

# Transport in heterogeneous media – diffusion, fractals and anomalous dynamics

Thomas Franosch

Arnold Sommerfeld Center for Theoretical Physics  
and Center for NanoScience (CeNS)  
Ludwig-Maximilians-Universität München

Theorie-Kolloquium, Oldenburg, April 16, 2009

Ludwig  
Maximilians  
Universität

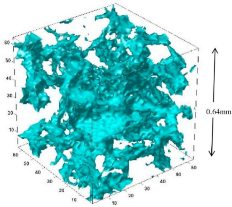


ARNOLD SOMMERFELD  
CENTER FOR THEORETICAL PHYSICS



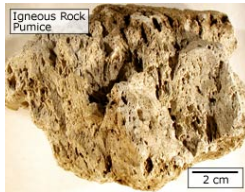
- 1 Motivation
- 2 Ant in the labyrinth
  - fractals
  - percolation
  - transport
- 3 Transport in heterogeneous media
  - Molecular Dynamics simulations
  - Mean-square displacement
  - VACF – two dimensions
  - Continuum Percolation Theory
  - Dynamic Scaling Hypothesis
  - Corrections to scaling

# Motivation- Transport in Disordered Media



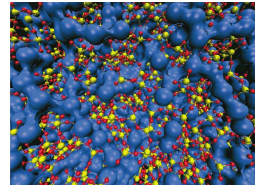
sandstone

Okabe & Blunt, PRE (2004)



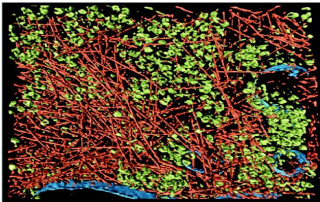
pumice

M. Nyman, TERC



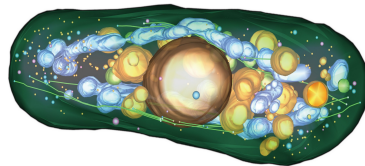
Na-silicate

A. Meyer *et al*, PRL (2004)



cellular crowding

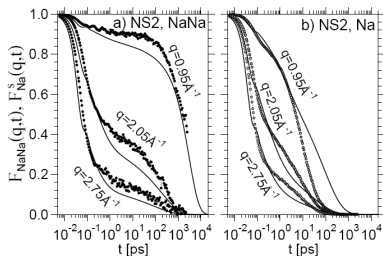
O. Medalia *et al* (2002) Science



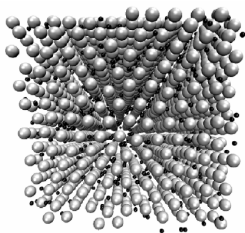
yeast

J. Höög, EMBL

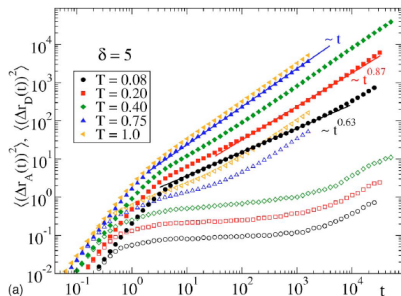
# Dense mixtures



Na-Silicate T. Voigtmann, J. Horbach



disparate Yukawa particles N. Kikuchi, J. Horbach



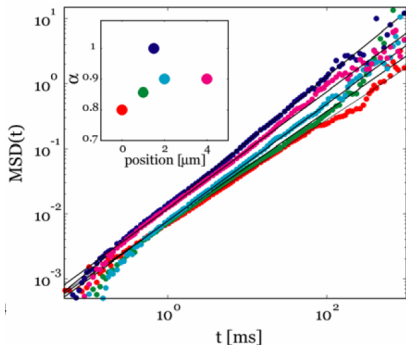
disparate soft spheres

A. Moreno, J. Colmenero

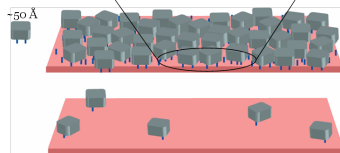
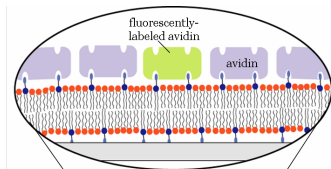
subdiffusive behavior in  
strongly disparate mixtures



# Membranes



- avidin binds irreversibly to biotinylated supported lipid bilayer (SLB)
- Fluorescent correlation spectroscopy (FCS)



M. Horton, J. Rädler, LMU

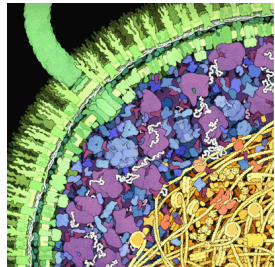
subdiffusive behavior in crowded membranes

# Molecular crowding

- Molecular crowding

"Molecular crowding is more accurately termed the **excluded volume effect**, because the mutual impenetrability of all solute molecules is its most basic characteristic. This nonspecific steric repulsion is always present, regardless of any other attractive or repulsive interactions that might occur between the solute molecules." **R. John Ellis 2001**

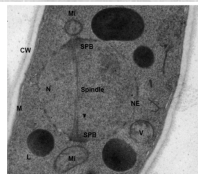
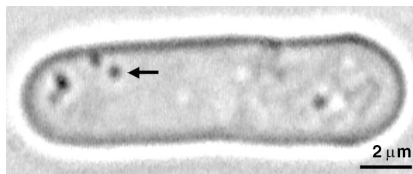
- 30% volume fraction by sugars, lipids, membranes
- **anomalous** transport in the cell
- chemical reactions are slow
- **apparent** density-dependent exponents?
  - alternatively: huge crossover regimes
  - origins: static **heterogeneities**, random traps, polymer networks



E-coli

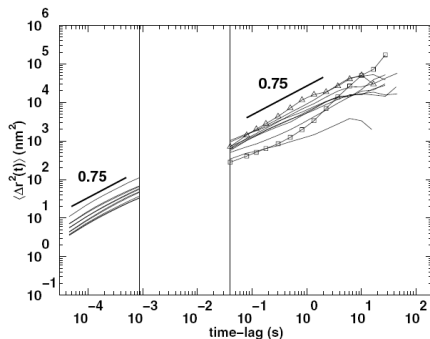
D. Goodsell

related: M. Weiss, M.J. Saxton



living fission yeast

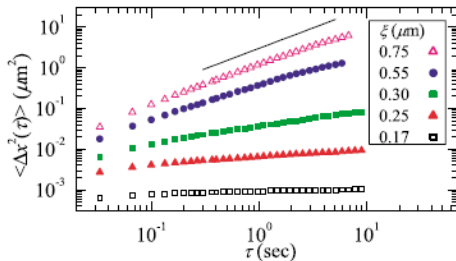
- small lipid granules that occur naturally in the cytoplasm
- particle tracking in video microscopy



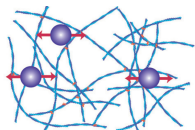
Iva Tolić-Nørrelykke, MPI-CBG

subdiffusive behavior in crowded cells

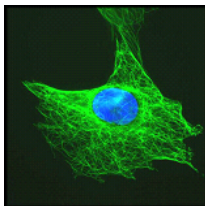
# Biopolymer Networks



Wong *et al*, PRL 2004

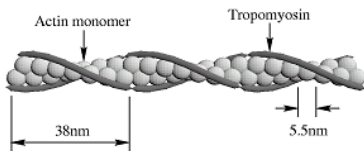


Bausch, Kroy



F-actin

Thomas Franosch



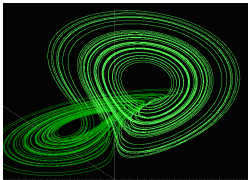
- multiple particle tracking
- entangled F-actin filament networks
- particle diameter *a* comparable to **mesh size**  $\xi$

**subdiffusive** behavior in crosslinked networks

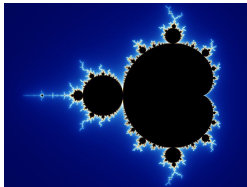
## Origin of anomalous transport?

- dense system constitutes course of **obstacles**
- long-living heterogeneities
- many length scales induce **hierarchy** of time scales
- Complex systems – also other mechanisms
  - distribution of sticking times
  - spectrum of relaxation times in polymers
  - glassy dynamics
  - phase separation
  - non-equilibrium, aging
  - **living**, active systems

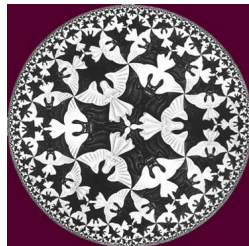
- 1 Motivation
- 2 Ant in the labyrinth
  - fractals
  - percolation
  - transport
- 3 Transport in heterogeneous media
  - Molecular Dynamics simulations
  - Mean-square displacement
  - VACF – two dimensions
  - Continuum Percolation Theory
  - Dynamic Scaling Hypothesis
  - Corrections to scaling



Lorenz attractor

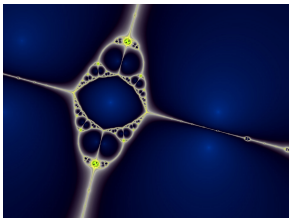


Mandelbrot set



Devils and Angels

M.C. Escher



bubbles fractal

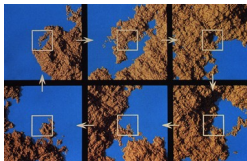


[www.fractalarts.com](http://www.fractalarts.com)

# fractals – picture gallery



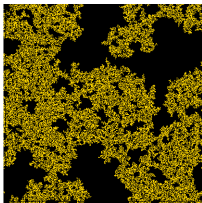
Romanesco



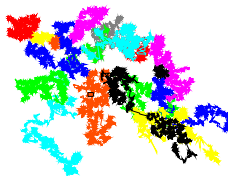
coastline



Sierpiński



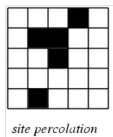
site percolation



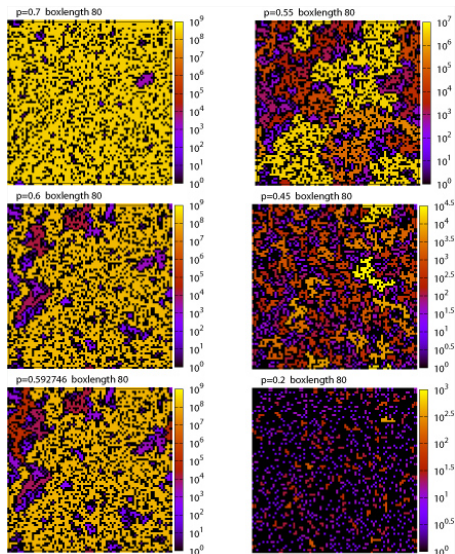
continuum percolation



# Site Percolation

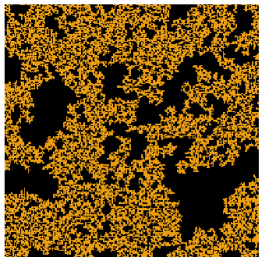


- sites are occupied with probability  $p$
- Occupied sites form clusters
- An **infinite** cluster is present above some threshold
- correlation length  $\xi(p)$ : size of the largest **finite** cluster

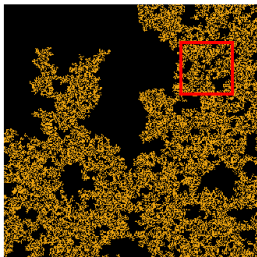


# Self-similarity

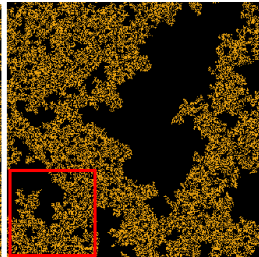
$p=0.592746$



L=200

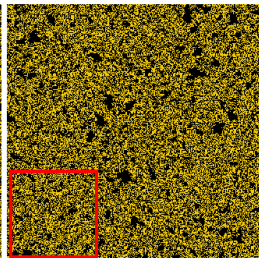
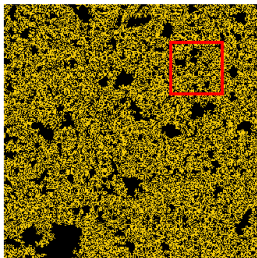
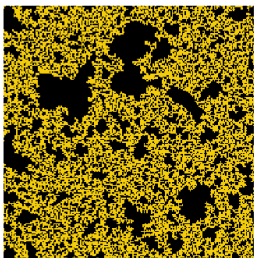


L=1000



L=3000

$p=0.6$



# Fractal dimension

- self-similarity at criticality
- mass of the infinite cluster

$$M(r) \sim r^{d_f}$$

$d_f$  is the **fractal dimension**

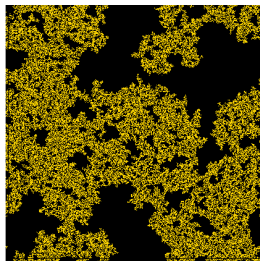
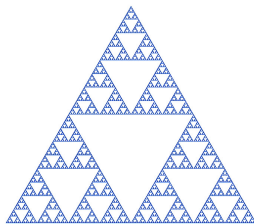
- Sierpiński gasket

$$d_f = \frac{\log 3}{\log 2}$$

- percolation

$$d_f = 91/48 \approx 1.9 \quad (d = 2)$$

$$d_f \approx 2.53 \quad (d = 3)$$



- self-similarity implies

- correlation length  $\xi \sim |p - p_c|^{-\nu}$
- infinite cluster  $P_\infty \sim (p - p_c)^\beta$
- mean finite cluster size  
 $\ell \sim |p - p_c|^{-\nu + \beta/2}$
- cluster size distribution at  $p = p_c$

$$n_s \sim s^{-\tau}$$

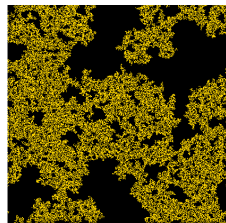
- similar to continuous phase transitions

- $p$  plays rôle of temperature
- $P_\infty$  order parameter

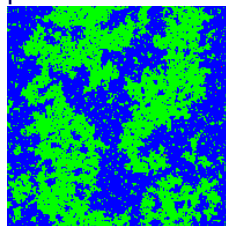
- scaling relations

- Fisher exponent and fractal dimension

$$\tau = 1 + d/d_f \quad d_f = d - \beta/\nu$$

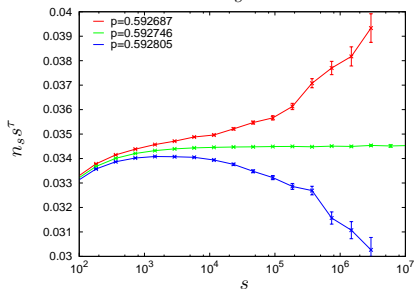
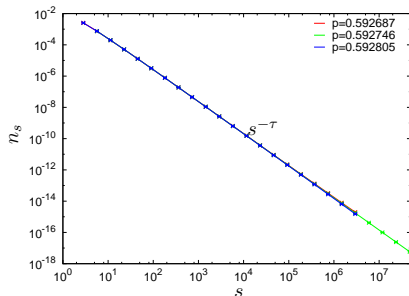


percolation



Ising model

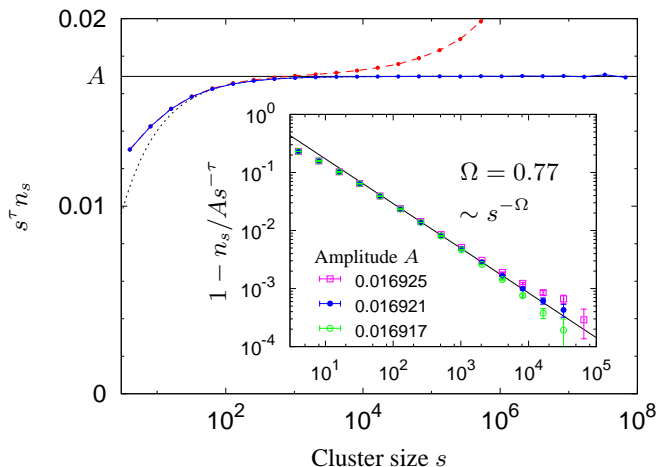
K. Binder and W. Kob, *Glassy Materials and Disordered Solids: An Introduction to Their Statistical Mechanics*



## Simulation based on Hoshen-Kopelman algorithm

- periodic boundary conditions reduce finite size correction
- box length  $L = 45,000$
- realizations 195,000
- critical density  $p_c = 0.5927460$

# power-law corrections

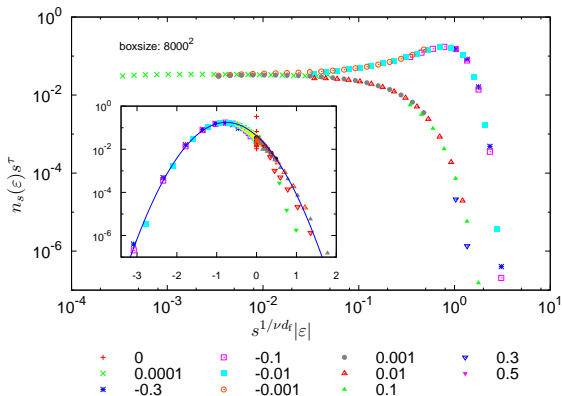


A. Kammerer,  
F. Höfling, and  
T. Franosch,  
EPL **84** (2008)  
66002

power-law corrections

$$n_s(p_c) = A s^{-\tau} (1 + B s^{-\Omega} + \dots) \quad s \rightarrow \infty$$

new universal correction exponent  $\Omega = 0.77$



## Scaling behavior

- distance  
 $\epsilon = (p - p_c)/p_c$
- all cluster alike
- compare size  
 $R_s \sim s^{1/d_f}$   
with correlation  
length  $\xi \sim |\epsilon|^{-\nu}$

$$n_s(\epsilon) = s^{-\tau} \hat{n}(\epsilon s^{1/\nu d_f})$$

$\hat{n}$ : scaling function, excellent data collapse

# Ant in the Labyrinth

## Transport on percolating systems

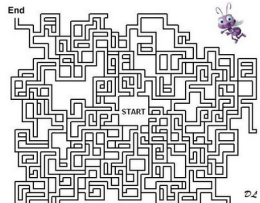
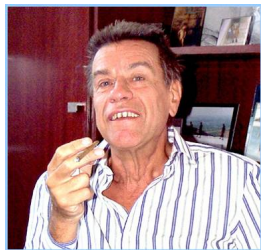
- random walker on occupied sites
  - ant in the labyrinth (de Gennes)
- fractal geometry causes anomalous transport
- mean-square displacement
  - all cluster average

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

- infinite cluster

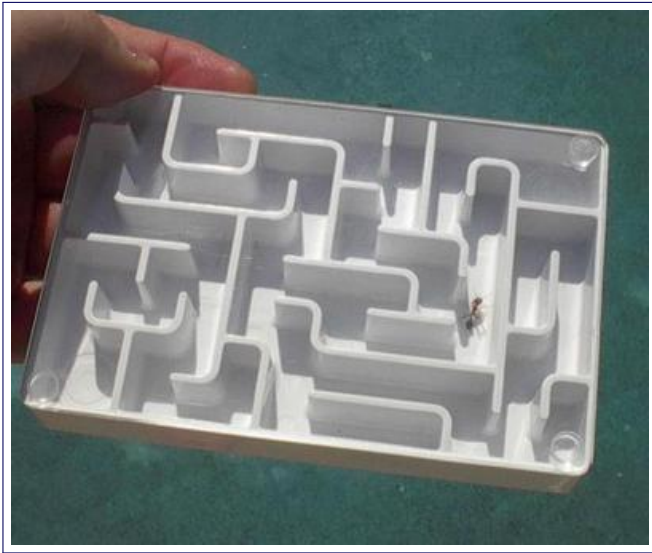
$$\delta r_{\infty}^2(t) \sim t^{2/d_w}$$

- **subdiffusive**
- up to where the system is homogeneous



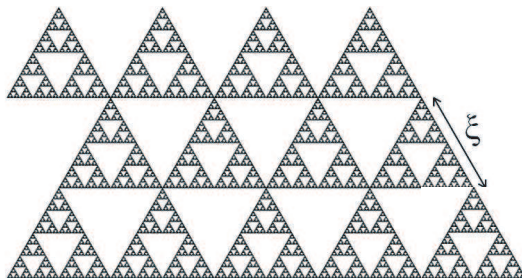


# Ant Farmer John



[www.AntFarmerJohn.com](http://www.AntFarmerJohn.com)

# Crossover to homogeneous System



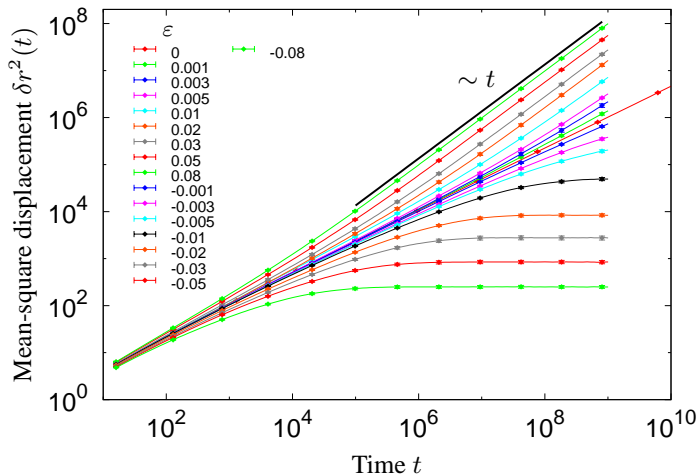
- crossover to diffusion at scale  $\xi$

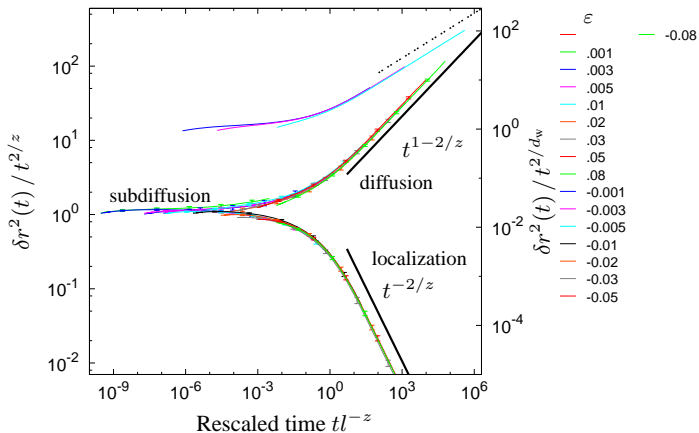
$$\delta r_{\text{Sierpiński}}^2 \sim \begin{cases} t^{2/d_w} & \text{anomalous for } t \ll t_\xi \\ t & \text{diffusive for } t \gg t_\xi \end{cases}$$

- crossover time  $t_\xi \sim \xi^{d_w/2}$
- walk dimension for Sierpiński  $d_w = \log 5 / \log 2$

D. ben Avraham and S. Havlin

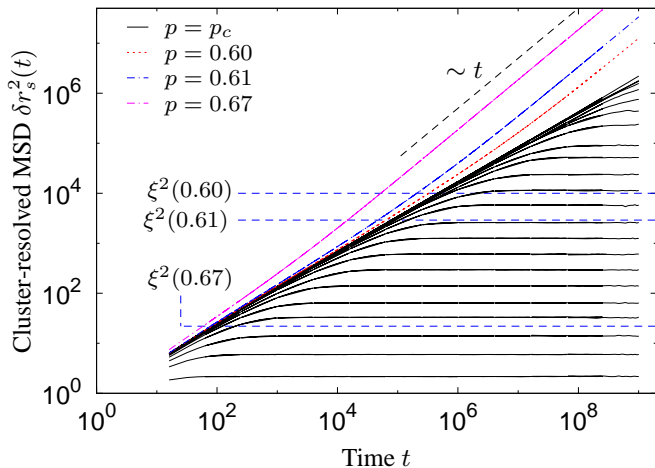
# mean-square displacement





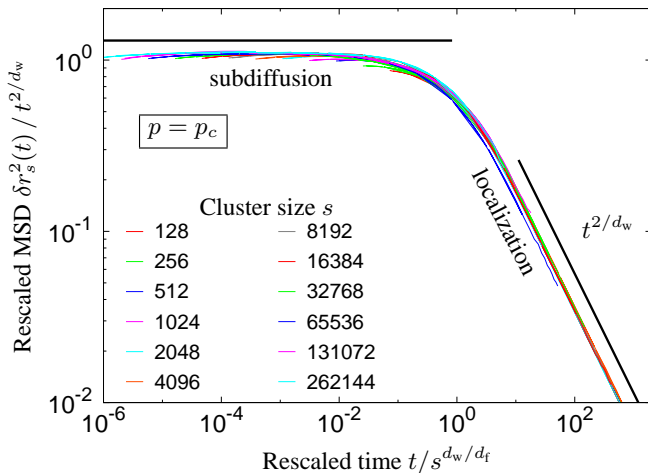
$$\text{Scaling: } \delta r^2(t; \epsilon) = t^{2/z} \delta \hat{r}_{\pm}^2(tl^{-z})$$

# cluster resolved transport



$$\delta r_s^2(t \rightarrow \infty) = R_s^2 \sim s^{2/d_t} \quad \delta r_\infty^2(t) \sim t^{2/d_w}$$

# cluster resolved transport



$$\text{Scaling: } \delta r_s^2(t) = t^{2/d_w} \delta \hat{r}_{\pm}^2(ts^{-d_w/d_f})$$

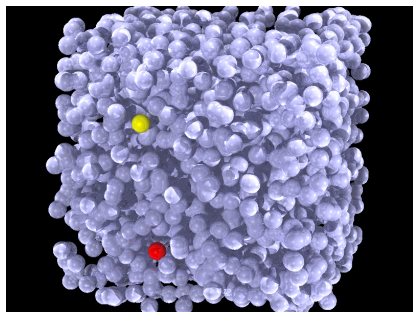
- 1 Motivation
- 2 Ant in the labyrinth
  - fractals
  - percolation
  - transport
- 3 Transport in heterogeneous media
  - Molecular Dynamics simulations
  - Mean-square displacement
  - VACF – two dimensions
  - Continuum Percolation Theory
  - Dynamic Scaling Hypothesis
  - Corrections to scaling

- classical gas of non-interacting, structureless particles
- **randomly** distributed, fixed obstacles:  
→ overlapping hard spheres

*Swiss Cheese model*

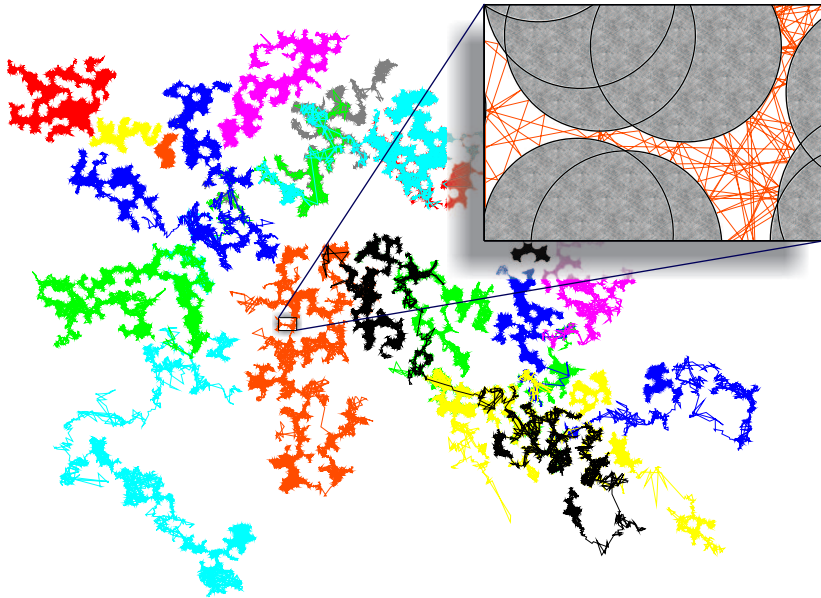


- ballistic motion, elastic scattering  
or **Brownian motion**



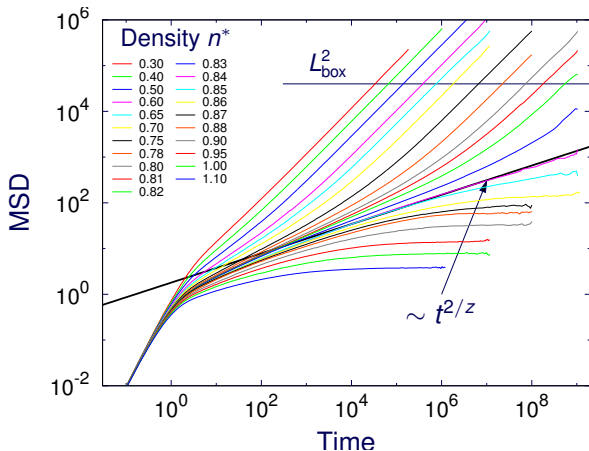
- relevant for transport in **disordered** media
- **single control parameter:**  
reduced obstacle density  
 $n^* = n\sigma^3$  ( $d = 3$ )





# Mean-Square Displacement

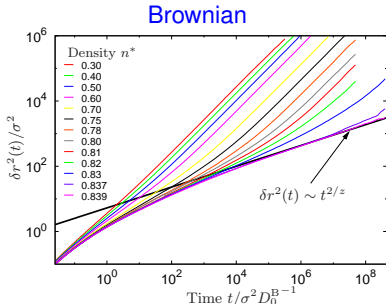
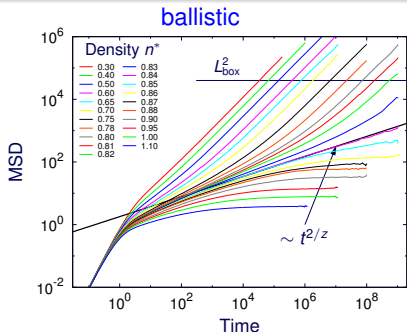
$$\delta r^2(t) = \langle |\mathbf{R}(t) - \mathbf{R}(0)|^2 \rangle \quad (\text{three dimensional system})$$



F. Höfling, T. Franosch, E. Frey, PRL **96**, 165901 (2006)

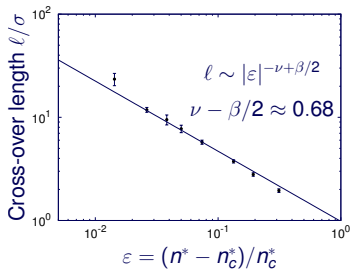
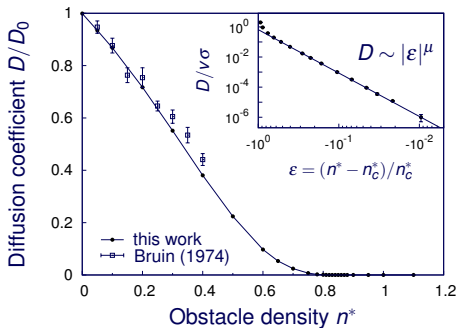
# Mean-Square Displacement

- two regimes for  $t \rightarrow \infty$ 
  - $n^* < n_c^* \rightarrow$  Diffusion  
 $\delta r^2(t) \simeq 6Dt$
  - $n^* > n_c^* \rightarrow$  Localization  
 $\delta r^2(t) \simeq \ell^2$
- close to  $n_c^*$ : intermediate time window until  
 $\delta r^2(t) \approx \ell^2$   
 $\rightarrow$  **subdiffusive** motion,  
 $\delta r^2(t) \sim t^{2/z}$
- at  $n^* = 0.84 \approx n_c^*$ :
  - anomalous diffusion five time decades
  - dynamic exponent  
 $z \approx 6.25$



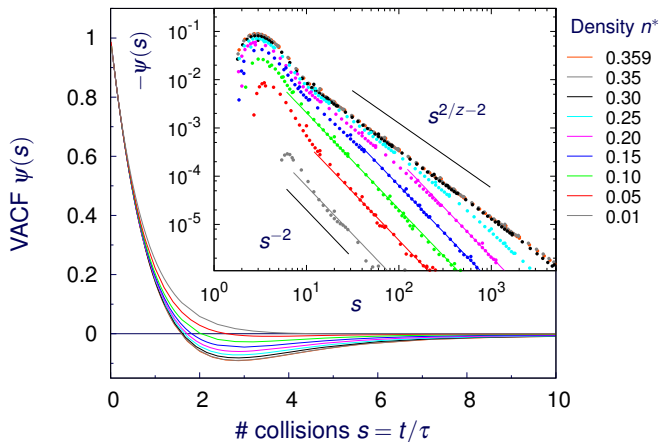
# Diffusion Coefficient

- $D$  vanishes as  $D \sim |\varepsilon|^\mu$ , exponent  $\mu \approx 2.88$
- $\ell$  diverges as  $\ell \sim |\varepsilon|^{-0.68}$
- critical density:  $n_c^* = 0.839(4)$ ,  $\phi_c = 0.9702(5)$



$$D_0 = v\sigma/3\pi n\sigma^3 \text{ (Boltzmann)}$$

# VACF – two dimensions



F. Höfling, T. Franosch, Phys. Rev. Lett. **98**, 140601 (2007)

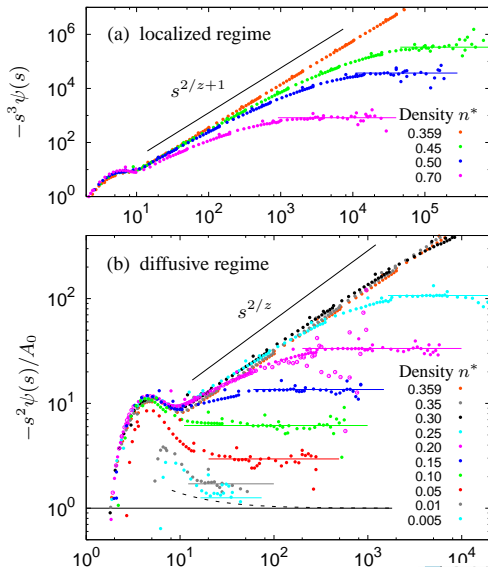
- Noise level  $10^{-7}$  (!), power-law over several decades
- density-dependent exponents or **crossover scenario**?

# VACF – rectification

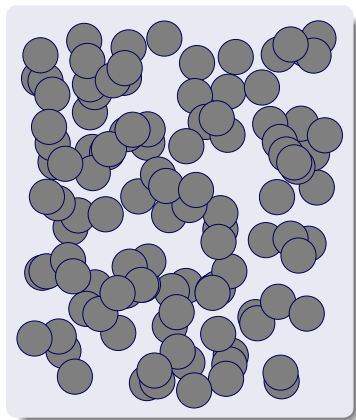
- crossover scenario
- cancellation at intermediate density,  $n^* \approx 0.1$   
Alder and Alley (1978)
- long-time tails in the **localized** regime due to power-law distributed exit rates from cul-de-sac

$$\psi(t) \sim t^{-3} \quad (d=2)$$

Machta and Moore (1985)



# Mapping to a Percolating Network

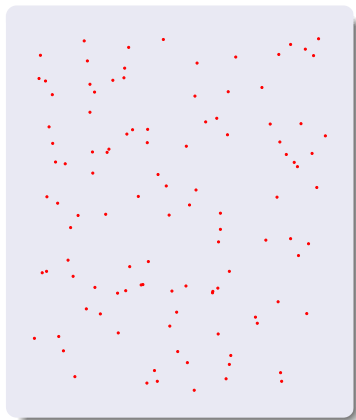


- Voronoi tessellation of the obstacle centers
- gap width  $\rightarrow$  bond strength  $W$
- bonds crossing the obstacles have zero strength
- random resistor network with fractal distribution of weak bonds,

$$\rho(W) \sim W^{-\alpha} \text{ for } W \rightarrow 0$$

Elam, Kerstein, and Rehr (1984)

# Mapping to a Percolating Network



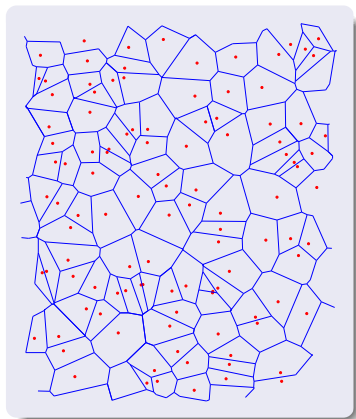
- Voronoi tessellation of the obstacle centers
- gap width  $\rightarrow$  bond strength  $W$
- bonds crossing the obstacles have zero strength
- random resistor network with fractal distribution of weak bonds,

$$\rho(W) \sim W^{-\alpha} \text{ for } W \rightarrow 0$$

Elam, Kerstein, and Rehr (1984)



# Mapping to a Percolating Network

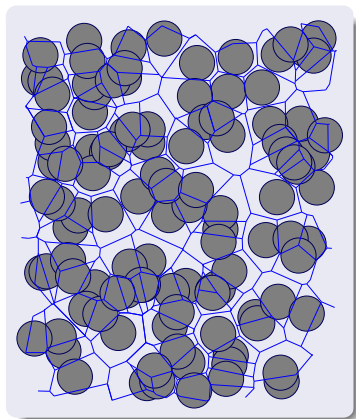


- Voronoi tessellation of the obstacle centers
- gap width  $\rightarrow$  bond strength  $W$
- bonds crossing the obstacles have zero strength
- random resistor network with fractal distribution of weak bonds,

$$\rho(W) \sim W^{-\alpha} \text{ for } W \rightarrow 0$$

Elam, Kerstein, and Rehr (1984)

# Mapping to a Percolating Network

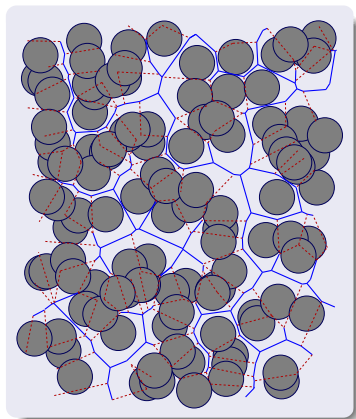


- Voronoi tessellation of the obstacle centers
- gap width  $\rightarrow$  bond strength  $W$
- bonds crossing the obstacles have zero strength
- random resistor network with fractal distribution of weak bonds,

$$\rho(W) \sim W^{-\alpha} \text{ for } W \rightarrow 0$$

Elam, Kerstein, and Rehr (1984)

# Mapping to a Percolating Network

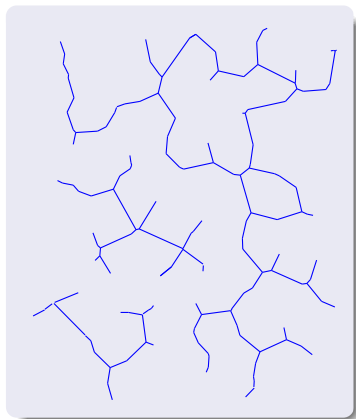


- Voronoi tessellation of the obstacle centers
- gap width  $\rightarrow$  bond strength  $W$
- bonds crossing the obstacles have zero strength
- random resistor network with fractal distribution of weak bonds,

$$\rho(W) \sim W^{-\alpha} \text{ for } W \rightarrow 0$$

Elam, Kerstein, and Rehr (1984)

# Mapping to a Percolating Network



- Voronoi tessellation of the obstacle centers
- gap width  $\rightarrow$  bond strength  $W$
- bonds crossing the obstacles have zero strength
- random resistor network with fractal distribution of weak bonds,

$$\rho(W) \sim W^{-\alpha} \text{ for } W \rightarrow 0$$

Elam, Kerstein, and Rehr (1984)

# Results of Continuum Percolation Theory

- critical exponents:

$$P \sim |n - n_c|^\beta, \quad \xi \sim |n - n_c|^{-\nu}, \quad D \sim |n - n_c|^\mu$$

- mean-cluster radius:  $\ell \sim |n - n_c|^{-\nu + \beta/2}$

- scaling relation:  $z - 2 = \mu / (\nu - \beta/2)$

- geometric exponents  $\nu$  and  $\beta$  are universal for lattice and continuum percolation Elam, Kerstein, and Rehr (1984)

- dynamical exponents as  $z$  and  $\mu$  not weak conductances **dominate** or **irrelevant** Halperin, Feng, and Sen (1985)

- hyperscaling relation:

$$\mu = (d - 2)\nu + 1 / (1 - \alpha) > \mu^{\text{lat}}, \quad (\alpha \text{ sufficiently large})$$

Straley (1982); Stenull and Janssen (2001)

# Results of Continuum Percolation Theory

- critical exponents:

$$P \sim |n - n_c|^\beta, \quad \xi \sim |n - n_c|^{-\nu}, \quad D \sim |n - n_c|^\mu$$

- mean-cluster radius:  $\ell \sim |n - n_c|^{-\nu + \beta/2}$

- scaling relation:  $z - 2 = \mu / (\nu - \beta/2)$

- geometric exponents  $\nu$  and  $\beta$  are universal for lattice and continuum percolation Elam, Kerstein, and Rehr (1984)

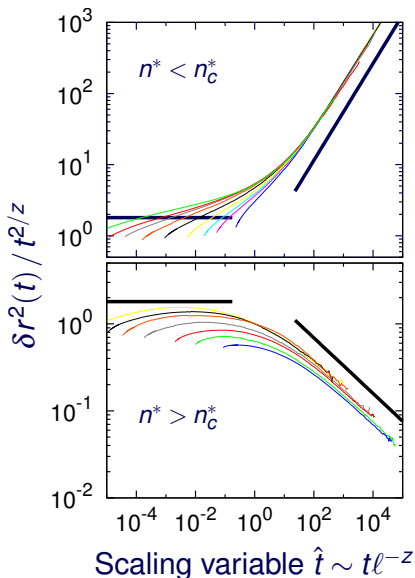
- dynamical exponents as  $z$  and  $\mu$  not weak conductances **dominate** or **irrelevant** Halperin, Feng, and Sen (1985)

- hyperscaling relation (3D Lorentz model):

$$\mu = \nu + 2 \quad (\alpha = \frac{1}{2})$$

Machta and Moore (1985)

# Testing the Dynamic Scaling Ansatz



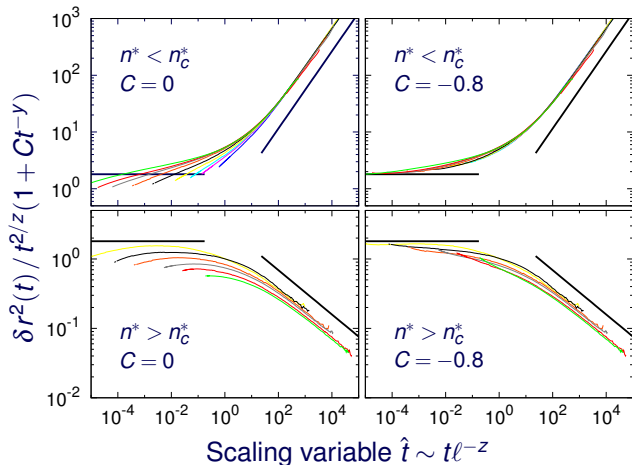
$$\delta r^2(t; \varepsilon) = t^{2/z} \delta \hat{r}_{\pm}^2(\hat{t})$$

- excellent data collapse in the diffusive regime
- rapid convergence towards large- $\hat{t}$  asymptotes
- small  $\hat{t}$ : asymptotic convergence as  $n^* \rightarrow n_c^*$
- corrections to scaling relevant for  $\hat{t} \ll 1$
- **apparent** density-dependent exponents

# Corrections to Scaling

corrections to scaling approximately

$$\delta r^2(t; \varepsilon) = t^{2/z} \delta \hat{r}_{\pm}^2(\hat{t}) (1 + Ct^{-y})$$



- new **universal** correction exponent  $y$
- data at  $n^* = 0.84$ :  $0.15 \lesssim y \lesssim 0.4$
- scaling plots for  $y = 0.34$  and  $C = -0.8$



- Corrections to scaling for the cluster distribution at criticality

$$n_s(\varepsilon = 0) = s^{-d/d_f - 1} [A + Bs^{-\Omega}]$$

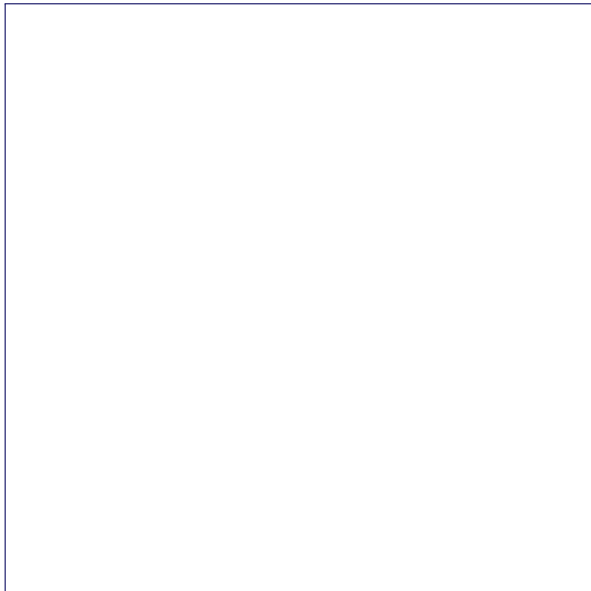
- Extensive Monte Carlo Simulation for Lattice Percolation  
 $\Omega = 0.64 \pm 0.02$
- Extended scaling hypothesis

$$y = \frac{d_f}{d_w} \Omega = \frac{\nu d - \beta}{z(\nu - \beta/2)} \Omega$$

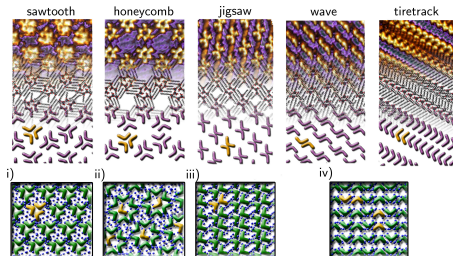
→  $y = 0.34$

## Anomalous Transport

- **Subdiffusion** quite common
  - heterogeneous media, strong size disparities
  - obstructed motion
- **Lorentz model** generic model for anomalous transport
  - Origin: **fractal nature** of the clusters
  - continuum percolation, random resistor network
  - mean-square displacement, non-Gaussian parameter
  - **also**: VACF (long-time tails), non-fickian diffusion, van Hove correlation function, intermediate scattering function,
- **exponents and scaling**
  - universality fixes exponents
  - large crossover regimes
    - **apparent** density-dependent exponents
  - analogy to **molecular crowding**

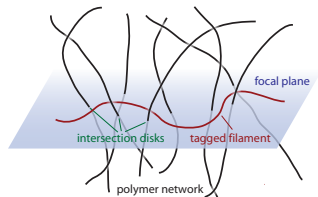


- molecular self-assembly



C. Rohr, M. Balbas-Gambra,  
K. Gruber, E. Constable, E. Frey,  
T. Franosch, B. Hermann

- entangled dynamics of a stiff rod

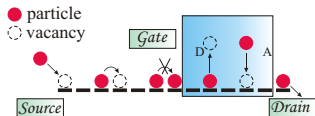
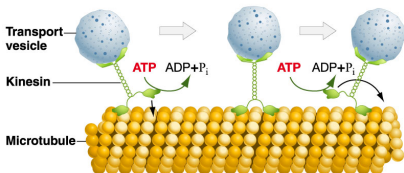


F. Höfling, E. Frey, and T. Franosch,  
PRL **101**, 120605(2008)  
T. Munk, F. Höfling, E. Frey, and  
T. Franosch, EPL **85**, 30003 (2009)

# Further Interests – Driven Transport

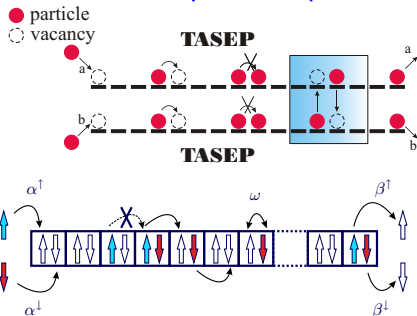
## Molecular Motors

Kinesin "walks" along a microtubule track



A. Parmeggiani, T. Franosch, E. Frey,  
PRL **90**, 086601 (2003)

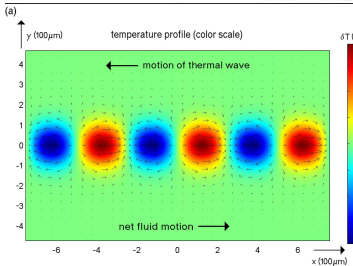
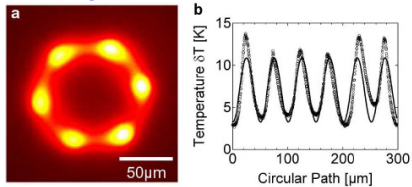
## Classical Spin Transport



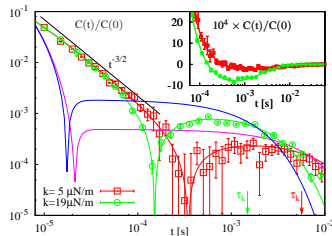
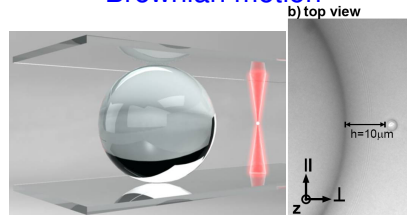
T. Reichenbach, T. Franosch, E. Frey,  
PRL **97**, 050603(2006).

# Further Interests – Hydrodynamics at Microscales

## Light-induced fluid flow



## Brownian motion



F.M. Weinert, J.A. Kraus, T. Franosch,  
D. Braun, PRL (2008)

S. Jeney, B. Lukić, J.A. Kraus,  
T. Franosch, L. Főrro, PRL (2008)

- **Anomalous transport**
  - Felix Höfling, Tobias Munk, Axel Kammerer (LMU München)
  - Joachim Rädler, Margaret Horton, Doris Heinrich (LMU München)
  - Thomas Voigtmann, Jürgen Horbach, Matthias Sperl, Andreas Mayer (DLR Cologne)
- **Nonequilibrium phase transitions**
  - Andrea Parmeggiani (Montpellier 2), Paolo Pierobon (Institut Curie), Tobias Reichenbach, Mauro Mobilia, Anna Melbinger (LMU München)
- **Order phenomena and self-assembly**
  - Andreja Šarlah (University of Ljubljana)
  - Bianca Hermann, Marta Balbás Gandra (LMU München)
- **Fluid dynamics on micro- and nano-scales**
  - Franz Weinert, Dieter Braun, Jonas Kraus (LMU München)
  - Branimir Lukić, Sylvia Jeney (EPFL)

Lehrstuhl Erwin Frey (LMU München)

# Appendix



## Two dimensional lattice percolation

- fractal dimension  $d_f = 91/48 \approx 1.90$
- correlation length  $\nu = 4/3$
- infinite cluster  $\beta = 5/36$
- Fisher exponent  $\tau = 187/91$

- **Non-equilibrium Transport**
  - Andrea Parmeggiani (CNRS-Université Montpellier 2)
  - Paolo Pierobon, Tobias Reichenbach, Mauro Mobilia, Anna Melbinger (LMU München)
- **Rods & Needles**
  - **Felix Höfling** (Hahn-Meitner Institute, LMU München)
  - Tobias Munk (LMU München)
- **Ordering in molecular crystals**
  - Andreja Šarlah (University of Ljubljana), Clemens Bechinger (University of Stuttgart)
  - Bianca Hermann, Marta Balbás Gamba LMU München (*new*)
- **Fluidics at the microscale** (*new*)
  - Dieter Braun, Jonas Kraus LMU München, Sylvia Jeney EPFL

*and for collaboration and continuous support*

**Erwin Frey** LMU München

# Van Hove self-correlation function

$$G(\mathbf{r}, t) := \langle \delta(\mathbf{R}(t) - \mathbf{R}(0) - \mathbf{r}) \rangle$$

- probability distribution of the particle positions
- diffusive peak vanishes
- sharp peak at a definite distance remains

vanHove-0,75

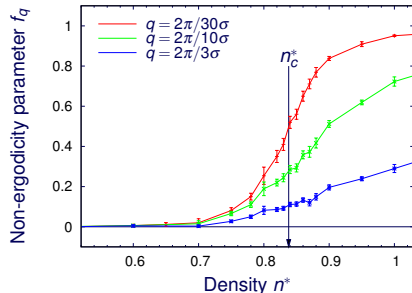
consider long-time limit:

- $G(\mathbf{r}, t \rightarrow \infty) \equiv 0 \rightarrow$  all particles can diffuse away
- finite peak  $\rightarrow$  localization of some particles
- coexistence of localized and diffusing particles below  $n_C^*$ , phase space is decomposed into finite and infinite subsets

# Non-ergodicity Parameters $f_q$

$$\Phi_q^S(t) := \langle e^{i\mathbf{q}\cdot(\mathbf{R}(t)-\mathbf{R}(0))} \rangle$$

- Fourier transform of  $G(\mathbf{r}, t)$
- incoherent inelastic scattering function  $\Phi_q^S(t)$
- non-ergodicity parameter:  
 $f_q := \Phi_q^S(t \rightarrow \infty)$



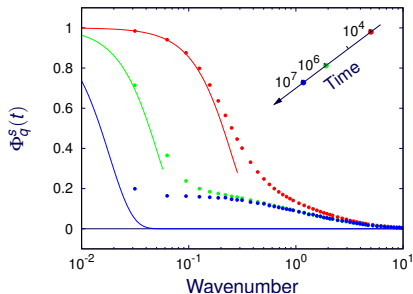
- $f_q > 0$  at *all* densities  
→ phase space is always decomposed
- transition affects  $f_q$  in next-to-leading order only:

$$f_q \sim \text{const} + |\varepsilon|^\beta$$

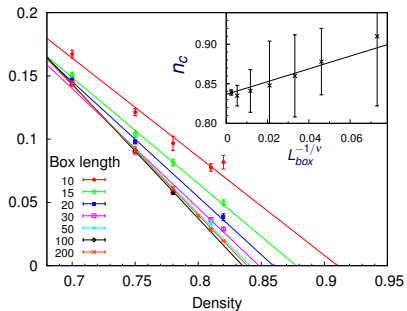
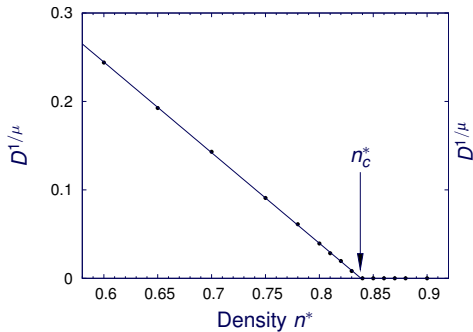
Kertész and Metzger (1983)

$$\Phi_q^S(t) := \langle e^{i\mathbf{q}\cdot(\mathbf{R}(t)-\mathbf{R}(0))} \rangle$$

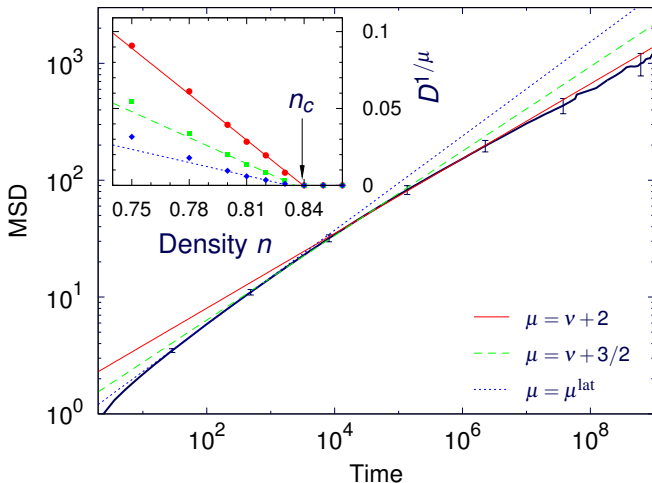
- Fourier transform of  $G(\mathbf{r}, t)$
- incoherent inelastic scattering function  $\Phi_q^S(t)$
- non-ergodicity parameter:  
 $f_q := \Phi_q^S(t \rightarrow \infty)$



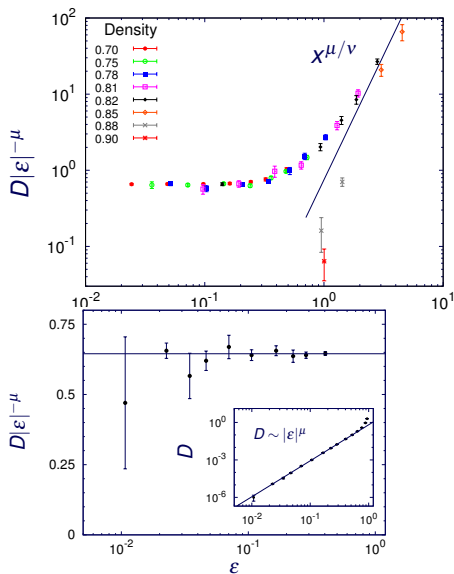
- Gaussian approximation:  
 $\Phi_q^S(t) \approx e^{-Dq^2t} \rightarrow f_q$  should vanish in diffusive systems
- in presence of non-Gaussian corrections:  
valid only for  $q \ll 1/\sqrt{Dt} \rightarrow$  breaks down as  $t \rightarrow \infty$



$\mu$	$n_c^*$	$\Delta n_c^*$
2.87	0.8388	0.0041
2.88	0.8390	0.0040
2.89	0.8392	0.0040



- $\mu = (d - 2)\nu + 1/(1 - \alpha)$
- compatible only with Machta and Moore
- $\alpha = 1/2$



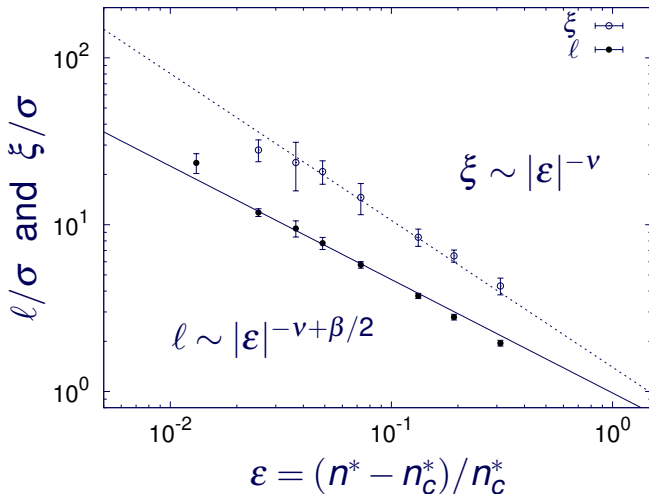
- Finite-size scaling prediction

$$D(\varepsilon; L) = |\varepsilon|^\mu \hat{D}^\pm(\xi/L)$$

- large boxes  
 $D(\varepsilon < 0; L \gg \xi) \sim |\varepsilon|^\mu$
- small boxes  
 $D(\varepsilon; L \ll \xi) \sim L^{-\mu/\nu}$

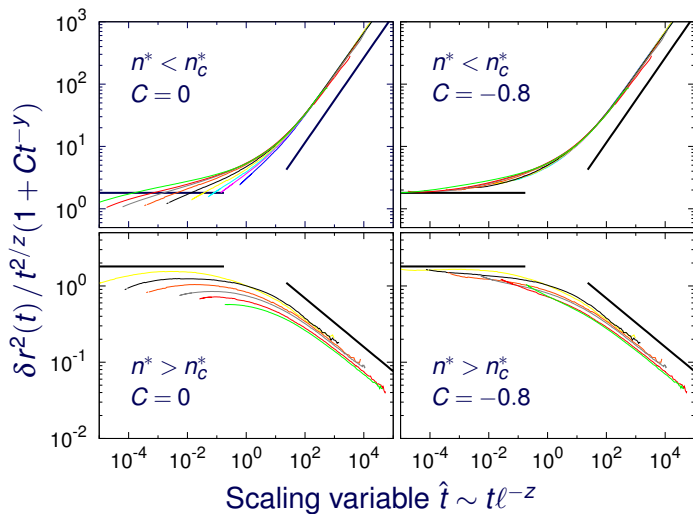


# Correlation Length



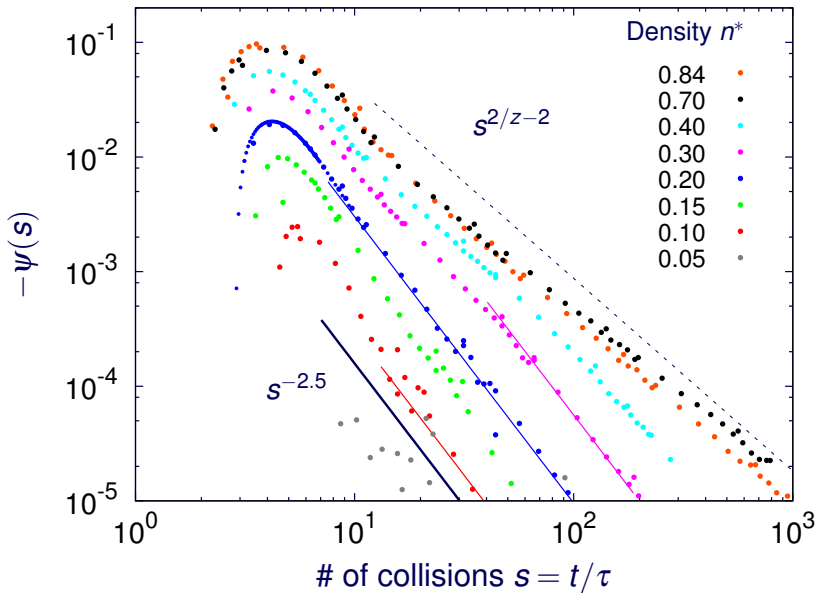
- Divergence of the correlation length
- $\xi$  extracted from non-gaussian parameter

# Corrections to Scaling



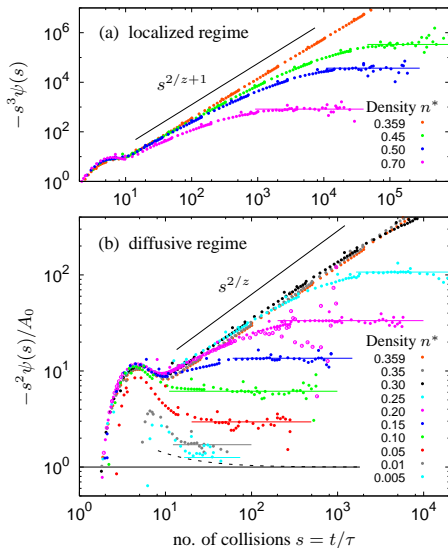
Correction-to-scaling exponent  $y = 0.34$

# VACF in 3d

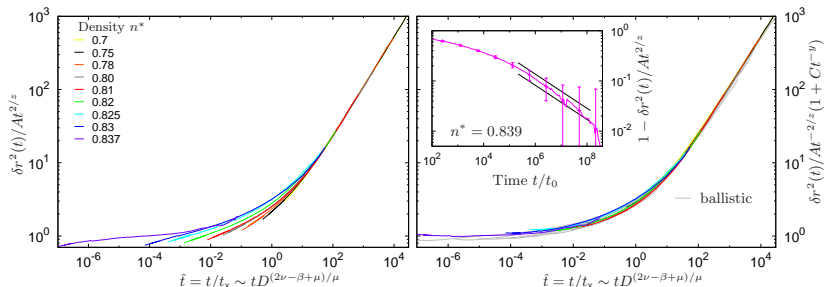


# Long-time tails

- rectification, **sensitive test**
- crossover scenario  
Götze, Leutheusser, and Yip (1981)
- cancellation effects
- growth close to  $n_c$
- low-density: difficult
- predicted tail in localized regime  
 $\psi(t) \propto t^{-3}$   
Machta and Moore (1985)
- prediction for super-Burnett, non-gaussian parameter
- 3d ..

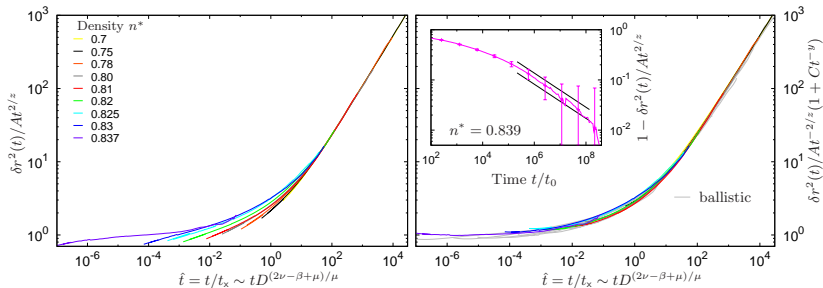


# Testing the Dynamic Scaling Ansatz



- leading order scaling  
 $\delta r^2(t; \varepsilon) = t^{2/z} \delta \hat{r}_{\pm}^2(\hat{t})$
- excellent data collapse  
in the **diffusive** regime
- small  $\hat{t}$ : asymptotic  
convergence as  
 $n^* \rightarrow n_C^*$

# Testing the Dynamic Scaling Ansatz



- leading order scaling  
 $\delta r^2(t; \varepsilon) = t^{2/z} \delta \hat{r}_{\pm}^2(\hat{t})$
- excellent data collapse in the **diffusive** regime
- small  $\hat{t}$ : asymptotic convergence as  $n^* \rightarrow n_c^*$

- extend scaling by irrelevant coupling:  
 $\delta r^2(t; \varepsilon) = t^{2/z} \delta \hat{r}_{\pm}^2(\hat{t}) [1 + t^{-y} \Delta_{\pm}(\hat{t})]$
- new **universal** correction exponent  $y$
- at criticality ( $\hat{t} = 0$ ):  
 $\delta r^2(t; \varepsilon) \propto t^{2/z} (1 + C t^{-y})$
- **approximate** correction function  
 $\Delta_{\pm}(\hat{t}) = C$
- scaling plots for  $y = 0.34$

# Non-Gaussian Parameter

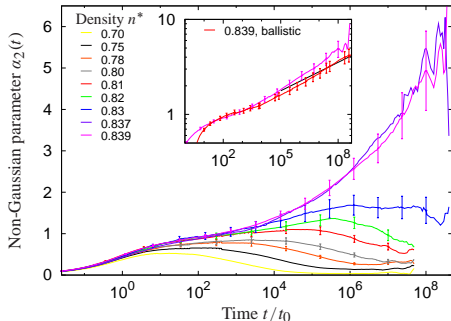
- mean-quartic displacement,  $\delta r^4(t; \varepsilon) = \int d\mathbf{r} r^4 G(\mathbf{r}, t; \varepsilon)$ :

$$\delta r^4(t) \sim t^{4/\tilde{z}} \quad (\varepsilon = 0), \quad \delta r^4(t \rightarrow \infty) \sim \begin{cases} \xi^2 \ell^2 & (\varepsilon > 0) \\ (Dt)^2 |\varepsilon|^{-\beta} & (\varepsilon < 0) \end{cases}$$

- different exponent  $\tilde{z} \approx 5.4 \neq z$

$$\alpha_2(t) := \frac{3}{5} \delta r^4(t) / [\delta r^2(t)]^2 - 1$$

- sensitive to **heterogeneities**
- diffusive regime:  $\alpha_2(\infty) > 0$
- critical law:  
 $\alpha_2(t) \sim t^{4/\tilde{z} - 4/z} \sim t^{0.097}$   
**divergent**



Brownian particles

- van Hove self-correlation function  $G(\mathbf{r}, t) = \langle \delta(\mathbf{R}(t) - \mathbf{R}(0) - \mathbf{r}) \rangle$   
→ Probability to travel distance  $\mathbf{r}$  in time  $t$

$$G(\mathbf{r}, t; \varepsilon) = \xi^{-\beta/\nu-d} \mathcal{G}_{\pm}(\mathbf{r}/\xi, tl^{-z})$$

- two diverging length scales:
  - correlation length  $\xi \sim |\varepsilon|^{-\nu}$  rescales geometry
  - cross-over length  $\ell \sim |\varepsilon|^{-\nu+\beta/2}$  rescales time
  - **three non-trivial exponents** → no CTRW, fractional FP, ...
- scaling ansatz for the MSD from  $\delta r^2(t; \varepsilon) = \int d\mathbf{r} r^2 G(\mathbf{r}, t; \varepsilon)$ :

$$\delta r^2(t; \varepsilon) = t^{2/z} \delta \hat{r}_{\pm}^2(\hat{t}), \quad \text{where } \hat{t} \sim tl^{-z}$$

- critical dynamics ( $\hat{t} = 0$ ) recovered:  $\delta r^2(t) \sim t^{2/z}$ .



# Corrections to Scaling

- extend scaling ansatz by including an irrelevant coupling:

$$\delta r^2(t; \varepsilon) = t^{2/z} \delta \hat{r}_{\pm}^2(\hat{t}) (1 + t^{-y} \Delta_{\pm}(\hat{t}))$$

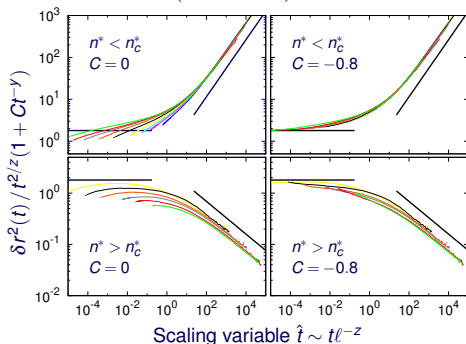
- new **universal** correction exponent  $y$

- at criticality ( $\hat{t} = 0$ ):  $\delta r^2(t; \varepsilon) \propto t^{2/z} (1 + C t^{-y})$

- data at  $n^* = 0.84$ :  
 $0.15 \lesssim y \lesssim 0.4$

- **approximate** correction function  $\Delta_{\pm}(\hat{t}) = C$

- scaling plots for  
 $y = 0.34$  and  $C = -0.8$



## Lorentz Model

- Molecular Dynamics simulations
  - first **accurate** data in the relevant regime
  - mean-square displacement, non-Gaussian parameter
  - **also**: VACF (long-time tails), non-fickian diffusion, van Hove correlation function, intermediate scattering function,
- anomalous transport over several decades
  - Origin: **fractal nature** of the clusters
  - continuum percolation
  - universality class of random resistor networks
- **exponents and scaling**
  - large crossover regimes
    - **apparent** density-dependent exponents
  - significant corrections to scaling
  - analogy to **molecular crowding**