FOR1394 DFG Research Unit
Nonlinear Response
to Probe Vitrification

Active microrheology: Brownian motion, strong forces and viscoelastic media

Matthias Fuchs
Fachbereich Physik, Universität Konstanz

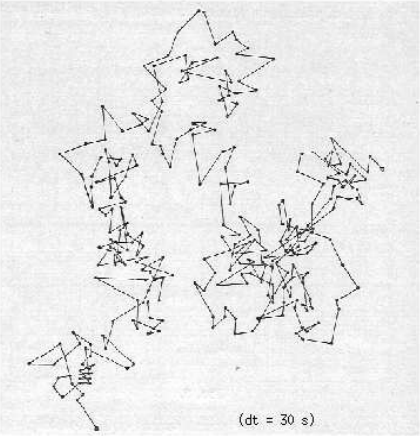
New Frontiers in Non-equilibrium Statistical Physics 2015

Outline

- Intro to dense colloidal dispersions
- Active microrheology: Theory
- Delocalization transition
- Length Scales & Populations
- Steady state motion
- Transient dynamics

Intro to dense colloidal dispersions

Brownian motion



The trajectory of a 0.53 micron particle
J. Perrin "Atoms" 1916

Jean Perrin
http://nobelprize.org/physics/laureates/1926

- Stokes–Einstein–Sutherland:

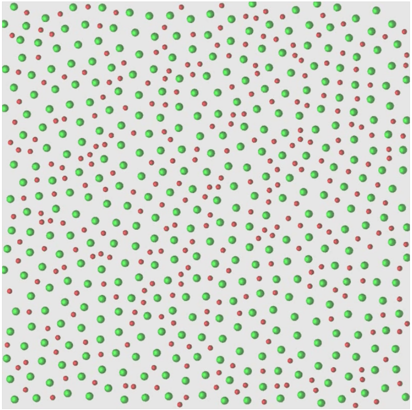
$$\boxed{D_0 = \frac{k_B T}{\zeta_0}}$$

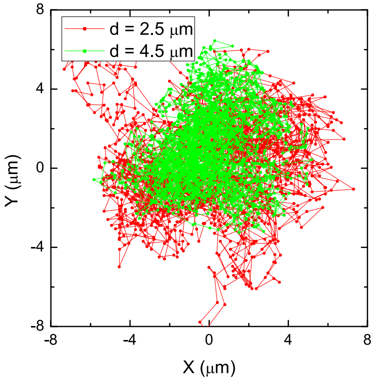
- mean squared displacement

$$\langle (\mathbf{r}(t) - \mathbf{r}(0))^2 \rangle = 6 D_0 t$$

Displacement fluctuations in glass

Binary glass in $d = 2$





finite displacements

$$\langle (\mathbf{r}_i(t) - \bar{\mathbf{r}}_i)^2 \rangle < \infty$$

with center of trajectory:

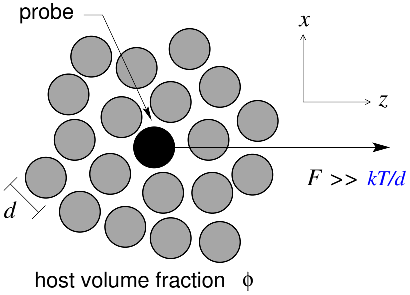
$$\bar{\mathbf{r}}_i = \frac{1}{\Delta t} \int_0^{\Delta t} dt\, \mathbf{r}_i(t)$$

[Klix, Ebert, Weysser, MF, Maret, Keim, Phys. Rev. Lett. **109**, 178301 (2012)]

Active microrheology: Theory

Active microrheology

- Hard-sphere suspension
- Fluid and glass states
- Probe displacement under strong forces
- Tails, heterogeneities etc.
- Parameters: ϕ and F



Microscopic (statistical) description

Model: Interacting Brownian particles, no HI ($D_0 = \frac{k_B T}{\zeta_0}$)

Smoluchowski equation:

$$\partial_t \Psi = \Omega \, \Psi$$

Micro rheology

$$\Omega = \sum_i^N D_0 \, \boldsymbol{\partial}_i \cdot (\boldsymbol{\partial}_i - \beta \mathbf{F}_i) + D_s \, \boldsymbol{\partial}_s \cdot (\boldsymbol{\partial}_s - \beta \mathbf{F}_s) - D_s \, \boldsymbol{\partial}_s \cdot \mathbf{F}^{\mathbf{ex}}$$

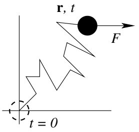
ITT force-velocity relation* (nonlinear, exact)

$$\zeta \langle \mathbf{v} \rangle_{t \to \infty} = F^{\mathrm{ex}} \quad , \quad \boldsymbol{\zeta} = \zeta_0 + \frac{1}{3 k_B T} \int_0^\infty dt \, \langle \mathbf{F}_s \, e^{\Omega_{\mathrm{irr}}^\dagger t} \, \mathbf{F}_s \rangle^{(e)}$$

[∗ Gazuz, Puertas, Voigtmann, MF, Phys. Rev. Lett. **102**, 248302 (2009)]

Microscopic (statistical) description (II)

Mode coupling approximation:



$G_s(\mathbf{r}, t)$ → Fourier transform → $\Phi^s_{\mathbf{q}}(t)$

probe distribution function — probe correlator

Evolution equation:

$$\partial_t \Phi^{\mathsf{s}}_{\mathbf{q}}(t) + \frac{1}{\tau_{\mathbf{q}}(F)} \Phi^{\mathsf{s}}_{\mathbf{q}}(t) + \int_0^t m_{\mathbf{q}}(t - t') \partial_{t'} \Phi^{\mathsf{s}}_{\mathbf{q}}(t') \mathrm{d}t' = 0$$

Parallel relaxation channels*:

$$m_{\mathbf{q}}(t) = \mathscr{F}\{\Phi^{\mathsf{s}}_{\mathbf{q}}(t), \Phi^{\mathsf{host}}_{\mathbf{q}}(t), F\}$$

Input (full MCT):
Equilibrium structure of the host at ϕ

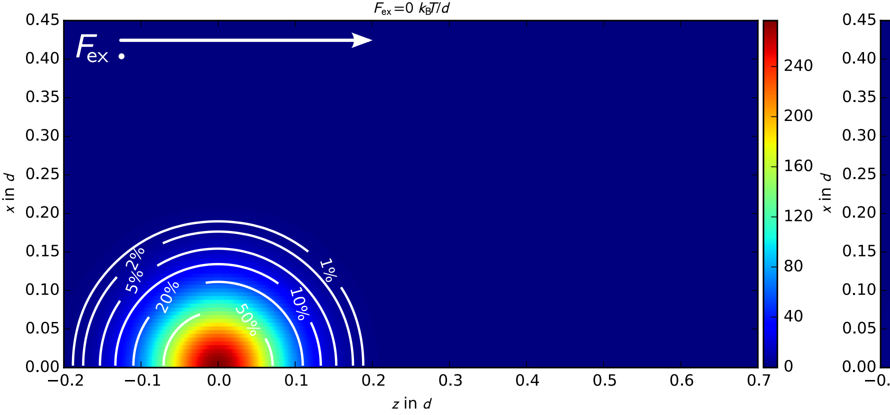
[* Lang et al., PRL **105**, 125701 (2010)]

Delocalization transition

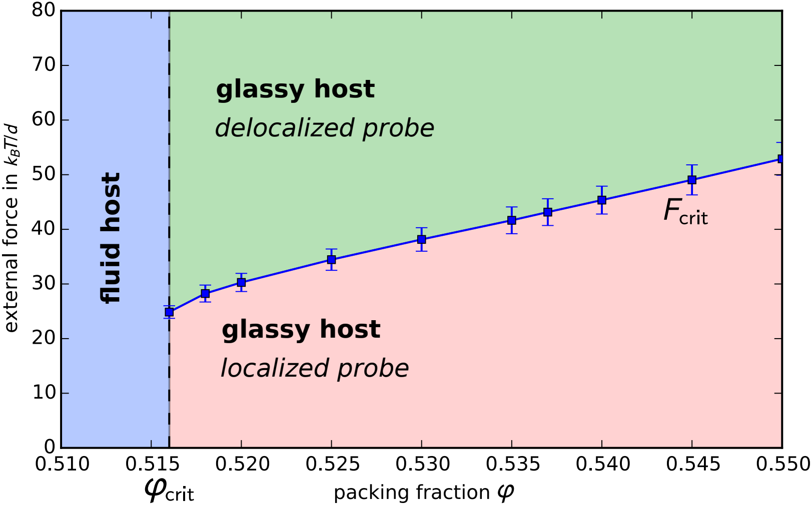
Tracer Nonergodicity Parameter in Real Space

$$f^s(\boldsymbol{r}, t) = \lim_{t\to\infty} \rho^s(\boldsymbol{r}, t)$$

$\varphi = .537$

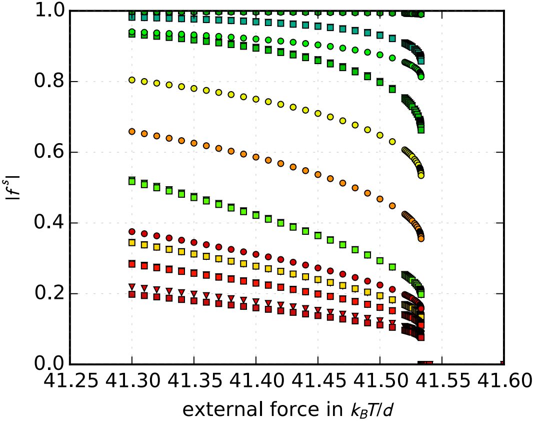


Delocalization transition: phase diagram (MCT)



Delocalization transition

$\phi = 0.537$



- type B transition

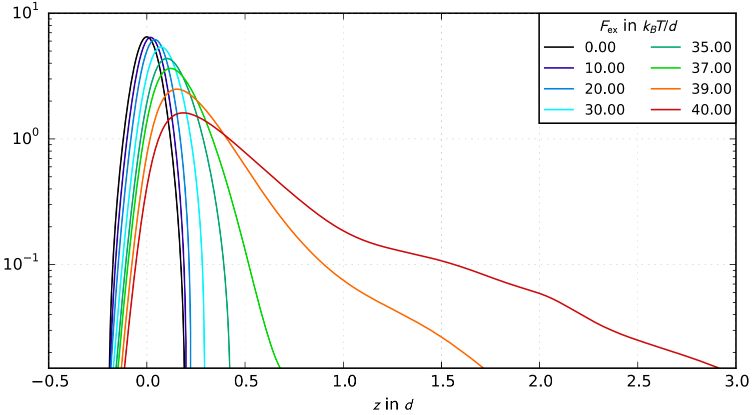
Length Scales & Populations

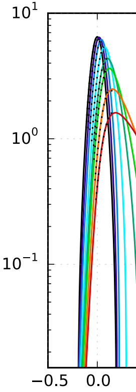
Exponential tails

FOR1394

Glassy host · $F \lesssim$ *threshold* · **localized probe** MCT, $\phi = 0.516$

Marginal probability distribution *parallel* to the force

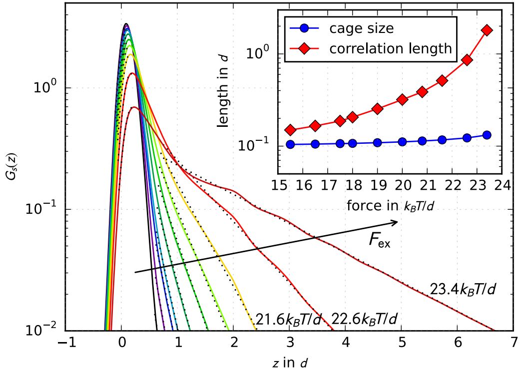




[A. Puertas (U. Almeria), unpublished]

Probe distribution function $G_s(z, t \to \infty)$

Localized probe

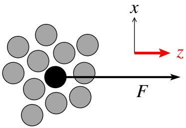


$$G_s^{\text{tail}} \propto \exp\left[-\frac{z}{\xi(F)}\right]$$

Growing correlation length ξ

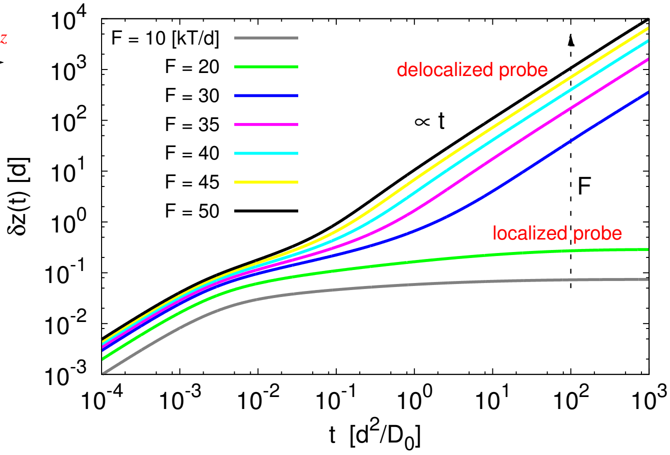
Steady state motion

Probe motion in force direction: average drift $\delta z(t)$



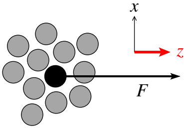
GLASSY HOST

MCT
$\phi = 0.516$



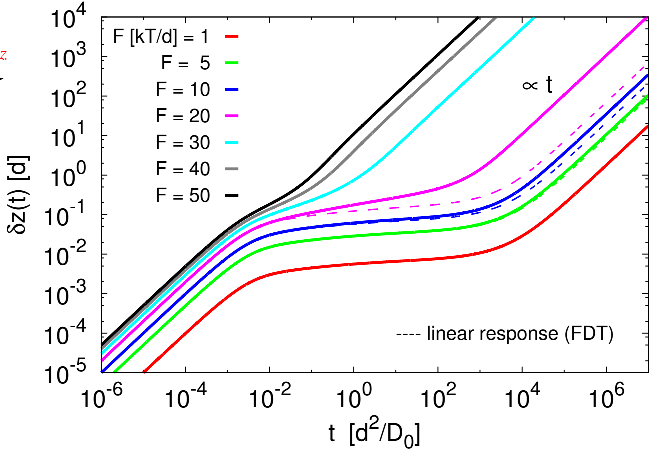
- Delocalization transition

Probe motion in force direction: average drift $\delta z(t)$



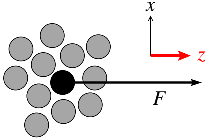
FLUID HOST

MCT
$\phi = 0.515$



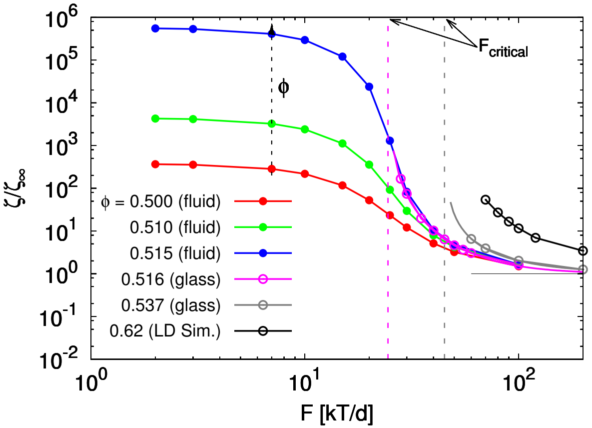
- Steady probe velocity for long times → friction coefficient

Nonlinear friction coefficient: $v = [\zeta(F)]^{-1} F$

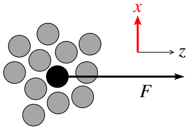


$\zeta_\infty = \zeta(F \to \infty)$

- Force-thinning

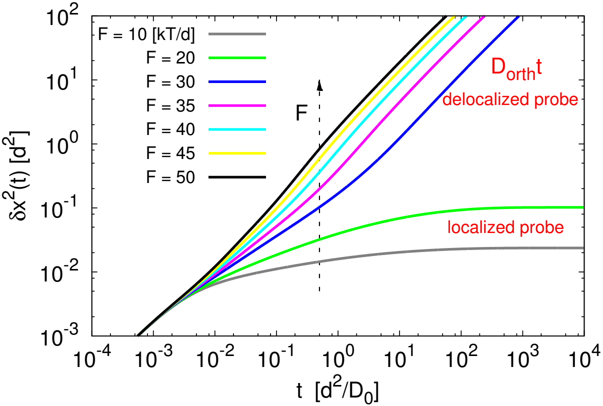


Probe motion in perpendicular direction: $\delta x^2(t)$



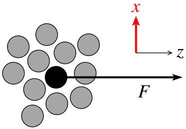
GLASSY HOST

MCT
$\phi = 0.516$

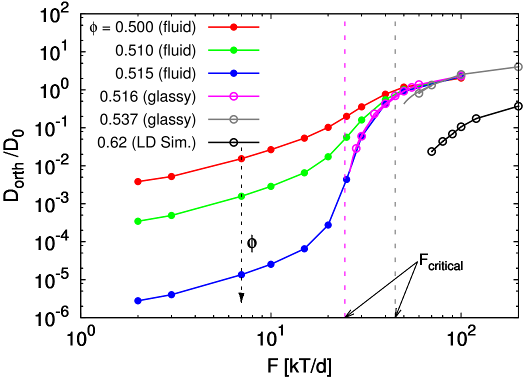


- Delocalization transition
- Diffusive motion for long times

Nonlinear diffusion coefficient - perpendicular direction



$$\delta x^2 \sim D_{\text{orth}} t$$



- Orthogonal diffusion enhanced by increasing F

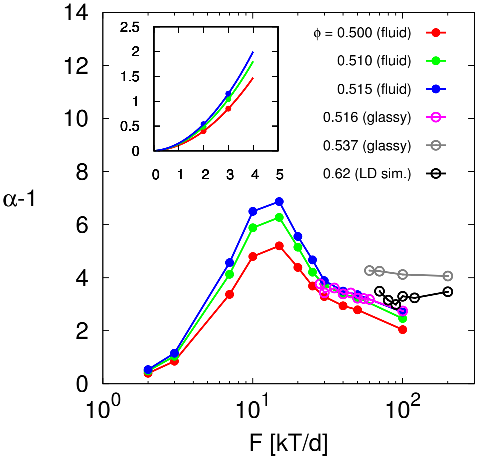
Stokes-Einstein Relation

Equilibrium:

$$\frac{D(\varphi)\zeta(\varphi)}{kT} = 1$$

Nonequilibrium:

$$\frac{D_{orth}(\varphi, F)\zeta(\varphi, F)}{kT} = \alpha(\varphi, F)$$



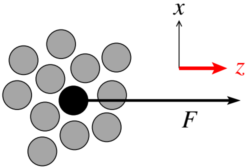
Transient dynamics

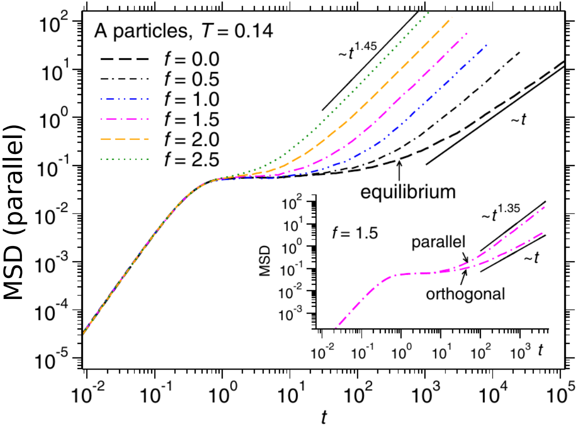
Probe motion in force direction: superdiffusion

MD Simulations - Yukawa mixture
Winter *et al.*, PRL **108** (2012)

MSD

$\delta z^2(t) - [\delta z(t)]^2$





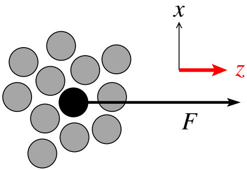
Probe motion in force direction: superdiffusion

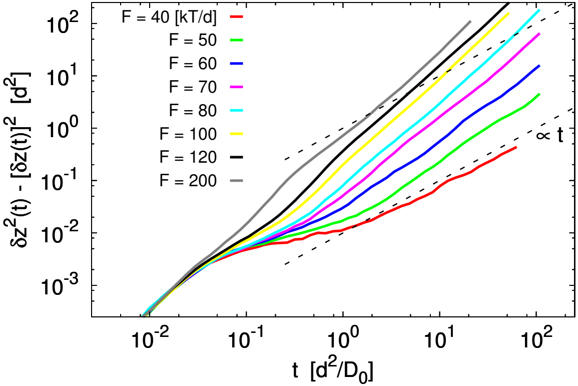
Langevin Dynamics simulations

$\phi = 0.62$

MSD

$\delta z^2(t) - [\delta z(t)]^2$





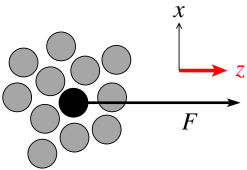
[A. Puertas (U. Almeria), unpublished]

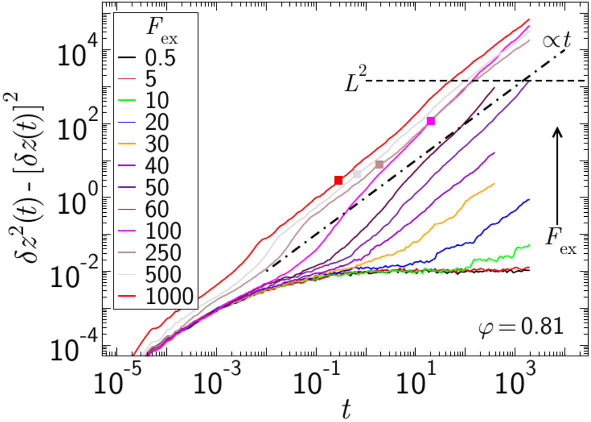
Probe motion in force direction: superdiffusion

Brownian Dynamics simulations (2D)

MSD

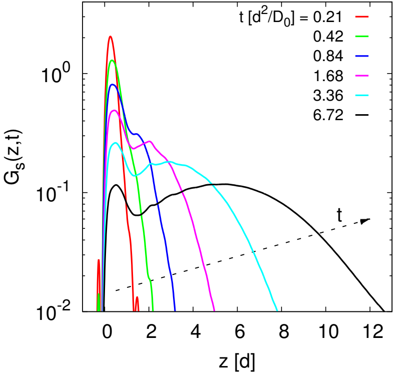
$\delta z^2(t) - [\delta z(t)]^2$





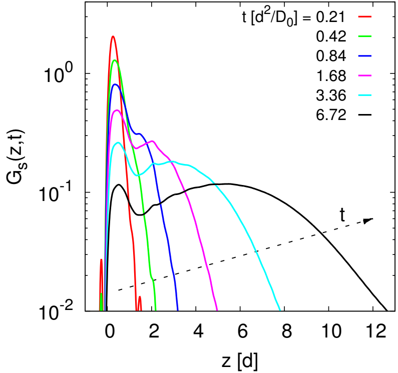
Evolution of the probe distribution function $G_s(z,t)$

Delocalized probe

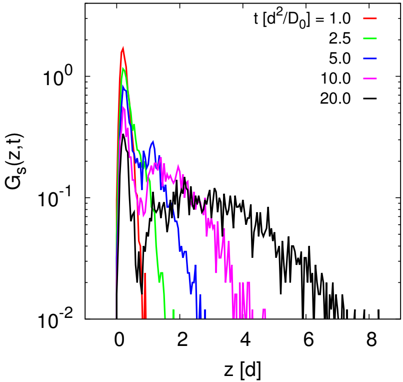


Evolution of $G_s(z,t)$: Delocalized probe

MCT, $\phi = 0.537$, $F = 50kT/d$



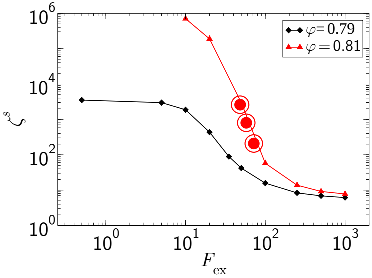
LD Simulations, $\phi = 0.62$, $F = 100kT/d$

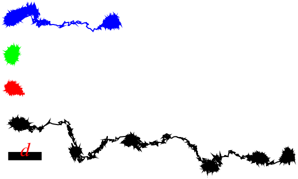


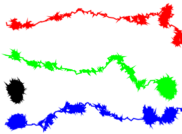
[A. Puertas (U. Almeria), unpublished]

Trajectories

- Brownian dynamics simulations
- individual trajectories in the glassy host
- intermittent dynamics

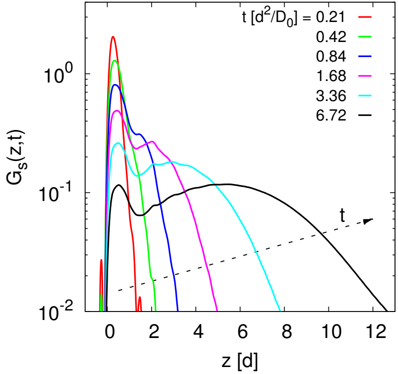






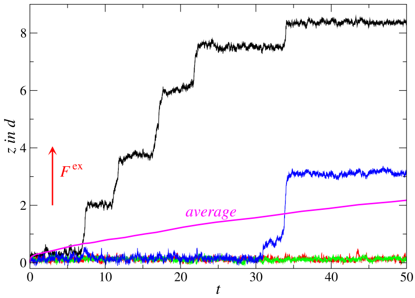
[Harrer, unpublished]

Evolution of $G_s(z,t)$: Delocalized probe

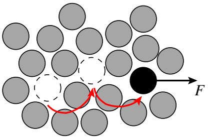


Probe trajectories

Brownian dynamics simulations



- Active and inactive probes



Conclusions

- micro-rheology delocalization threshold $F_c^{\text{ex}} \gg k_B T/\sigma$
- force-induced diffusion $\perp$ force
- "two population" behavior close to depinning
- rare large excursions on diverging length scale

Acknowledgements

- Gustavo Abade, Markus Gruber (Konstanz)
- Antonio Puertas (Almeria)
- Thomas Voigtmann (DLR, Düsseldorf)
- Igor Gazuz, Christian Harrer, Manuel Gnann (Konstanz)
- David Winter, Jürgen Horbach (Düsseldorf)
- movies/discussion: Peter Keim (Konstanz), Stefan Egelhaaf (Düsseldorf), Rut Besseling (Edinburgh), Eric Weeks (Emory)

Deutsche Forschungsgemeinschaft DFG

FOR1394 **DFG Research Unit Nonlinear Response to Probe Vitrification**

Thank you for your attention