UNIVERSITY OF TWENTE.

PCF Twente
Physics of Complex Fluids

(viscous)

dissipation in confined liquid films

Sissi de Beer, Dirk van den Ende, Frieder Mugele

Physics of Complex Fluids

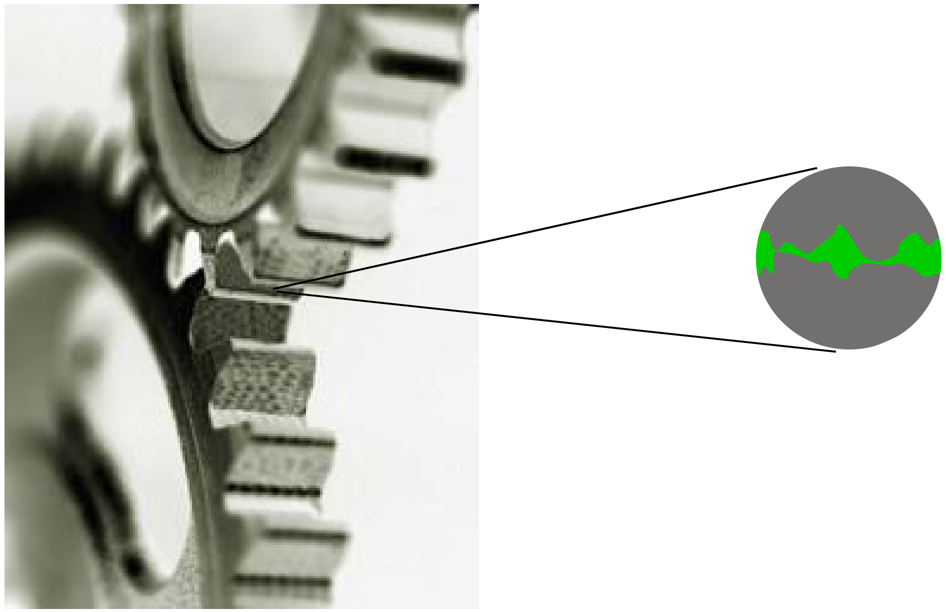
University of Twente

Wouter den Otter, Wim Briels

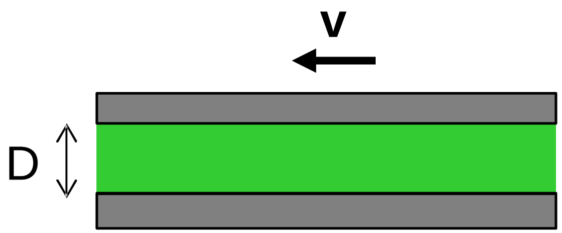
Computational Biophysics

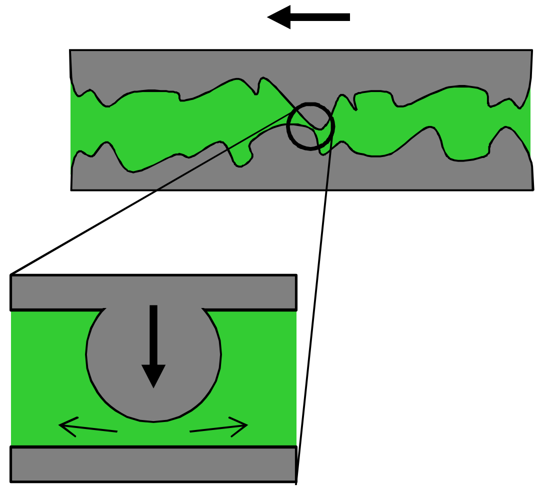
MESA+ INSTITUTE FOR NANOTECHNOLOGY

FOM



nanolubrication





sliding friction
(Couette flow)

$$F_C = \mu A \frac{v}{D}$$

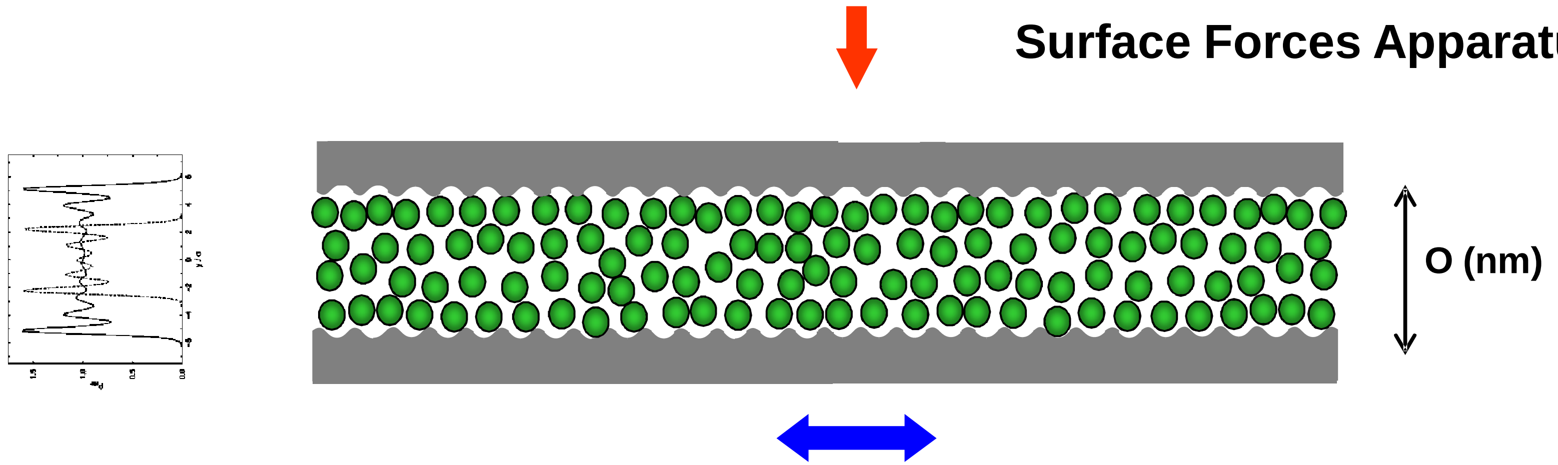
squeeze-out damping
(Reynolds damping)

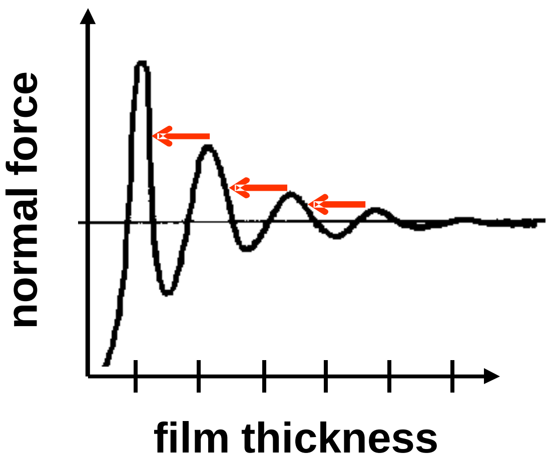
$$F_R = \frac{6\pi \mu R^2}{D}\dot{D}$$

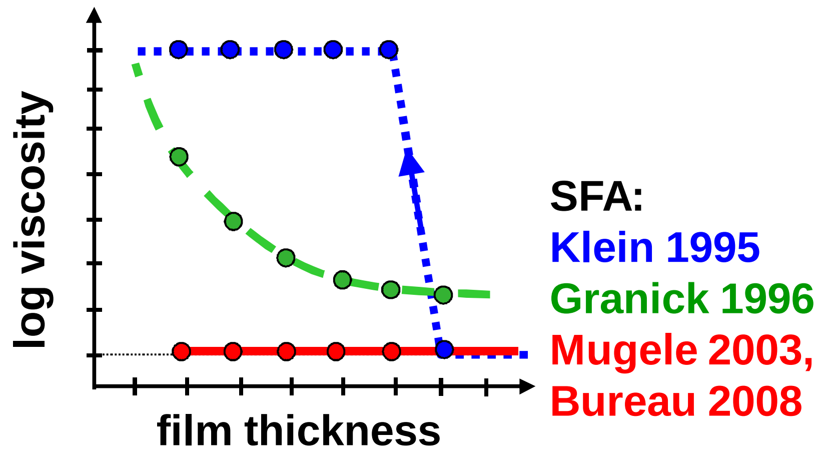
the menu

- confined simple liquids – macroscopic contacts (SFA)
- dynamic AFM measurements on confined liquids:
  non-monotonic evolution of dissipation
- Molecular Dynamics simulations of simple confined Lennard-Jones fluids
  - *non-monotonic evolution of dissipation*
  - *anisotropy of force fluctuations & dissipation*
  - *breakdown of Stokes-Einstein relation*

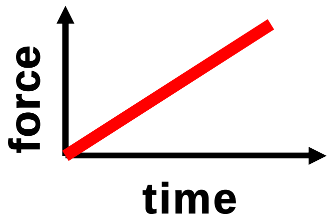
force measurements on confined liquid films

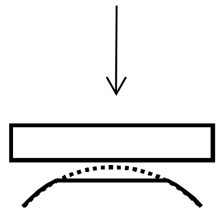


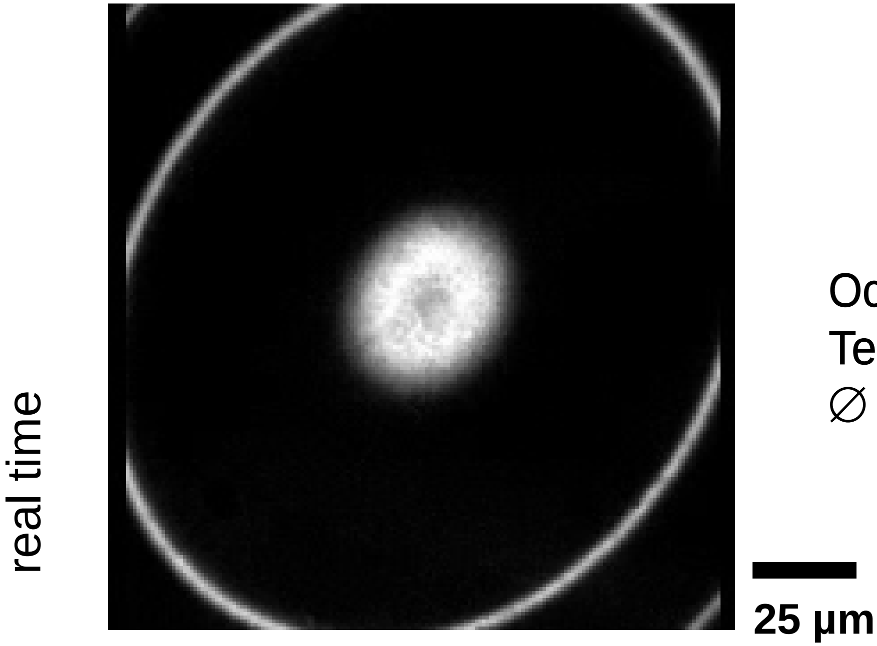


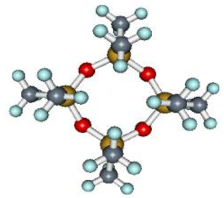


layer-by-layer squeeze-out in macroscopic contacts (SFA)

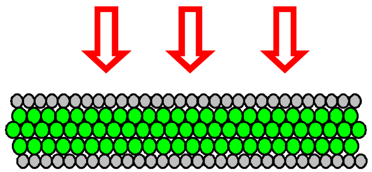


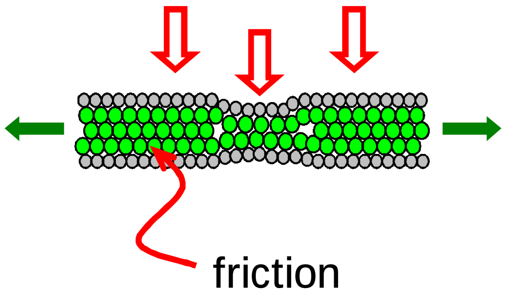






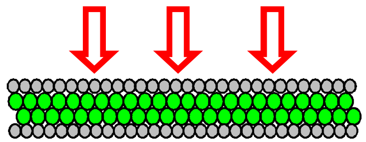
OctaMethylCyclo-
TetraSiloxane
$\varnothing$: ≈ 9 Å





2D potential flow: $\Delta\Phi = 0$

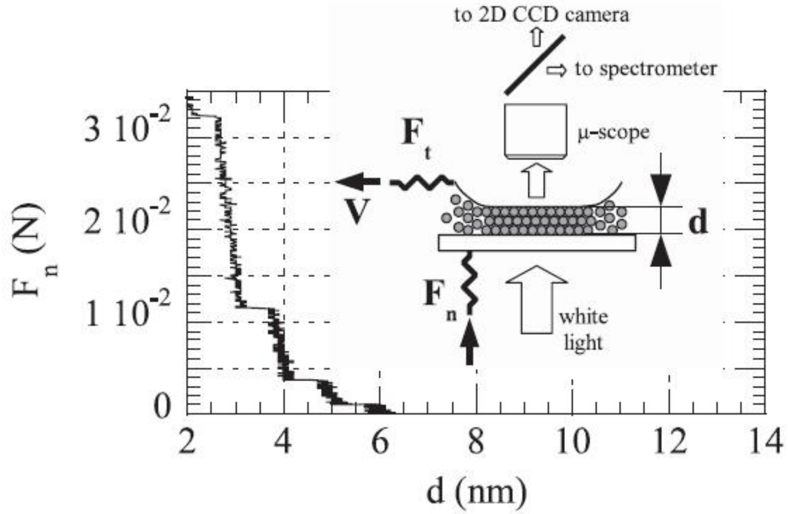
$$\Phi = - p_{2D} / \left(r_{2D} h_{eff}\right)$$

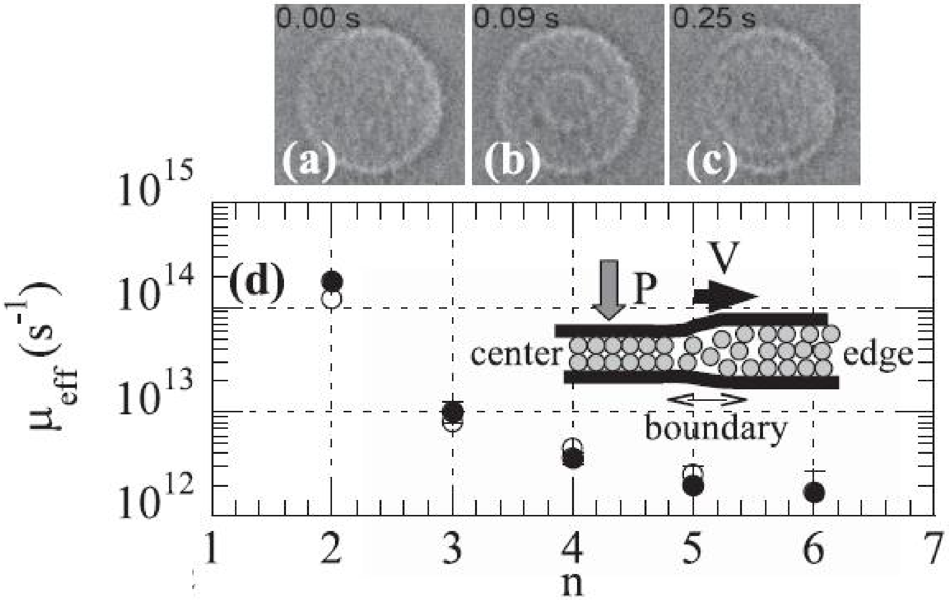


Persson, Tosatti
PR B 1994

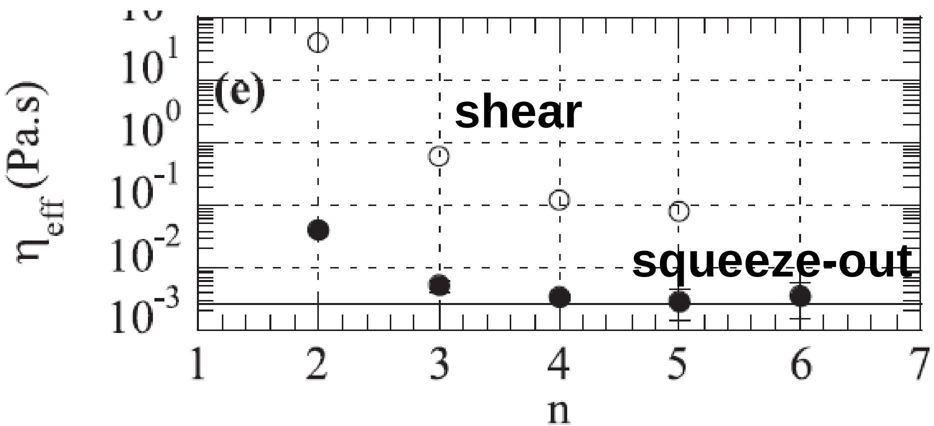
combined squeeze-out and shear measurements

L. Bureau, PRE 2008; PRL 2010

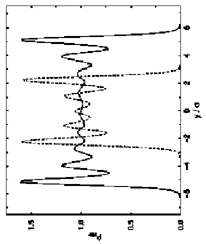


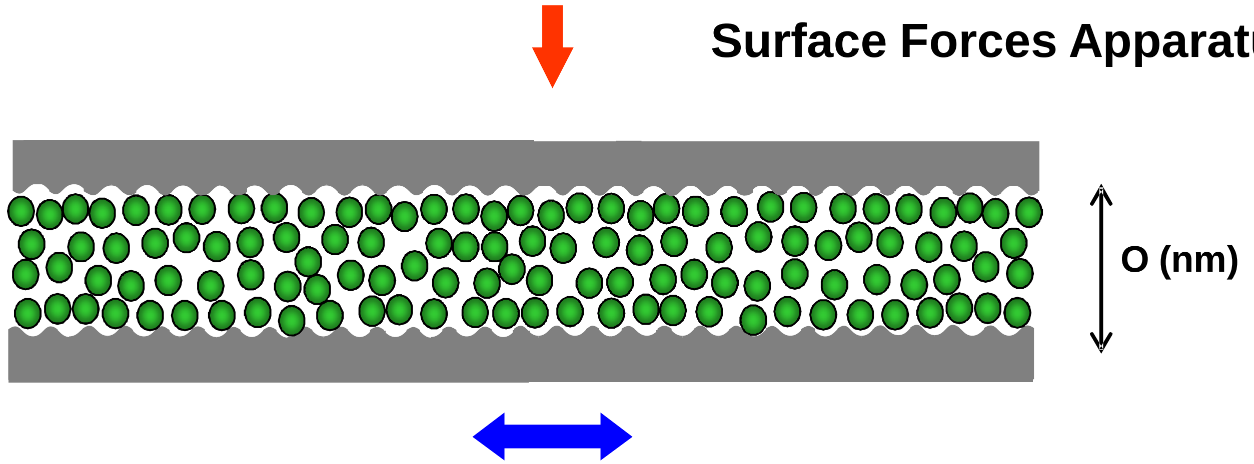


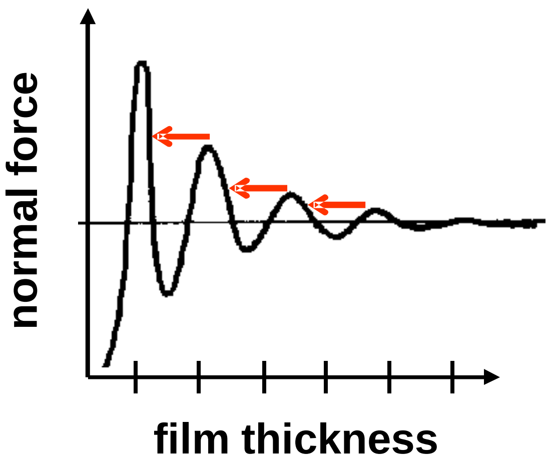
- q no stick slip motion
- q squeeze-out: $h \approx h_{bulk}$
- q shear: $h \approx 100\mathrm{x}h_{bulk}$

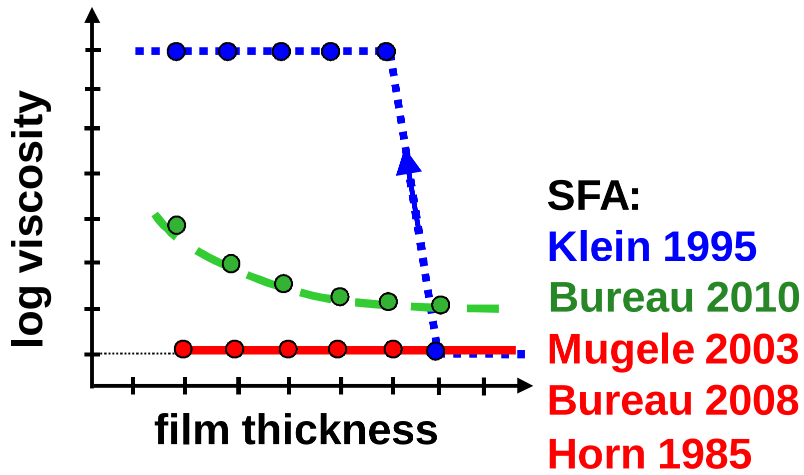


force measurements on confined liquid films

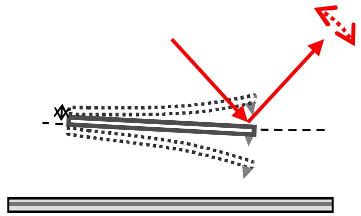




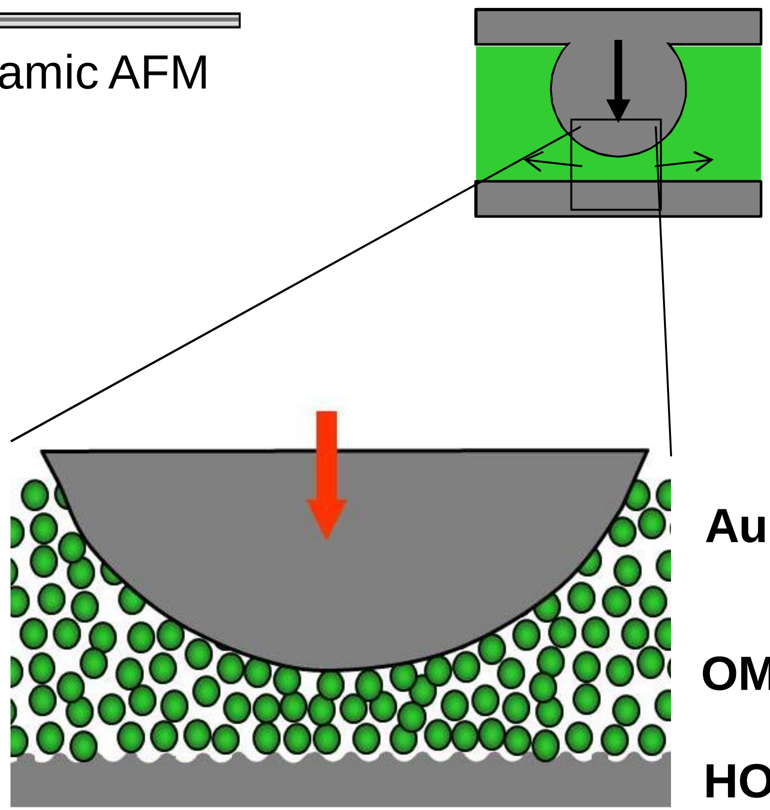




nano-asperity contacts

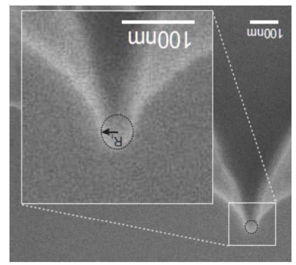


dynamic AFM



squeeze-out damping
(Reynolds damping)

$$F_R = \frac{6\pi \mu R^2}{D}\dot{D}$$



R=17…35nm

PRL 96, 086105 (2006) PHYSICAL REVIEW LETTERS week ending 3 MARCH 2006

Oscillatory Dissipation of a Simple Confined Liquid

Abdelhamid Maali,* Touria Cohen-Bouhacina, Gérard Couturier, and Jean-Pierre Aimé
CPMOH, Université Bordeau... 351 Cours de la Libération, 33405 Talence, France
(Received 3... ...005; published 3 March 2006)

PRL 97, 179601 (2006) PHYS...

Comment on "Oscillatory Dissipation of a ... Confined Liquid"

S. J. O'Shea
Institute of Materia...
3 Research Link, Si...

APPLIED PHYSICS LETTERS 93, 011909 (2008)

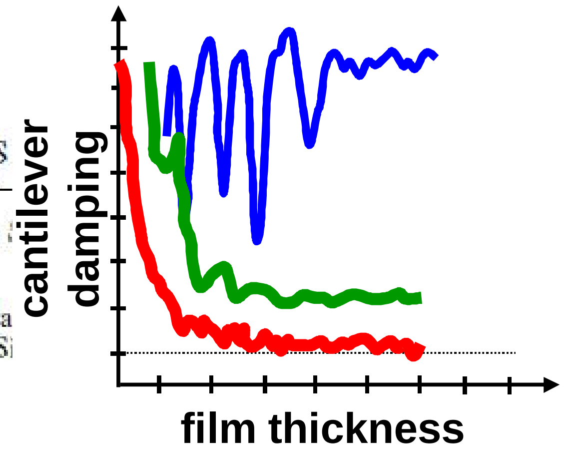
Artifact-free dynamic atomic force microscopy reveals monotonic dissipation for a simple confined liquid

G. B. Kaggwa,[1,a)] J. I. Kilpatrick,[1] J. E. Sader,[2] and S. P. Jarvis[1]

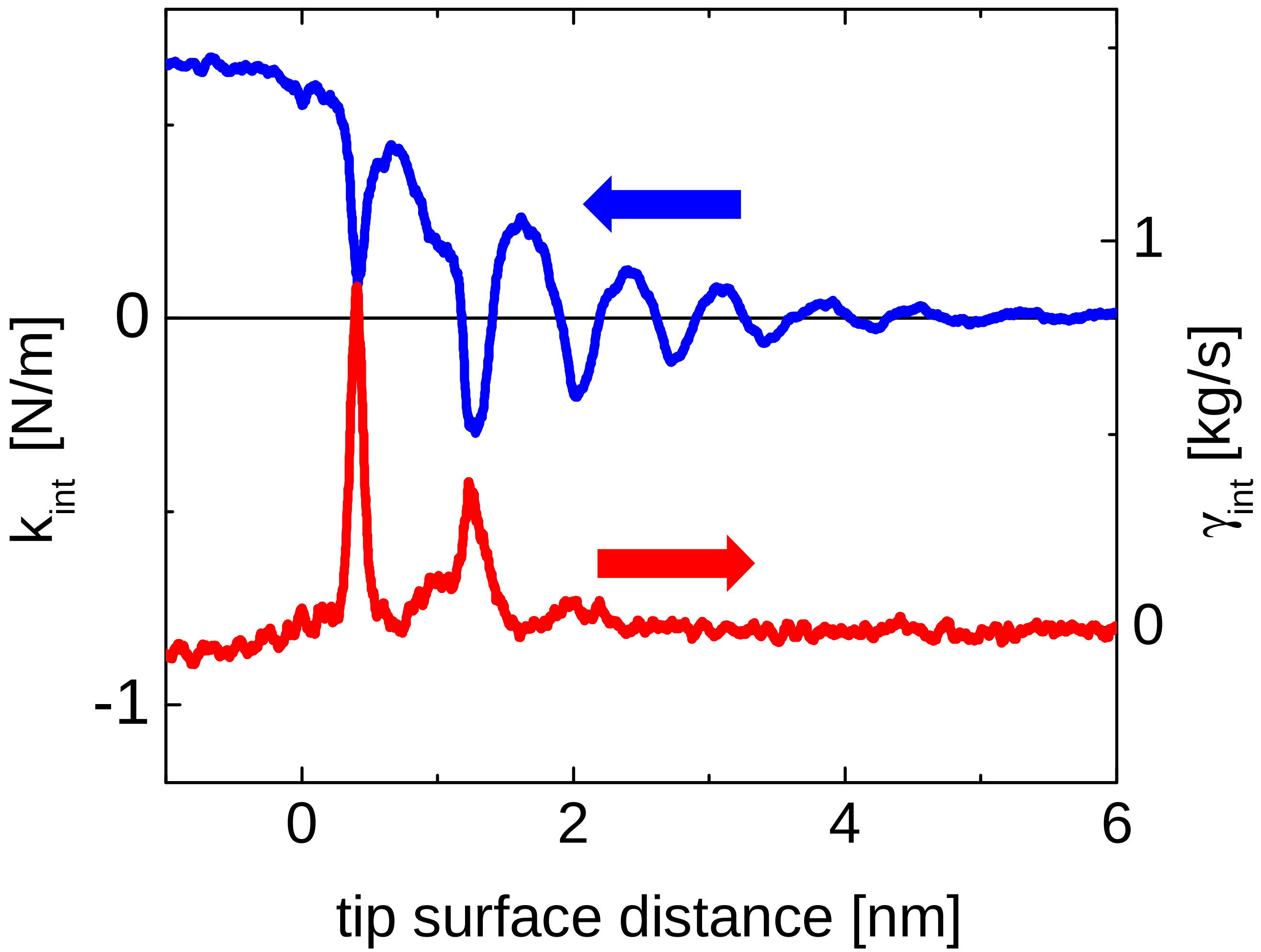
Langmuir 2006, 22, 6485–6488

Solid or Liquid? Solidification of a Nanoconfined Liquid under Nonequilibrium Conditions

Shivprasad Patil,† George Matei,† Ahmet Oral,‡ and Peter M. Hoffmann*,†

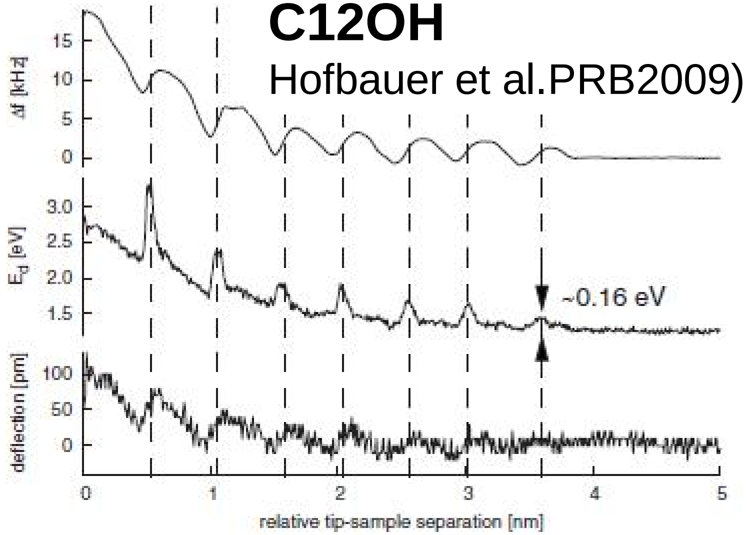


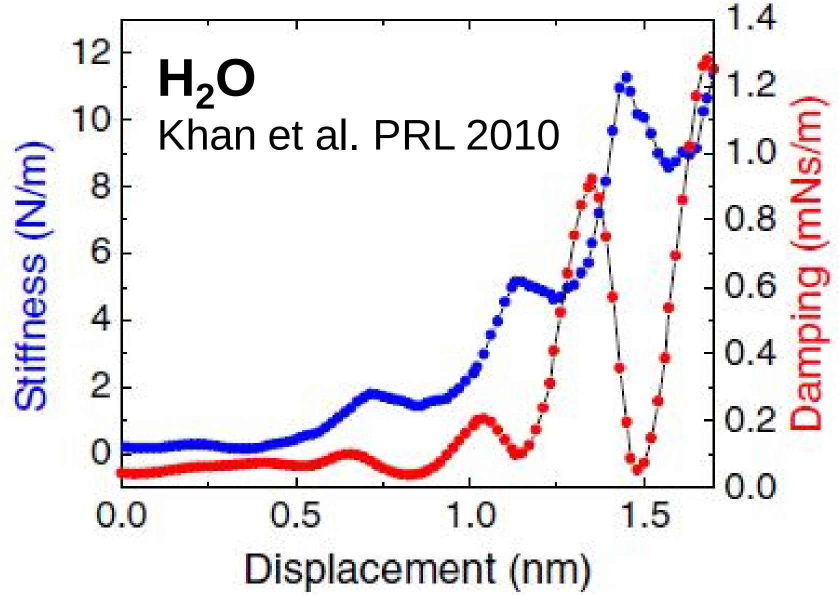
tip-sample interaction in OMCTS

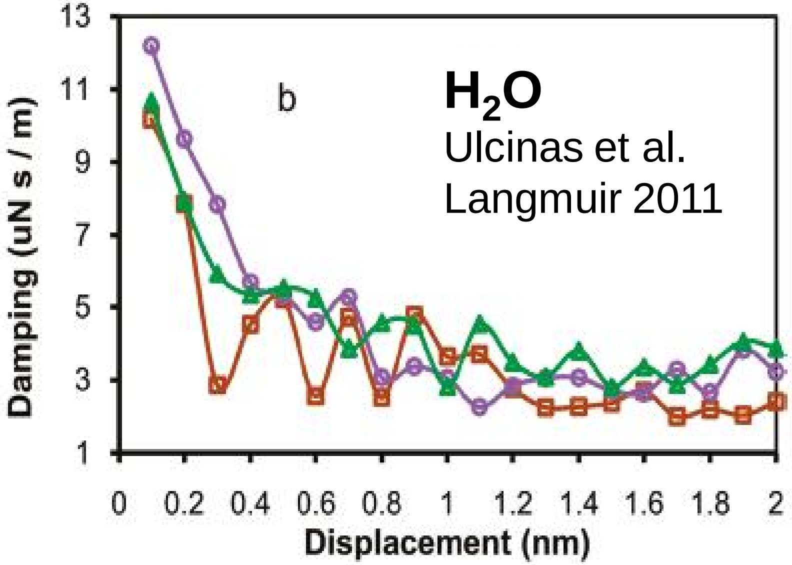


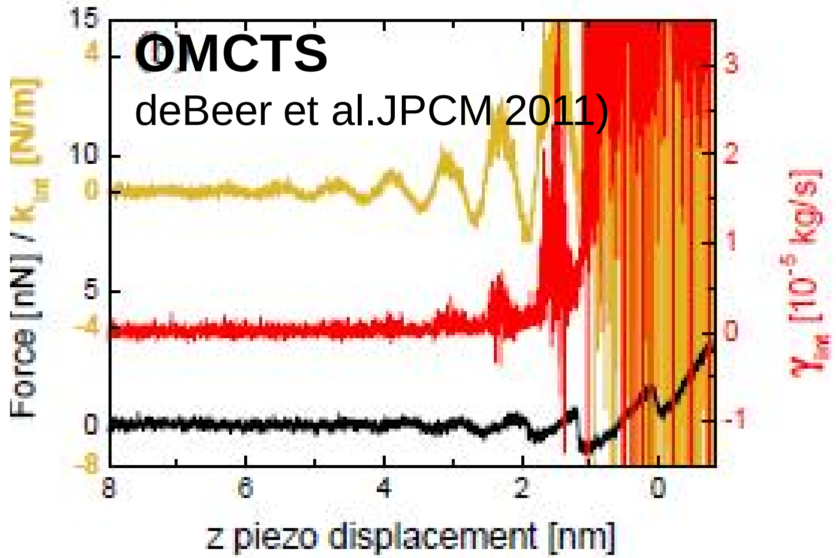
deBeer et al. Nanotechnology 2010

a growing consensus









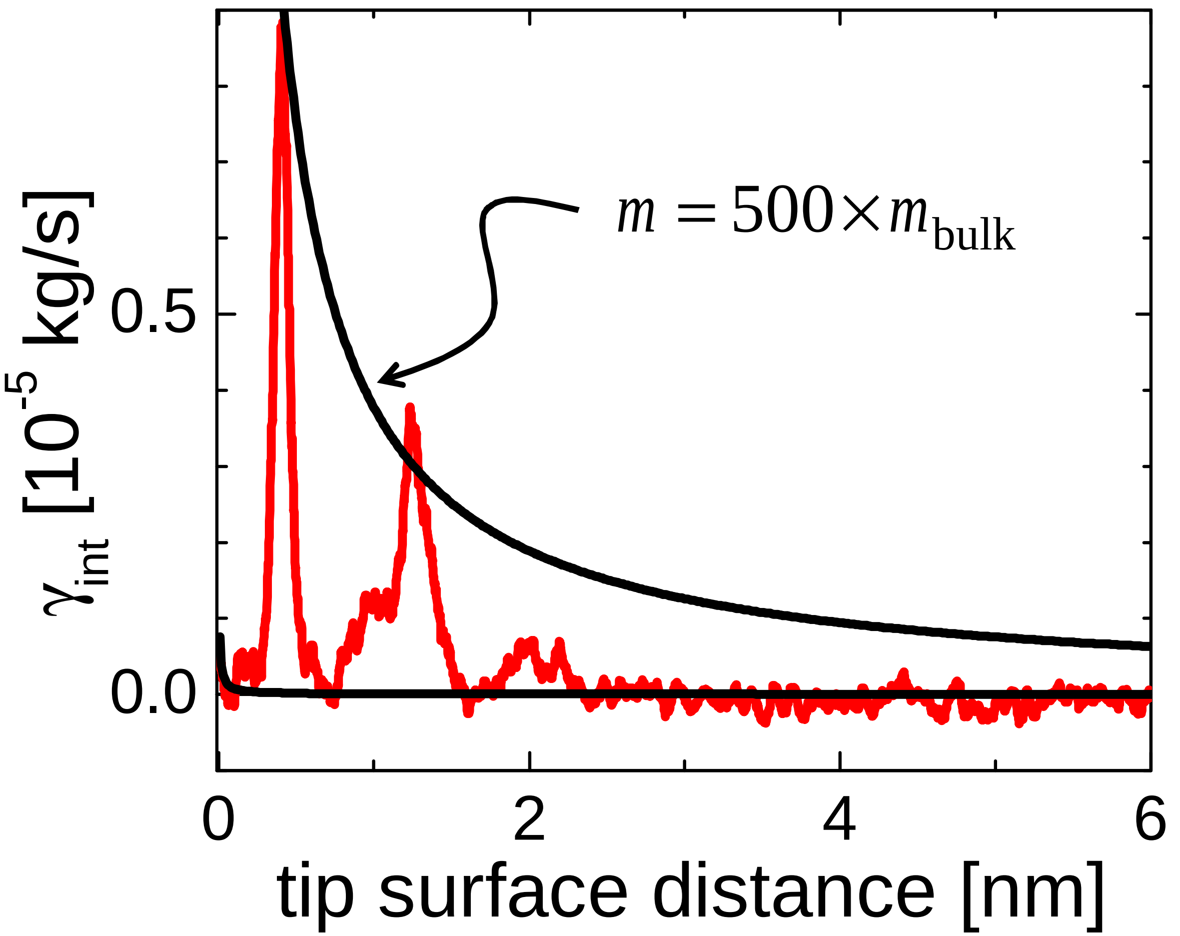
UNIVER

open issues

- relative position of maxima in stiffness & damping
- dependence on tip shape
- dependence on approach rate
- interpretation in term of solidification, glass transition, viscoelastic behavior, …
- epitaxy

how to interpret the excess dissipation?

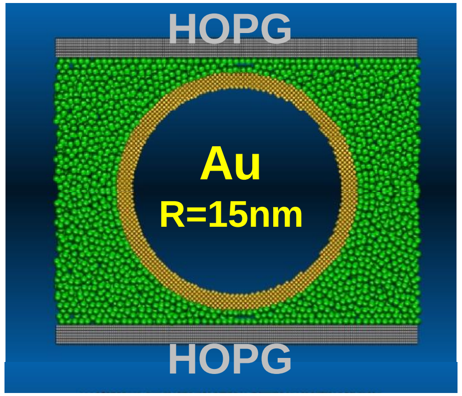
hydrodynamic damping **(Reynolds):** $\gamma_{hydro} = F_R / \dot{D} = \frac{6\pi\mu R^2}{D}$



molecular dynamics simulations

prior art:
Thompson , Robbins
Landmann, Lynden-Bell,
Schön

in collaboration with Wouter den Otter & Wim Briels



3dim

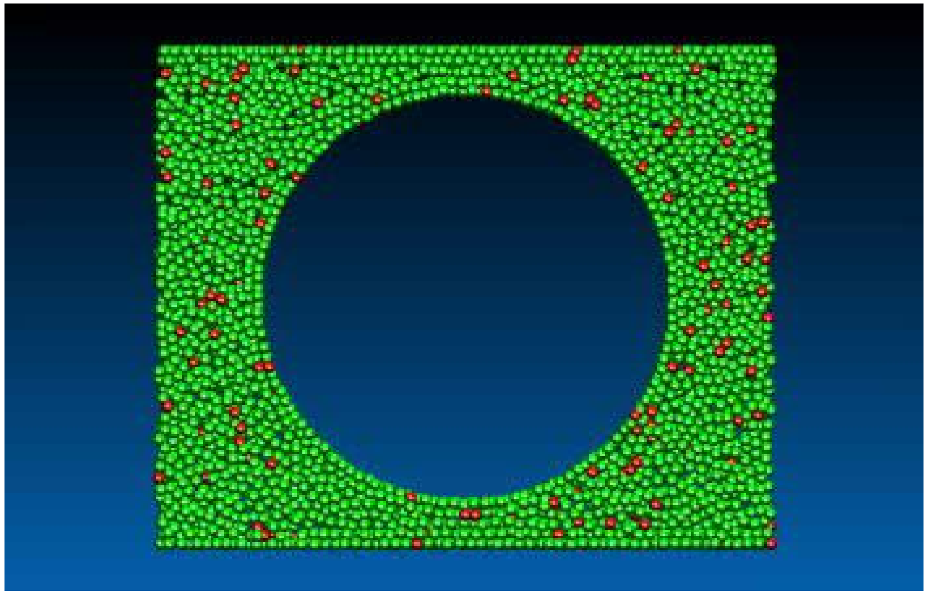
Lennard-Jones particles (≈10^5)

$$V_{ij}(r) = 4e_{ij}\left(\left(\frac{s_{ij}}{r}\right)^{12} - \left(\frac{s_{ij}}{r}\right)^{6}\right)$$

T=300K

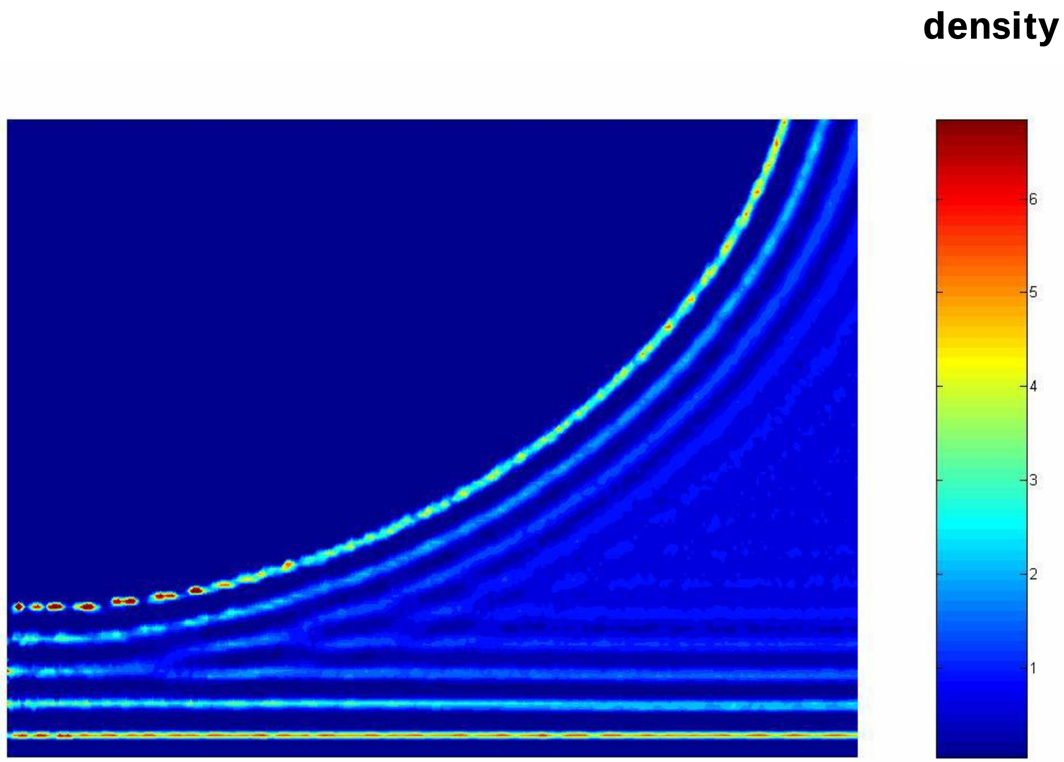
$s_{OMCTS} = 0.77\,\mathrm{nm}$

60ns PT equilibration
10ns NVE simulation

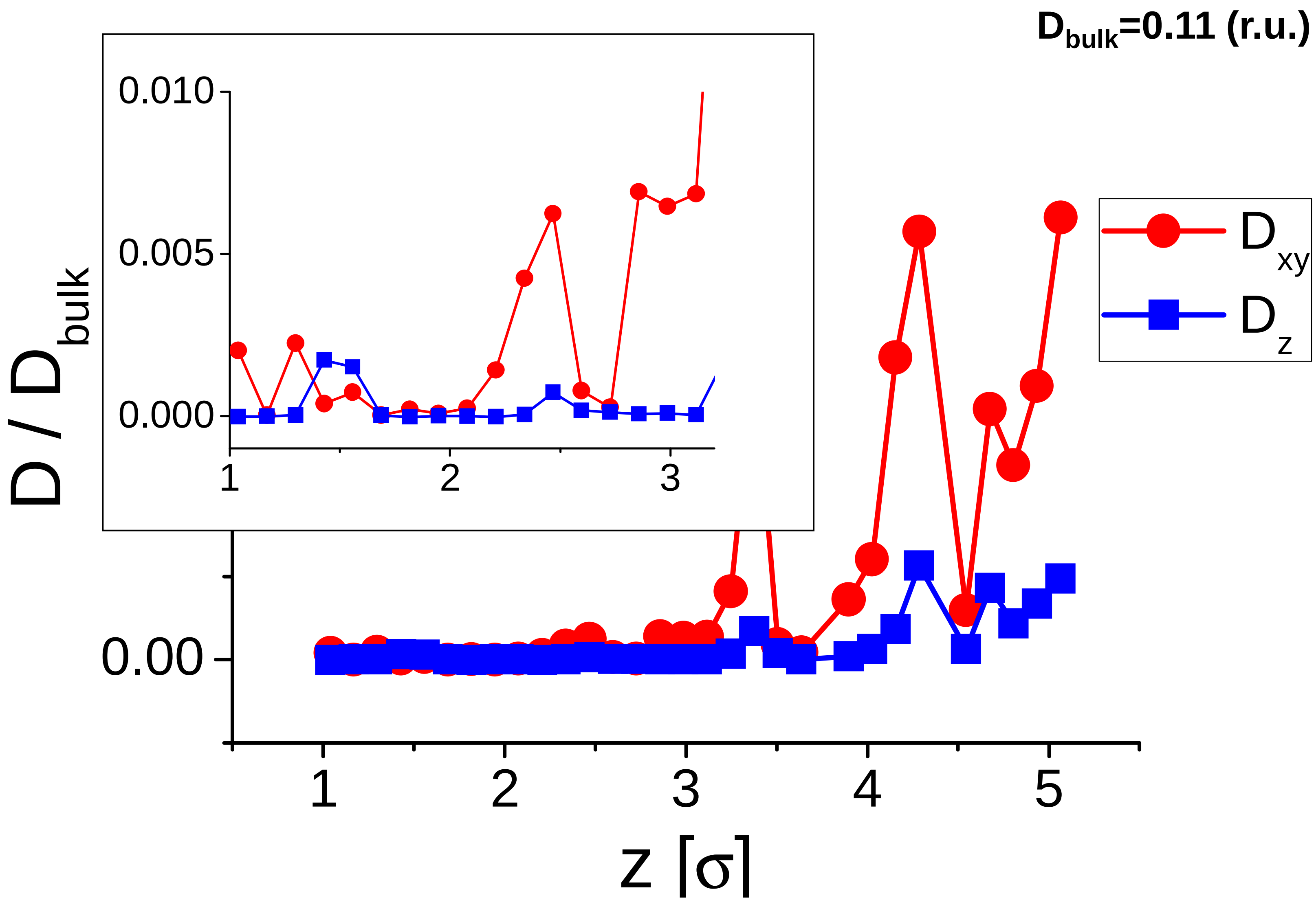


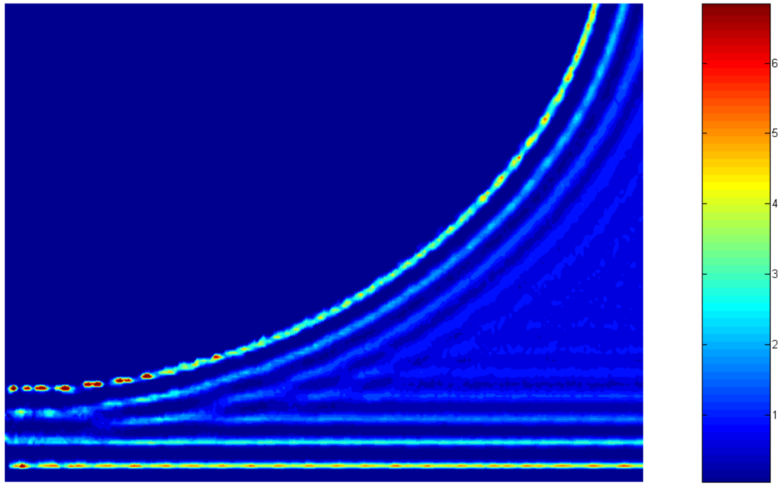
d=5 s

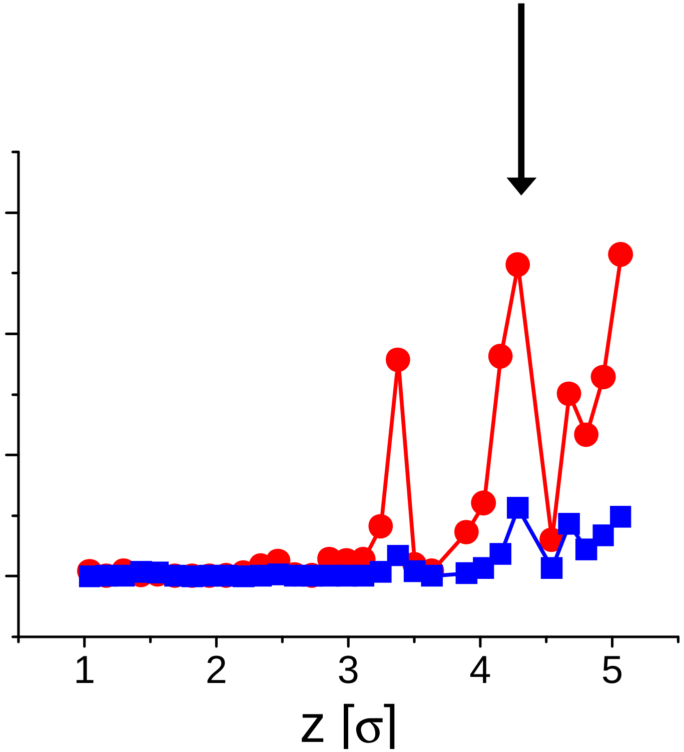
density profiles vs. film thickness



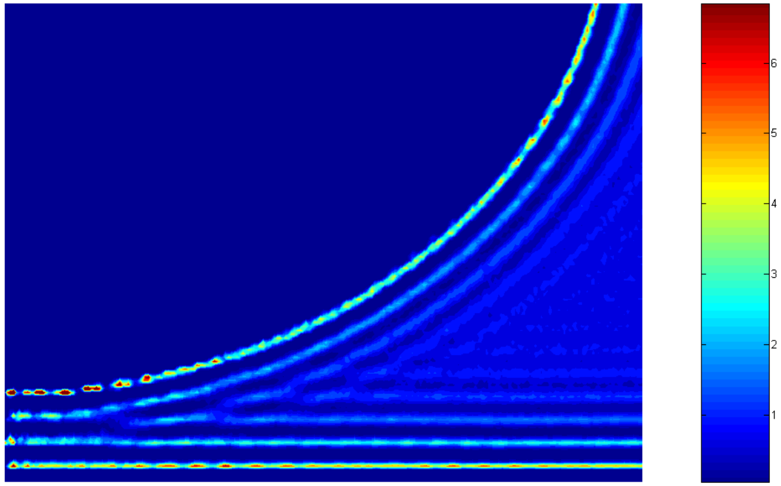
diffusion vs. layer thickness

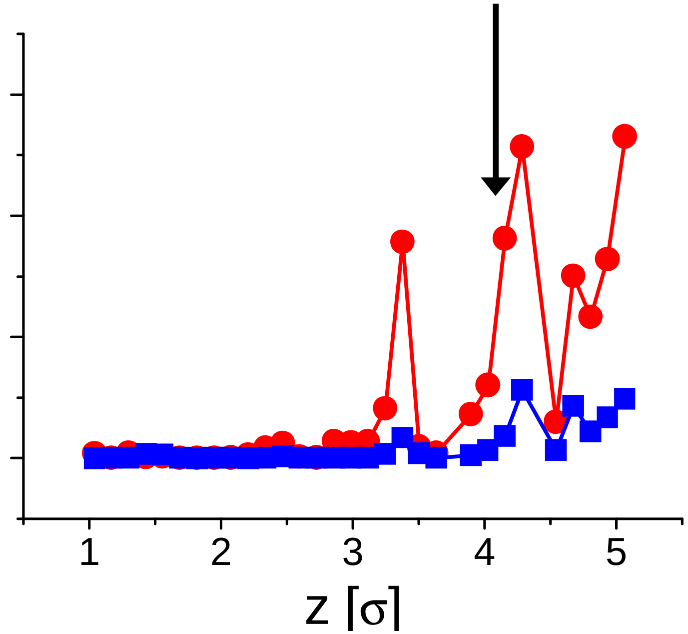




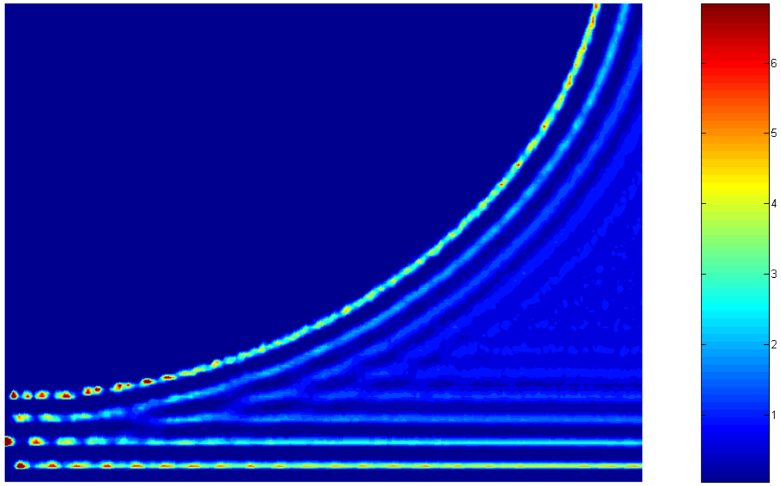


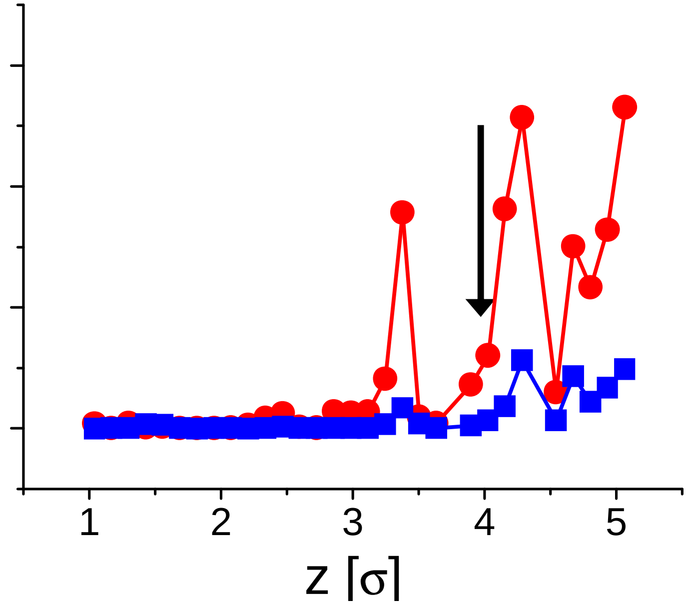
UNIVERSITY OF TWENTE.



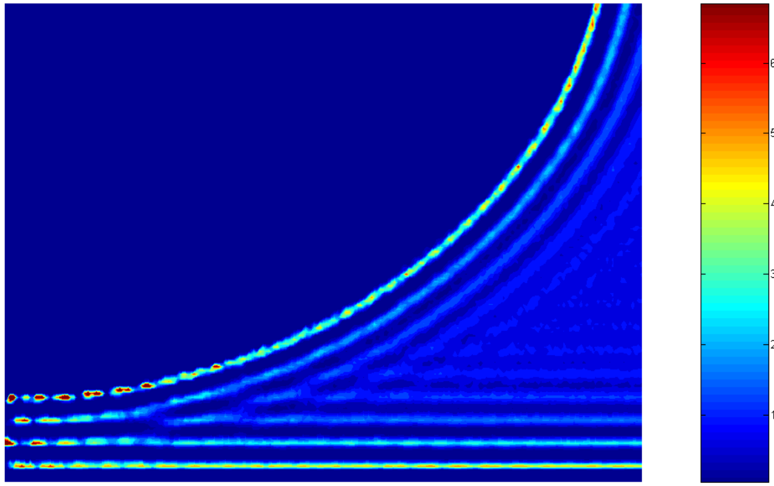


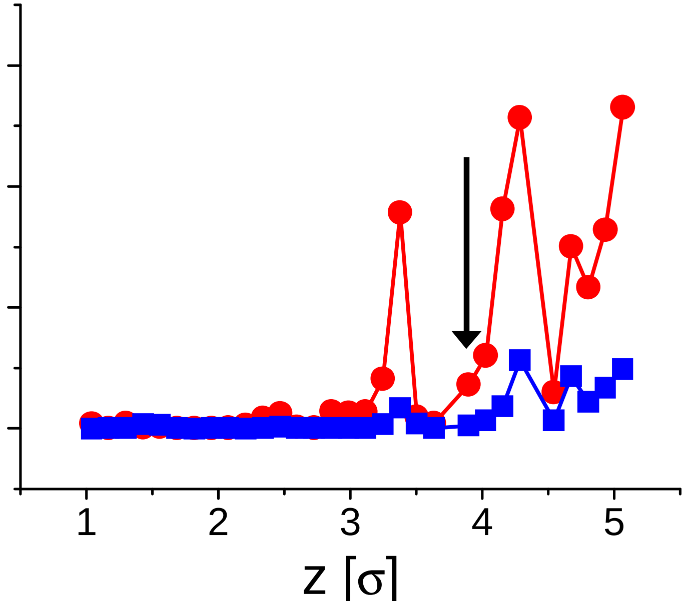
UNIVERSITY OF TWENTE.



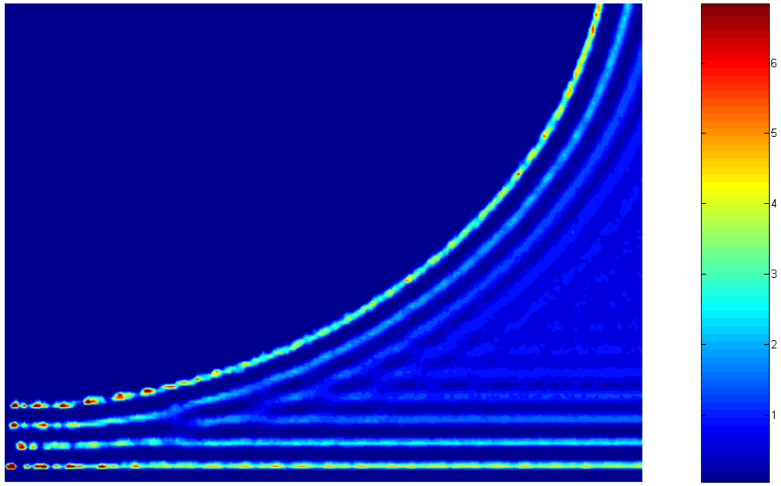


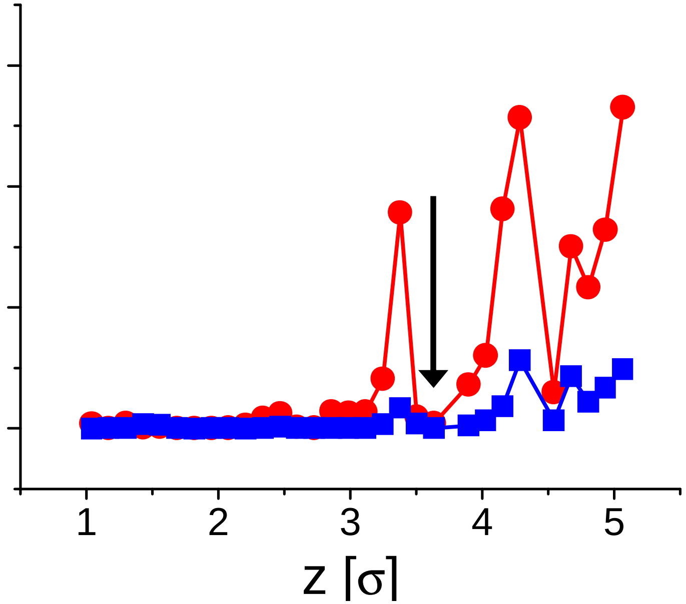
UNIVERSITY OF TWENTE.



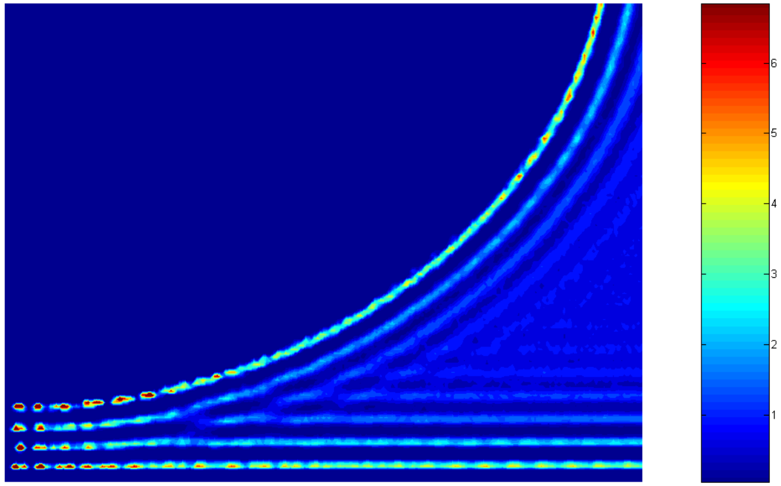


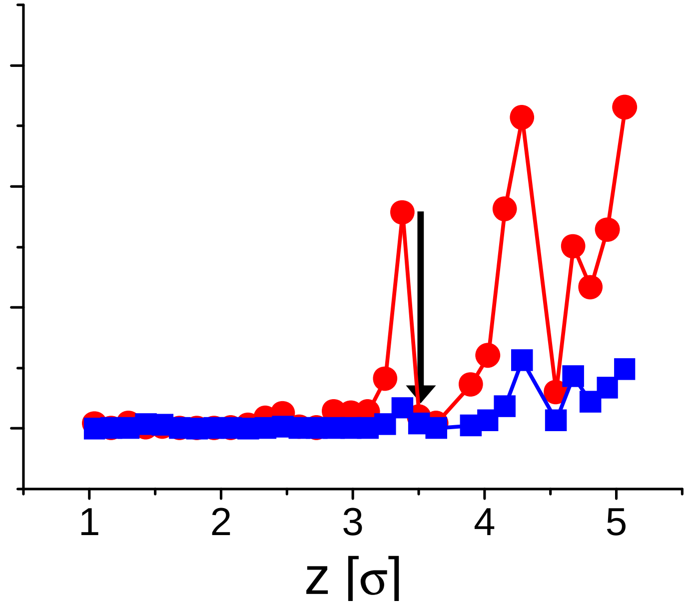
UNIVERSITY OF TWENTE.

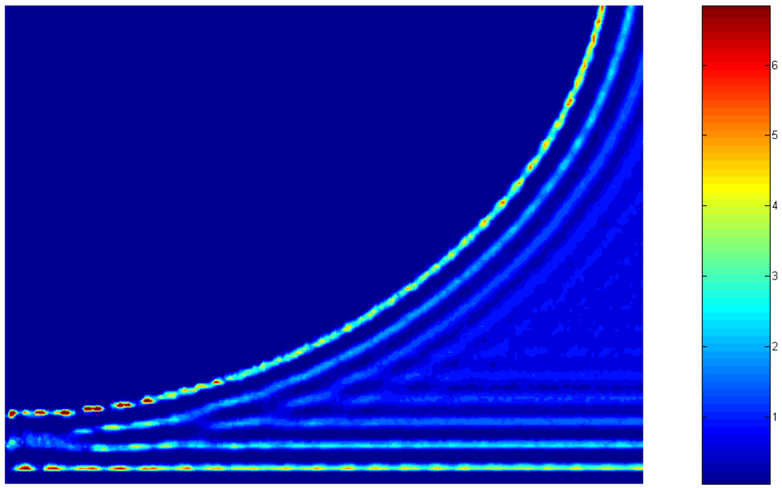


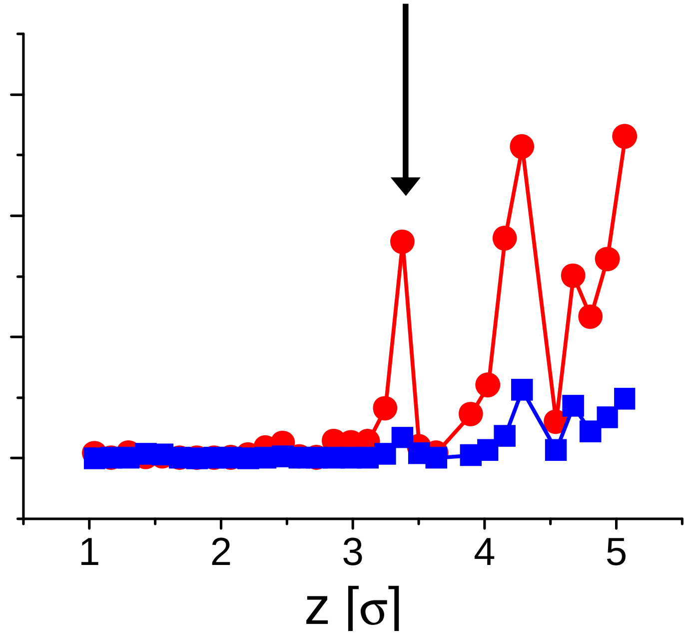


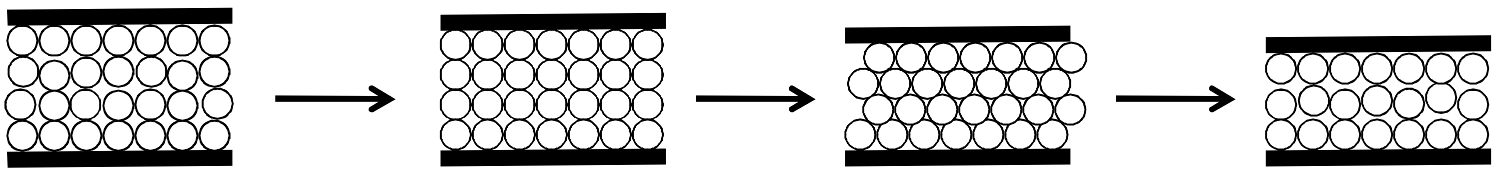
UNIVERSITY OF TWENTE.


1
2
3
4
5
6


1
2
3
4
5
z [σ]

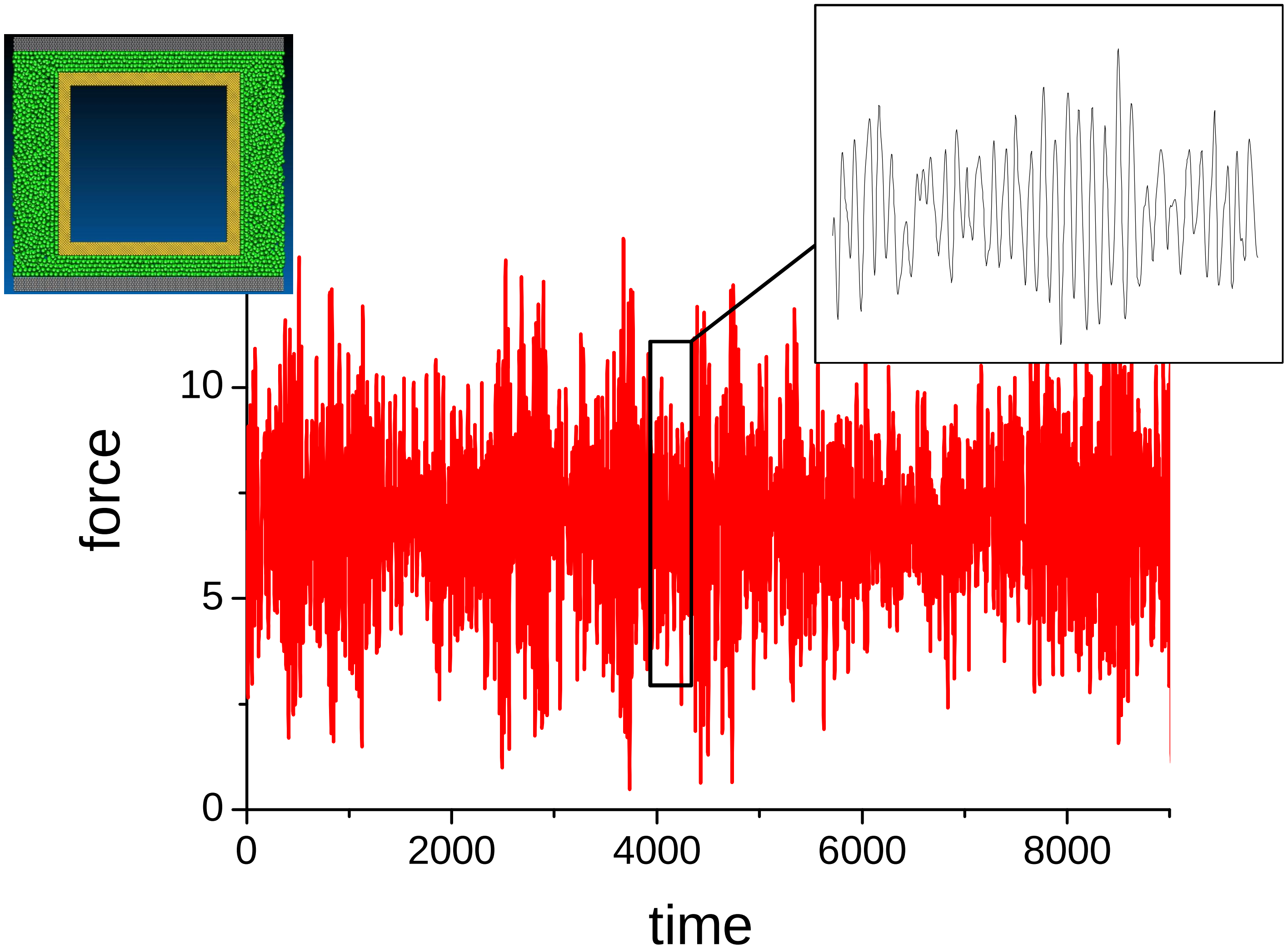



1
2
3
4
5
z [σ]



but AFM does not measure diffusion

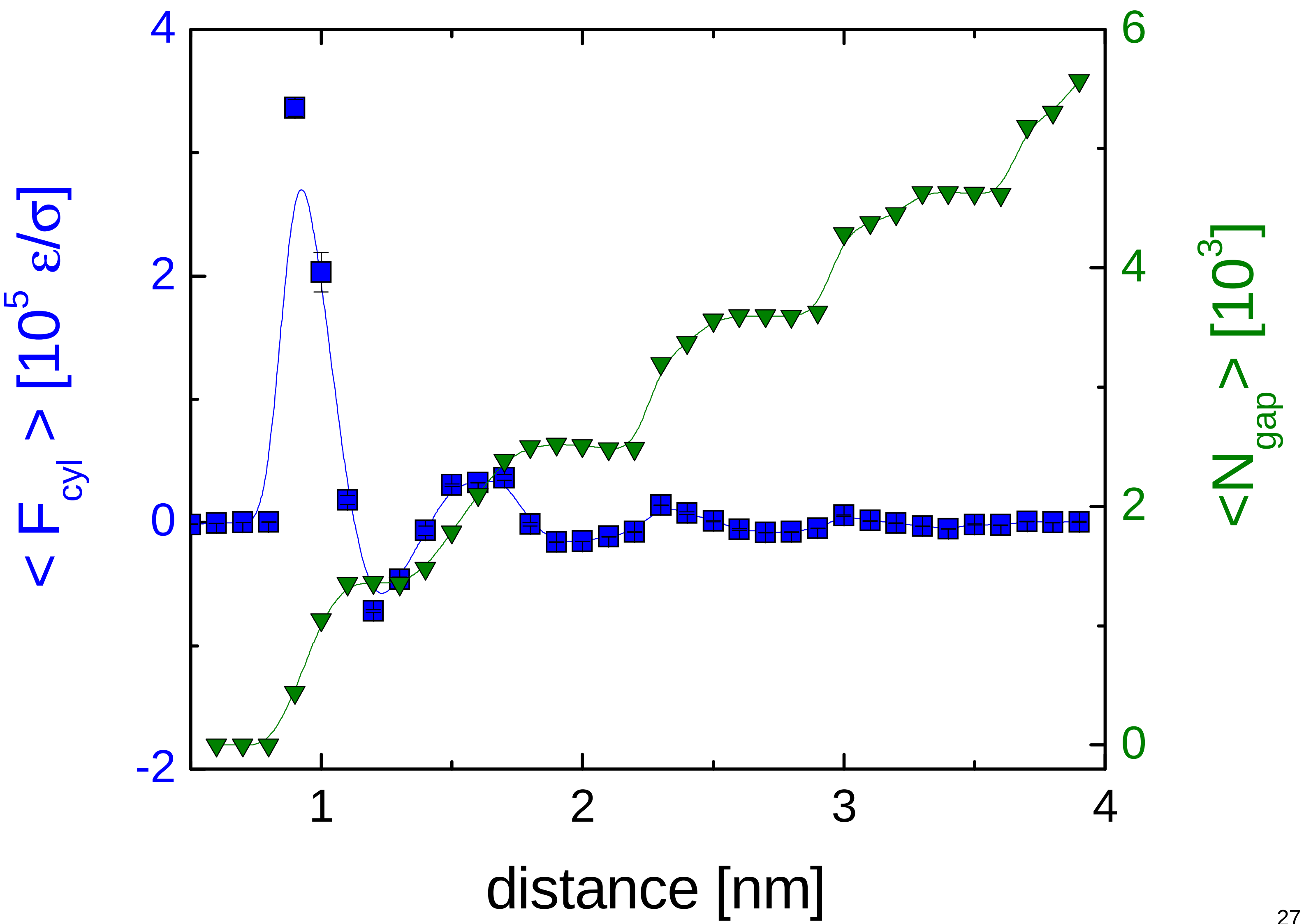
the force on the tip is noisy



fluctuation-dissipation theorem: noise à damping

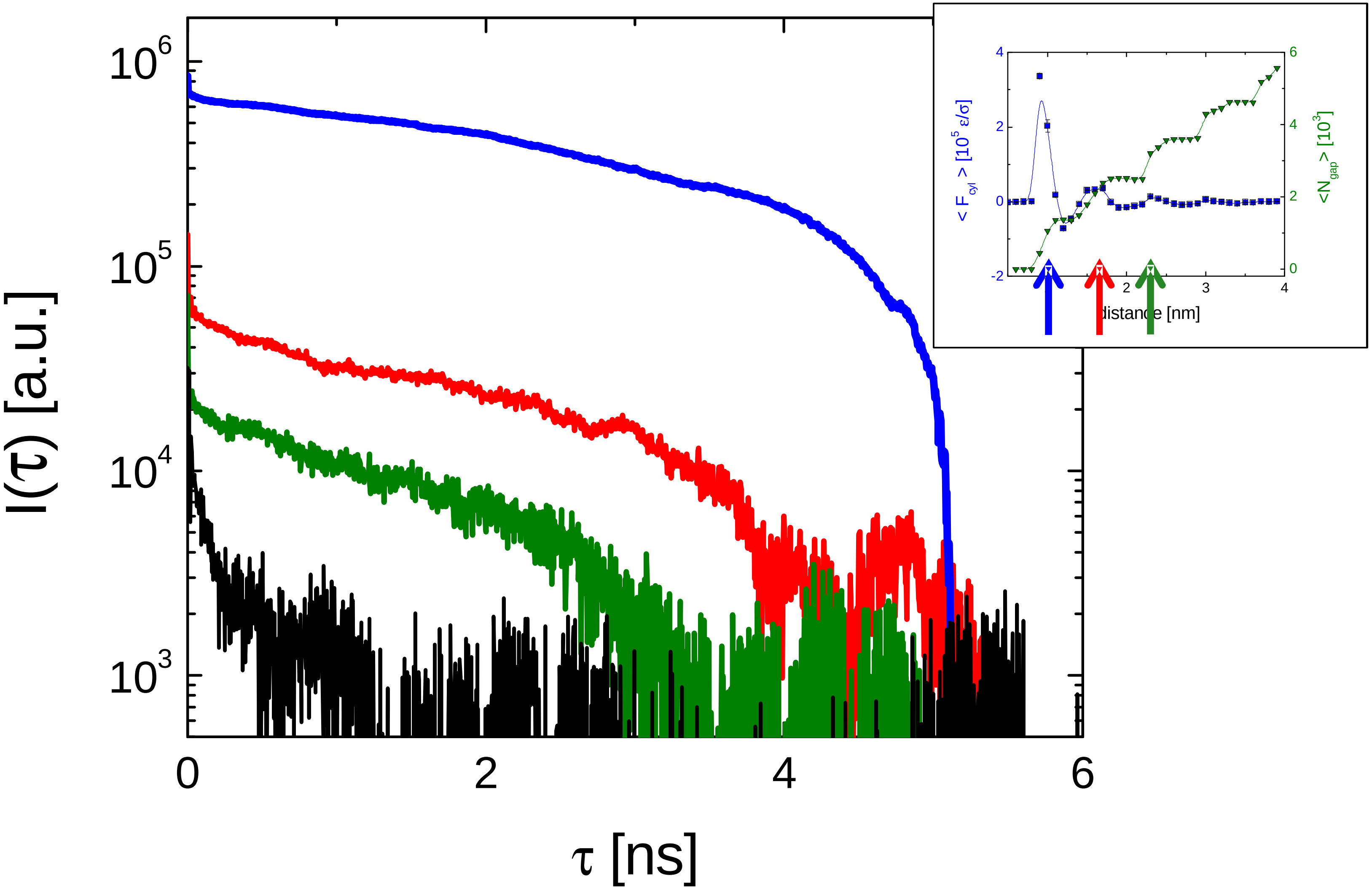
(without excessive shear rates!)

mean force and number of particles



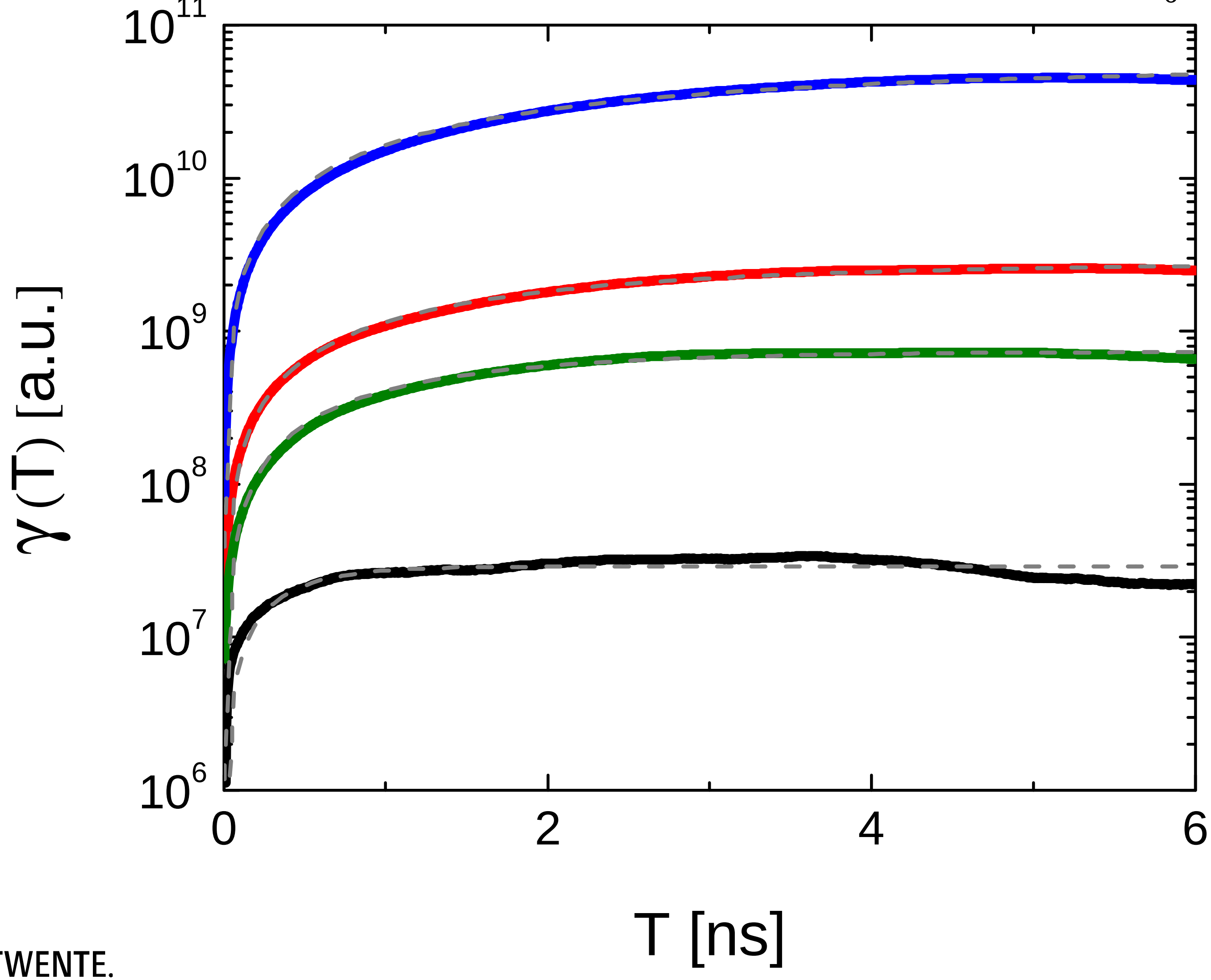
noise correlations

time autocorrelation: $I(t) = \langle dF(0)\, dF(t) \rangle \qquad \left(dF(t) = F(t) - \langle F \rangle\right)$

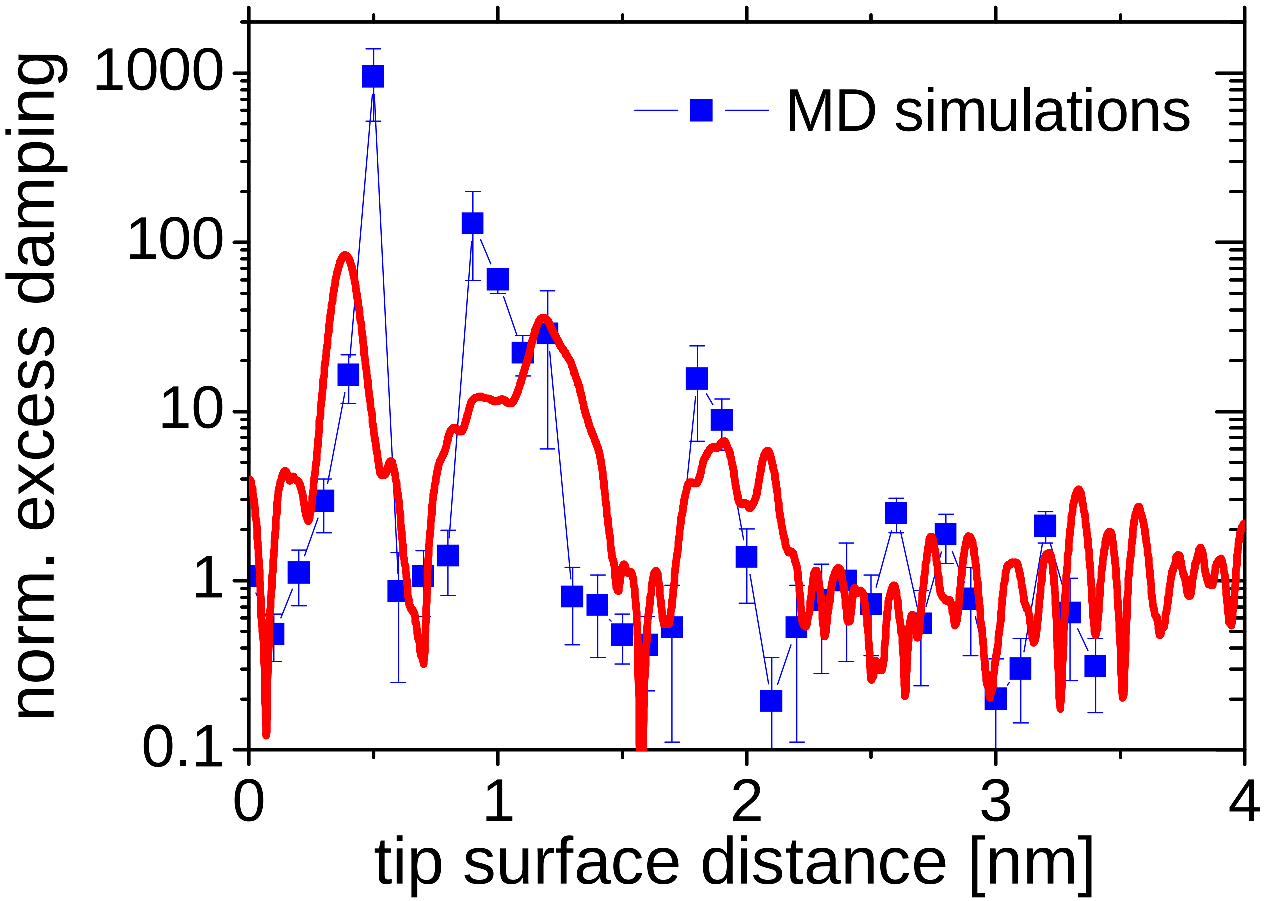


damping coefficient

fluctuation-dissipation theorem: $g = \lim_{T\to\infty} g(T) = \lim_{T\to\infty} \frac{1}{k_B T}\int_0^T I(t)\,dt$

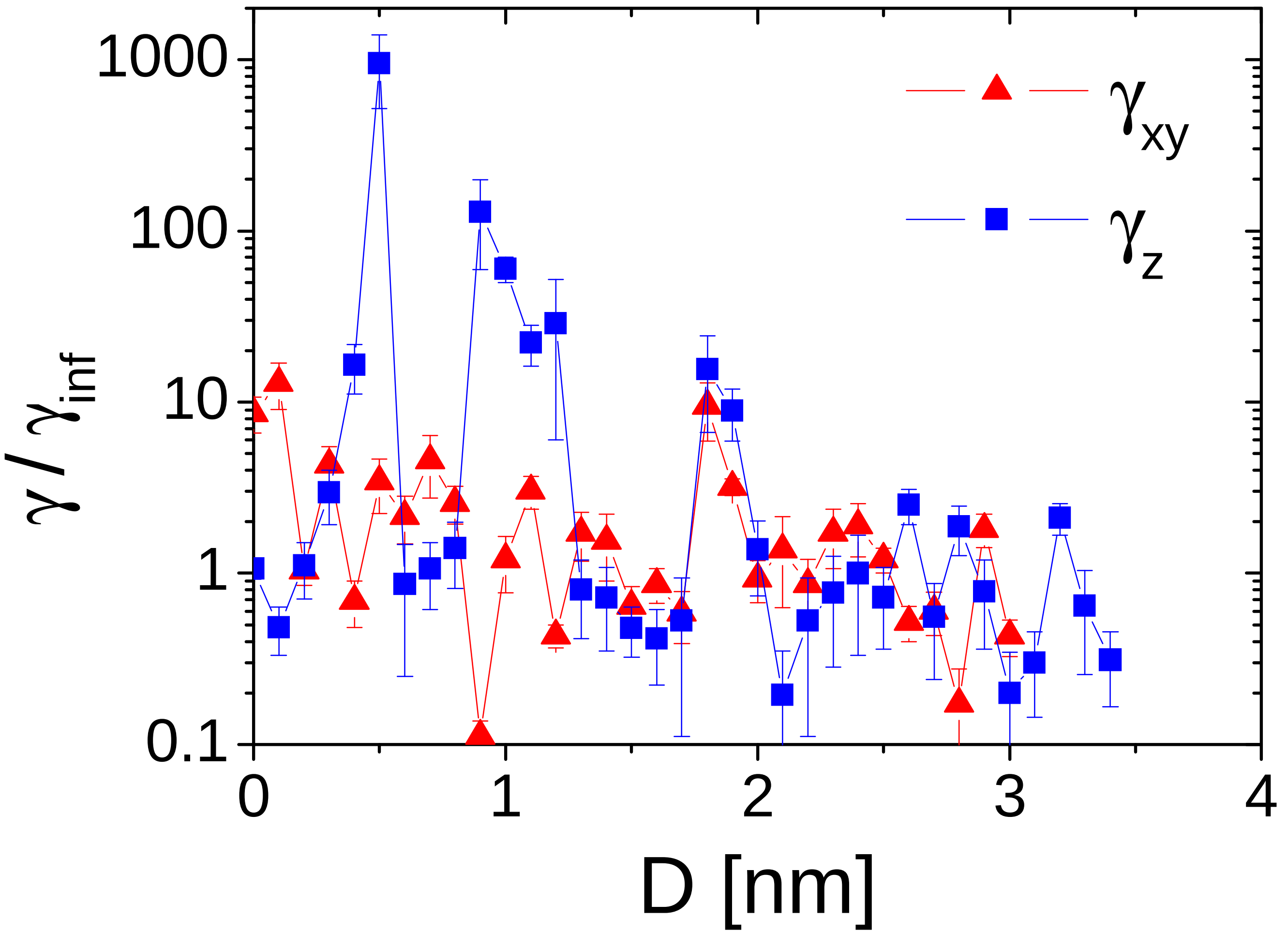


damping coefficient



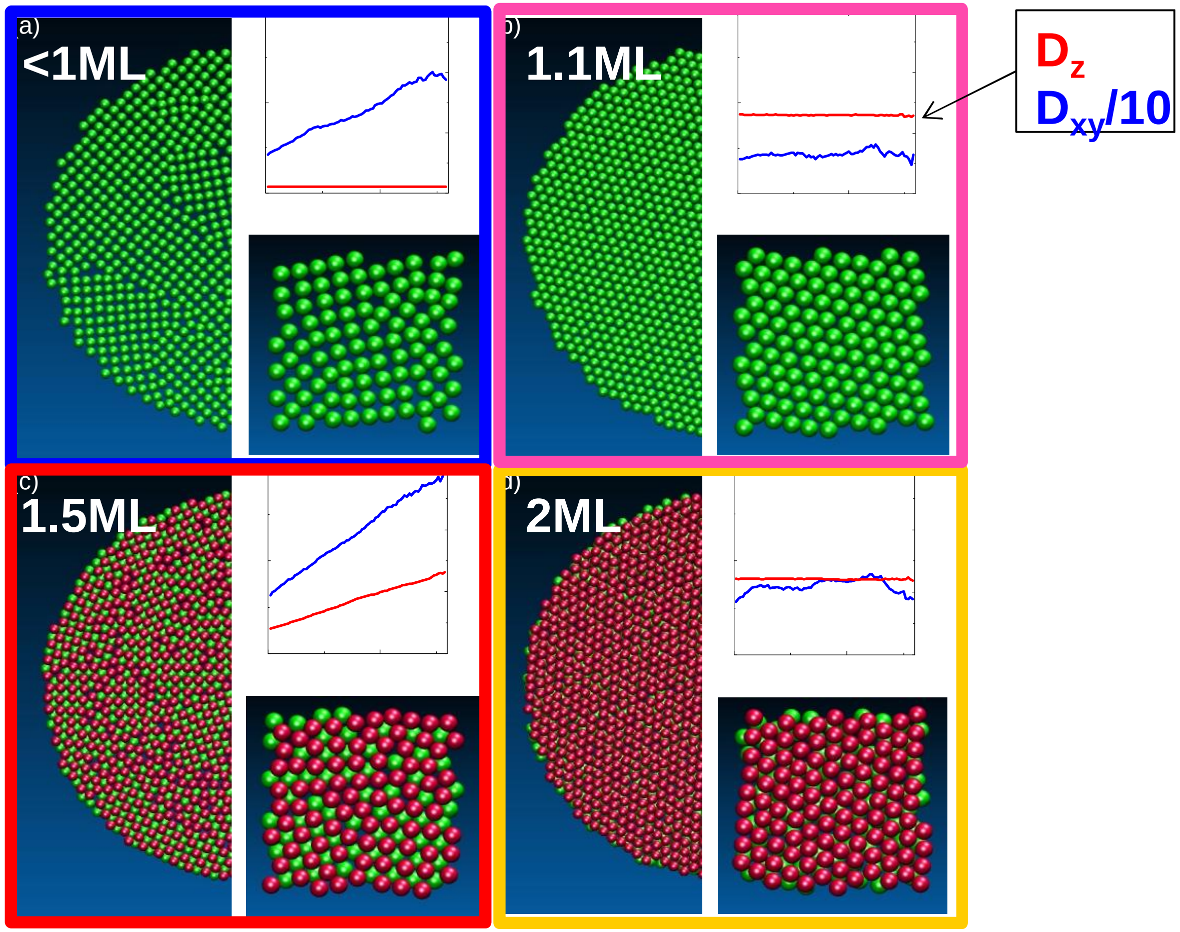
anisotropy matters

lateral damping shows little structure



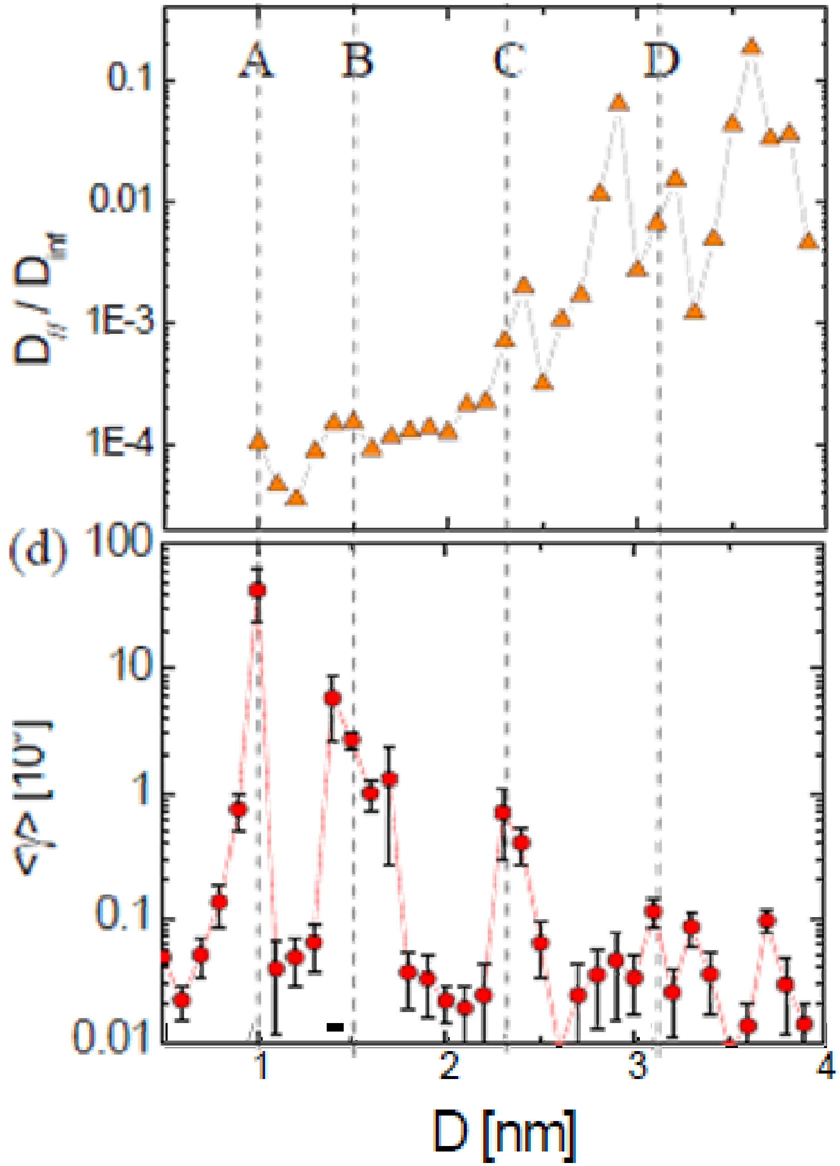
structure matters

disordered films display stronger diffusivity & damping



very heterogeneous dynamics

Stokes-Einstein relation does not hold

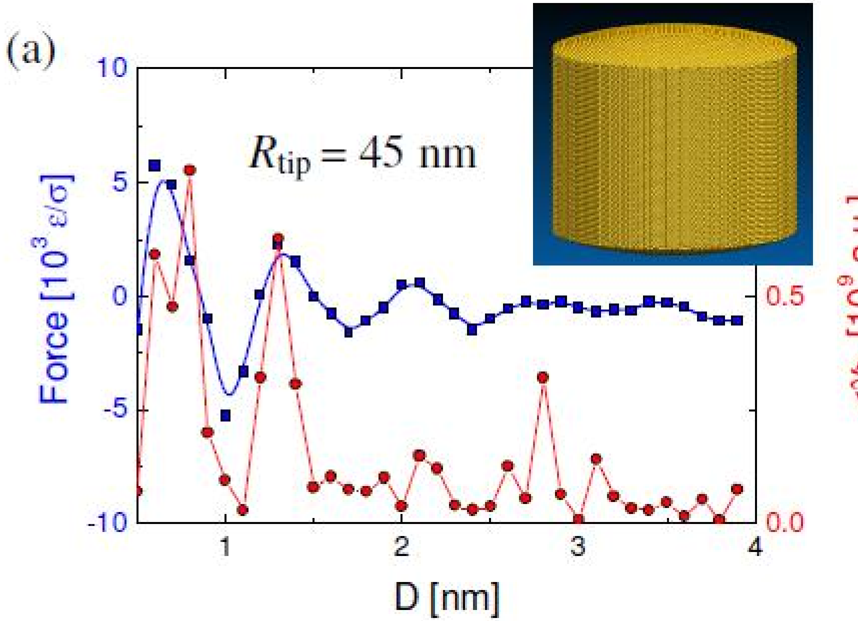


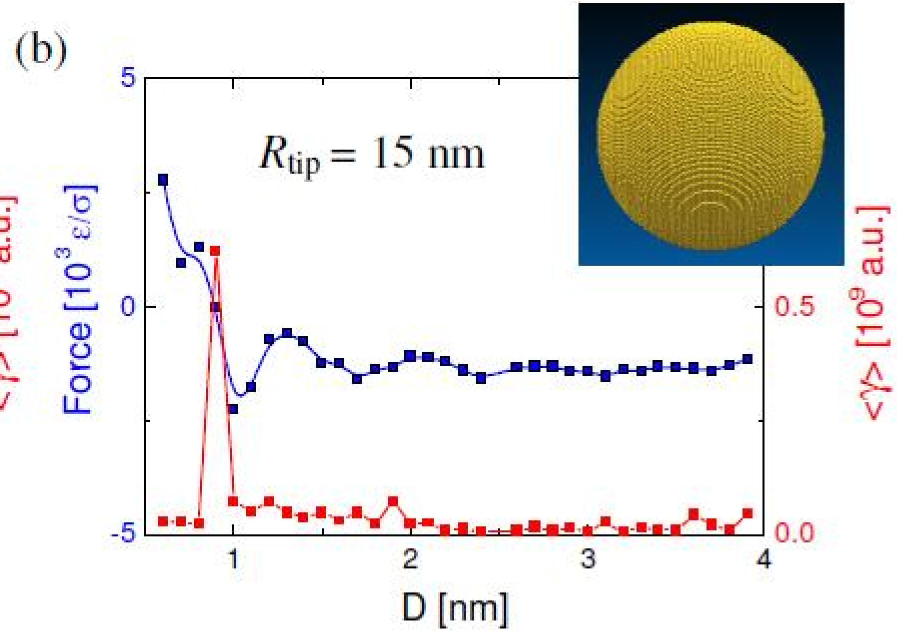
$$D \propto \frac{1}{mobility} \propto \frac{1}{g}$$

$$g = \frac{6p\ m R^2}{h}$$

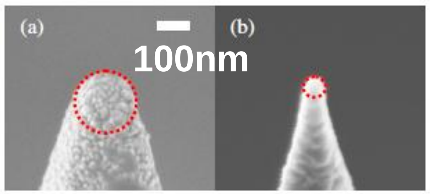
à continuum hydrodynamic description of fluid is no longer applicable

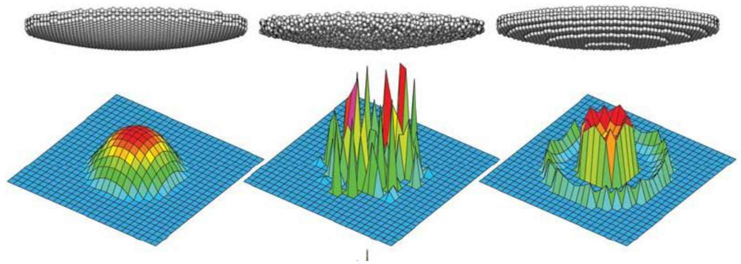
tips size matters





more complex behavior for more complex tip shapes?





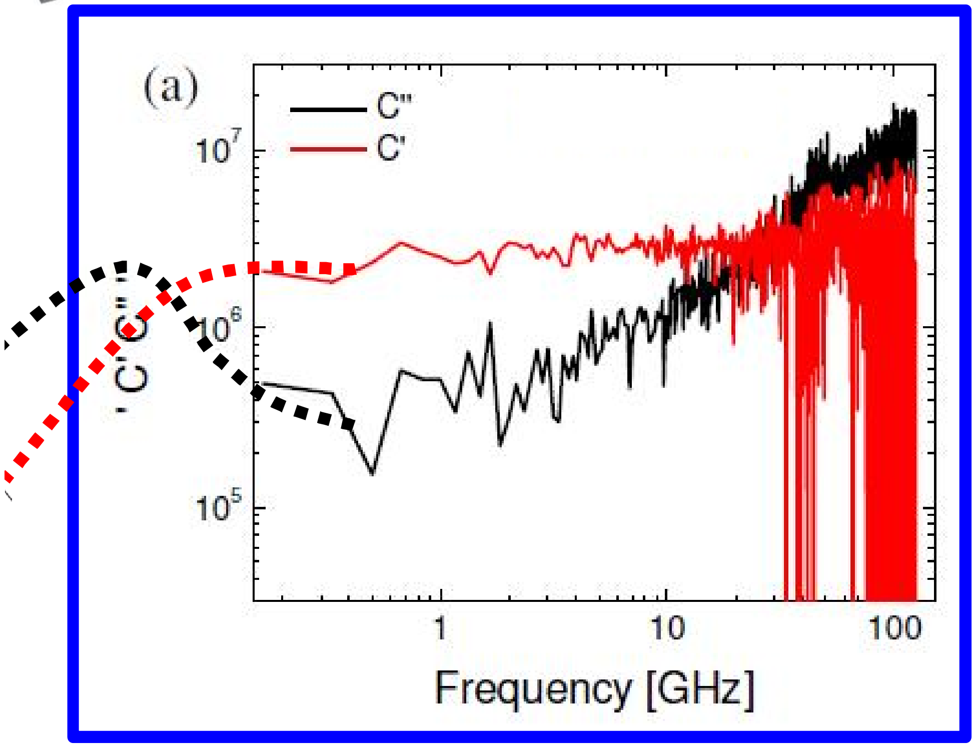
Luan, Robbins 2005

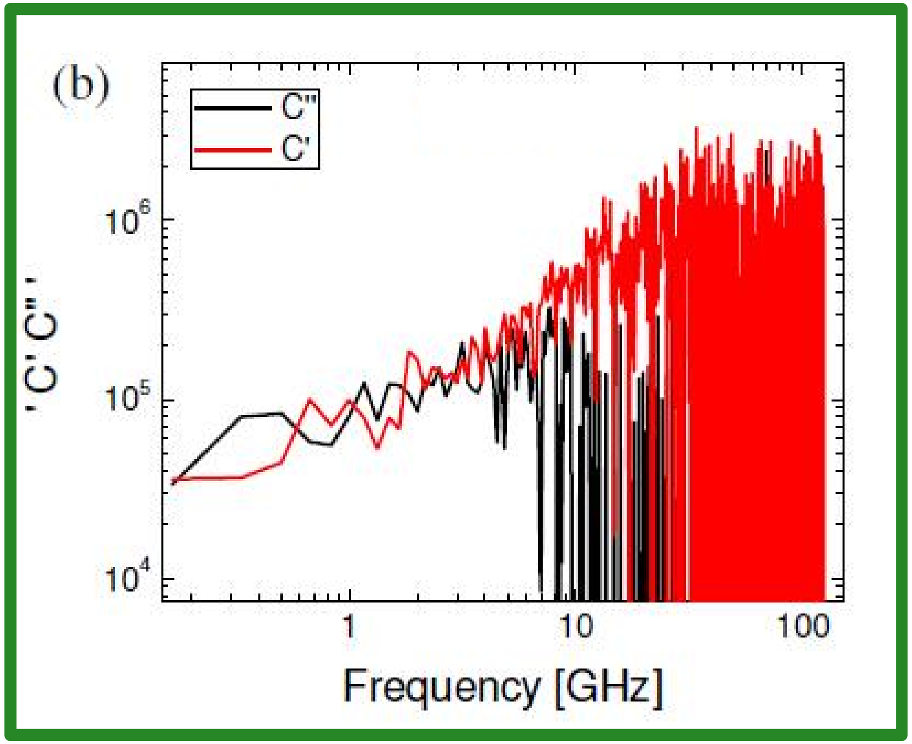
outlook: nano-rheology

$$C' = \int V(t) \cos wt \; dt \qquad C'' = \int V(t) \sin wt \; dt$$

elastic response **dissipative response**

on peak **off peak**





conclusions

- nano-confined liquids display non-monotonous dissipation
- disordered layer structure entails excess damping & diffusivity (violation of the Stokes-Einstein relation)
- well-ordered layers display solid-like structure and little dissipation (linear response hardly sensitive to solidification)
- strong anisotropy between z- and xy-directions

come to Leiden next April

Lorentz Center workshop

Fundamental Aspects of Friction and Lubrication

April 16 – 20

co-organized by the PI's FOM program FaF (Fundamentals of Friction)

M. Müser, B. Persson, R. Carpick, R. Bennewitz, E. Riedo, G. Hummer, T. Fukuma, J. O'Shea, L. Nicola, S. de Beer, A. Schirmeisen, E. Meyer, L. Bureau