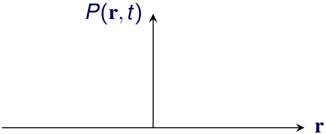
$$\langle[\mathbf{r}(t)-\mathbf{r}(0)]^2\rangle = 6Dt$$

- solves diffusion equation

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

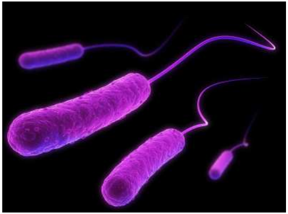
→ statistical interpretation of diffusion
central limit theorem

Albert Einstein (1905)



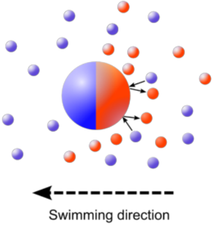
same time: Marian v. Smoluchowski

Self-propelled agents



Bacteria using flagella to swim

Paramecium uses cilia

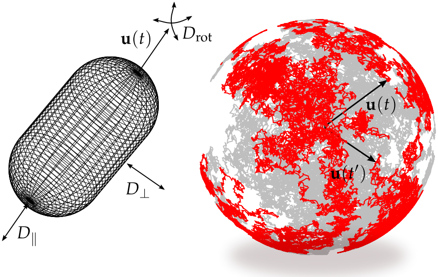


self-propelled Janus particle

- 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

Model set-up

Active Brownian Particle



- Active propulsion with constant velocity v along the long axis $\mathbf{u}, |\mathbf{u}| = 1$
- Rotational diffusion D_{rot}
- Anisotropic translational diffusion $D_\parallel, D_\perp$
- ignores microscopic origin of propulsion, effective description
- simplistic model encoding persistent random walk
  persistence length $\ell = v^2/D_{\mathrm{rot}}$, persistence time $\tau = v/D_{\mathrm{rot}}$

Stochastic equations

Anisotropic active Brownian particle

$$\mathrm{d}\mathbf{u} = -2D_{\mathrm{rot}}\mathbf{u}\,\mathrm{d}t - \sqrt{2D_{\mathrm{rot}}}\,\mathbf{u} \times \xi\,\mathrm{d}t$$

$$\mathrm{d}\mathbf{r} = v\,\mathbf{u}\,\mathrm{d}t + \left[\sqrt{2D_{\|}}\,\mathbf{u}\mathbf{u} + \sqrt{2D_{\perp}}(\mathbb{I} - \mathbf{u}\mathbf{u})\right]\eta\,\mathrm{d}t$$

Position fixed velocity orientation

$$\langle \xi_i(t)\xi_j(t')\rangle = \langle \eta_i(t)\eta_j(t')\rangle = \delta_{ij}\delta(t - t')$$

independent Gaussian white noise

- multiplicative noise (Itō)
- translational anisotropy $\Delta D = D_{\|} - D_{\perp}$
  mean diffusion coefficient $\bar{D} = (D_{\|} + 2D_{\perp})/3$
- 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 $\mathrm{Pe} = va/D_{\mathrm{rot}}$

Smoluchowski equation

conditional probability density $\mathbb{P}(\Delta\mathbf{r},\mathbf{u},t|\mathbf{u}_0)$ (Green function)

Perrin equation (Markov process)

$$\partial_t\mathbb{P} = D_{\mathrm{rot}}\Delta_{\mathbf{u}}\mathbb{P} - v\mathbf{u}\cdot(\partial_{\mathbf{r}}\mathbb{P}) + \partial_{\mathbf{r}}\cdot\left[D_{\parallel}(\partial_{\mathbf{r}}\mathbb{P}) - \Delta D(\mathbb{I}-\mathbf{uu})\cdot(\partial_{\mathbf{r}}\mathbb{P})\right]$$

orientational diffusion

active propulsion

anisotropic diffusion $\Delta D = D_{\parallel} - D_{\perp}$

- Smoluchowski equation for non-equilibrium dynamics
- coupling between orientation, active propulsion, and translation
- spatial Fourier transform $\tilde{\mathbb{P}}(\mathbf{k},\mathbf{u},t|\mathbf{u}_0) = \int \mathrm{d}^3\Delta r \exp(-i\mathbf{k}\cdot\Delta\mathbf{r})\mathbb{P}(\Delta\mathbf{r},\mathbf{u},t|\mathbf{u}_0)$

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

  reminiscent of quantum pedundulum
  exact solution in terms of spheroidal wave functions
- solution for intermediate scattering function

$$F(\mathbf{k},t) = \langle\exp(-i\mathbf{k}\cdot\Delta\mathbf{r}(t))\rangle = \int_{S^2}\mathrm{d}\mathbf{u}\int_{S^2}\frac{\mathrm{d}\mathbf{u}_0}{4\pi}\tilde{\mathbb{P}}(\mathbf{k},\mathbf{u},t|\mathbf{u}_0)$$

Christina Kurzthaler *et al*, Sci. Rep. (2016)

Spheroidal wave functions

Separation ansatz

- choose coordinates $\mathbf{k}$ in z-direction, parametrize $\mathbf{u} = (\sin\vartheta\cos\varphi, \sin\vartheta\sin\varphi, \cos\vartheta)$, write $\eta = \cos\vartheta$
- separation ansatz yields superposition of eigenfunctions

$$\tilde{P}(\mathbf{k},\mathbf{u},t|\mathbf{u}_0) = \frac{1}{2\pi} e^{-D_\perp k^2 t} \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} e^{im(\varphi-\varphi_0)} \mathrm{Ps}_\ell^m(c,R,\eta)\mathrm{Ps}_\ell^m(c,R,\eta_0)\exp(-A_\ell^m D_{\mathrm{rot}} t)$$

- generalized spheroidal wave functions $\mathrm{Ps}_\ell^m(R,c,\eta)$

$$\left[\frac{\mathrm{d}}{\mathrm{d}\eta}\left((1-\eta^2)\frac{\mathrm{d}}{\mathrm{d}\eta}\right) + R\eta - c^2\eta^2 + \frac{m^2}{1-\eta^2} + A_\ell^m\right]\mathrm{Ps}_\ell^m(c,R,\eta) = 0$$

active propulsion
$R = -ikv/D_{\mathrm{rot}}$

anisotropy
$c^2 = \Delta D k^2/D_{\mathrm{rot}}$

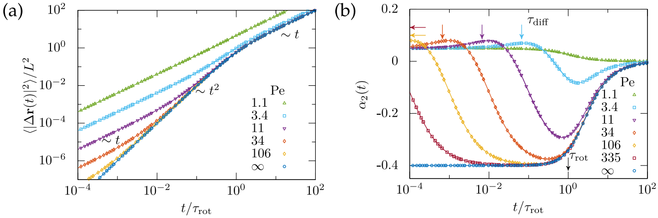
eigenvalue

- deformations of (associated) Legendre polynomials $\mathrm{P}_\ell^m(\eta)$
- Intermediate scattering function

$$F(k,t) = \frac{1}{2\pi} e^{-D_\perp k^2 t} \sum_{\ell=0}^{\infty} \exp(-A_\ell^m D_{\mathrm{rot}} t) \left[\int_{-1}^{1} \mathrm{d}\eta\, \mathrm{Ps}_\ell^0(c,R,\eta)\right]^2$$

Low-order moments

Mean-square displacement and non-gaussian parameter



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

$$F(k,t) = 1 - \frac{k^2}{3!}\langle|\Delta\mathbf{r}(t)|^2\rangle + \frac{k^4}{5!}\langle|\Delta\mathbf{r}(t)|^4\rangle + \mathscr{O}(k^6)$$

- MSD initially translational diffusion dominates, persistent swimming, effective diffusion

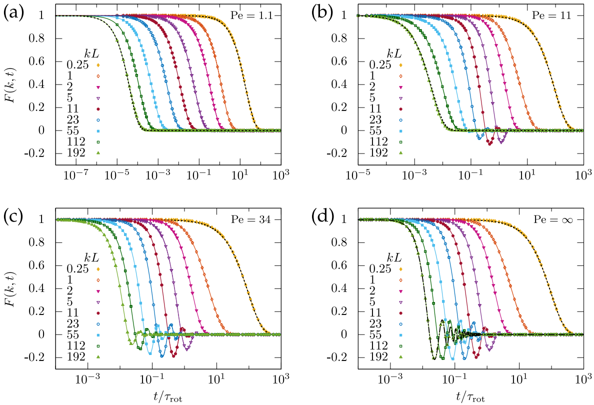
$$\langle|\Delta\mathbf{r}(t)|^2\rangle = 6\bar{D}t + \frac{v^2}{2D_{\rm rot}^2}\left(e^{-2D_{\rm rot}t} + 2D_{\rm rot}t - 1\right)$$

- Non-gaussian parameter

$$\alpha_2(t) = \frac{3\langle|\Delta\mathbf{r}(t)|^4\rangle}{5\langle|\Delta\mathbf{r}(t)|^2\rangle^2} - 1$$

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

Intermediate scattering function



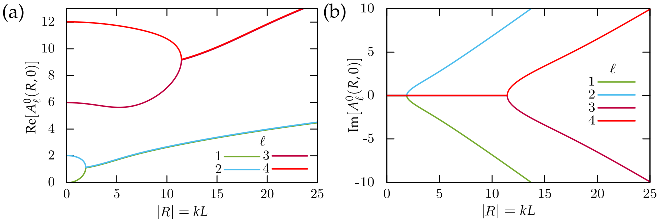
- characteristic oscillations emerge at intermediate wavenumbers fingerprint of persistent swimming

$$F(k,t) = \left\langle \frac{\sin(k|\Delta\mathbf{r}(t)|)}{k|\Delta\mathbf{r}(t)|} \right\rangle$$

- large wavenumbers anisotropic translational diffusion
- small wavenumbers effective diffusion $D_{\text{eff}} = \bar{D} + v^2/6D_{\text{rot}}$

Oscillations

How can oscillations emerge?



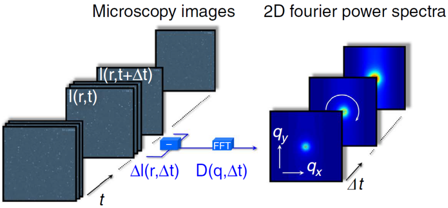
- Intermediate scattering function sum of relaxing exponentials?

$$F(k,t) = \frac{1}{2\pi} e^{-D_\perp k^2 t} \sum_{\ell=0}^{\infty} \exp(-A_\ell^m D_{\mathrm{rot}} t) \left[\int_{-1}^{1} \mathrm{d}\eta \, \mathrm{Ps}_\ell^0(c,R,\eta) \right]^2$$

- Eigenvalue problem is non-hermitian, eigenvalues become complex bifurcations in the eigenvalues, fingerprint of active motion

Christina Kurzthaler *et al*, Sci. Rep. (2016)

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

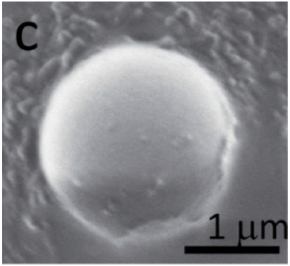
intermediate scattering function

camera noise

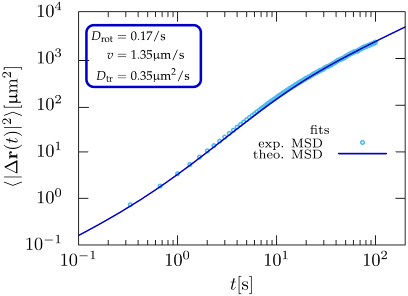
- complementary to single particle tracking (shorter times)

Comparison to Experiments

Janus particles



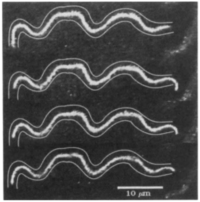
Brown & Poon, Soft Matter (2014)



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

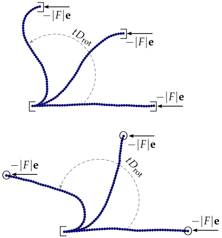
- so far: extract motility parameters from MSD

Semiflexible polymer



F-actin solution

J. Käs *et al*, Nature (1994)



Kurzthaler *et al* (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 $\mathbf{u} = \mathrm{d}\mathbf{r}/\mathrm{d}s$

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

κ bending rigidity

stetching force

- partition sum path integral

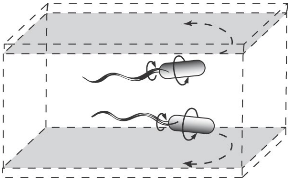
$$Z(\mathbf{u}_L L, \mathbf{u}_0, 0) = \int_{\mathbf{u}(0)=\mathbf{u}_0}^{\mathbf{u}(L)=\mathbf{u}_L} \mathscr{D}[\mathbf{u}(s)] \exp(-\mathscr{H}/k_B T)$$

- path integral solved by Fokker-Planck equation

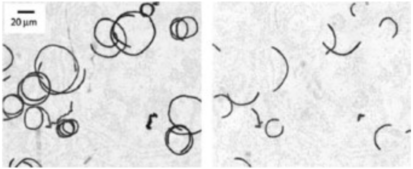
$$\partial_s Z(\mathbf{u}, s|\mathbf{u}_0 0) = \left[\frac{1}{\ell_p} \Delta_{\mathbf{u}} + \mathbf{F} \cdot \mathbf{u} \right] Z(\mathbf{u}, s|\mathbf{u}_0 0)$$

Circle swimmers

Escherichia coli



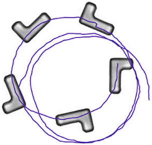
DiLuzio *et al* Nature (2005)

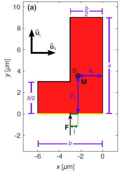


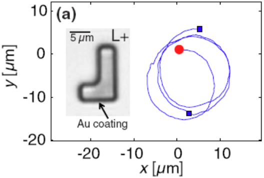
Lauga *et al* Biophys. J. (2006)

- Chirality of flagellar motion
- Hydrodynamic coupling close to boundaries
  → circular motion

Asymmetric self-propelled particles



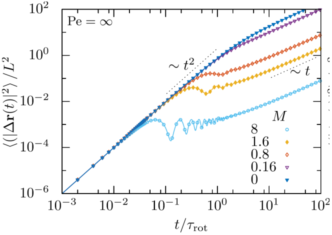


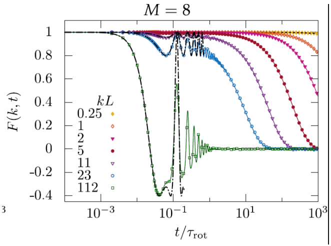


Kümmel *et al* PRL (2013)

- Propulsion force not at center
  → torque
- Hydrodynamic coupling close to boundaries circular motion

Circle swimmers





- angular drift velocity ω
- quality factor $M = \frac{\omega/2\pi}{D_{\text{rot}}}$

non-trivial oscillations

isotropy $F(k,t) = \langle \mathrm{J}_0(k|\Delta\mathbf{r}(t)|)\rangle$

exact solution in terms of generalizations of Mathieu functions

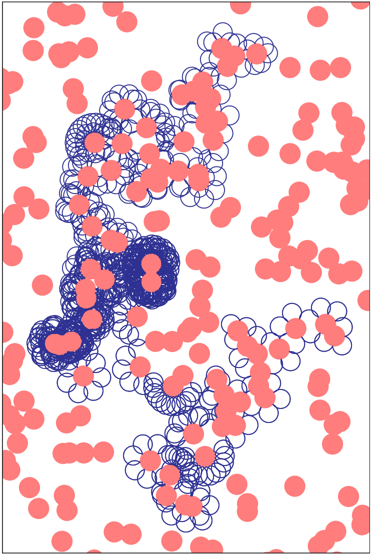
Fokker-Planck equation (2D)

$$\partial_t \mathbb{P} = -v\mathbf{u}\cdot\partial_{\mathbf{r}}\mathbb{P} - \omega\partial_\vartheta\mathbb{P} + \partial_{\mathbf{r}}\cdot(\mathbf{D}\cdot\partial_{\mathbf{r}}\mathbb{P}) + D_{\text{rot}}\partial_\vartheta^2\mathbb{P}$$

Kurzthaler *et al* Soft Matter

Disordered environment

Skipping orbits

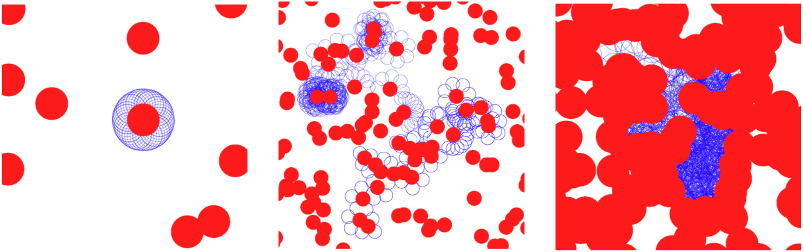


- independently distributed obstacles exclusion radius σ
  dimensionless scatterer density
  $n^* = n\sigma^2$
  → **Lorentz model**
- idealize to pure circular motion
  → deterministic orbits
  dimensionless trajectory curvature
  $B = \sigma / R$
- **skipping orbits** along edges of scatterer clusters

long-range transport
by edge percolation

Phases

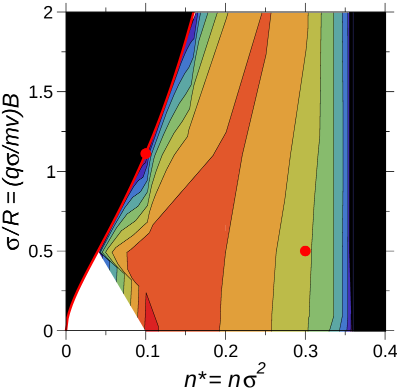
Phase behavior



- low scatterer density:
  skipping orbit around isolated clusters only
  → insulating phase
- intermediate density:
  skipping orbits percolate through entire system
  → diffusive phase
- high scatterer density:
  void space consists only of finite pockets
  → localized phase

Purely geometric transition

Phase diagram



isodiffusivities on log scale

$$D = \lim_{t\to\infty} \frac{1}{4}\frac{\mathrm{d}}{\mathrm{d}t}\delta r^2(t)$$

- Transition line to localized phase independent of curvature
  → conventional percolation of void space

$$n_c^* = 0.359081\ldots$$

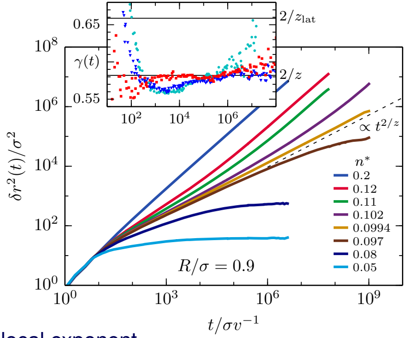
critical exponents correspond to conventional percolative transport on lattices

- Delocalization transition

$$n_m(R) = n_c^* \sigma^2/(\sigma + R)^2$$

→ percolation of disks+halo
exact result!, 2 significant digits
Kuzmany & Spohn, PRE (1998)

Mean-square displacements



local exponent

$$\gamma(t) = \frac{\mathrm{d} \log \delta r^2(t)}{\mathrm{d} \log t}$$

anamalous transport at $n^*_m = n^*_m(R)$

$$\gamma = \lim_{t\to\infty} \gamma(t) = 0.581 \pm 0.005$$

W. Schirmacher *et al*, *Phys. Rev. Lett.* (2015)

- mean-square displacement (MSD)

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

- *subdiffusive* close to $n^*_m = n^*_m(R)$

$$\delta r^2(t \to \infty) \sim t^{2/z}$$

  critical exponent $z = 2/\gamma$ measured value $z = 3.44 \pm 0.03$

- different from random walkers on percolating lattices
  $z \neq z_{\rm lat} = 3.036 \pm 0.001$
- divergent localization length in insulating phase

$$\ell^2 = \delta r^2(t \to \infty)$$

New dynamic universality class

Résumé

Self-propelled particles

- active Brownian particle a minimal model for persistent swimmer
- orientational motion strongly coupled to translational dynamics
- exact solution in terms of eigenfunctions
- analytic expressions for mean-square displacement, fourth moment
- space-resolved dynamics – intermediate scattering functions directly measurable
  unprecedented agreement with experiments
- maps to semiflexible polymers – exact force-extension relation
- bacteria – include run-and-tumble motion, renewal processes
- swimmers with constant torque – circular motion
- external force – gravitaxis, optical trapping
- walls, confinement, porous media
  critical phenomena – new universality classes