High Frequency Microrheology Measurement

Active Method:

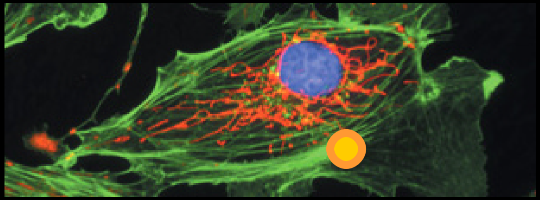
Magnetic microrheometer – Baush, BJ 1998
Huang, BJ 2002

Passive Method:

Single particle tracking – Mason, PRL 1995
Yamada, BJ 2000
Multiple particle tracking – Crocker, PRL 2000

Microrheology of Complex Fluid

- ❑ Rheology: Science of the deformation & flow of matter
- ❑ Microrheology
  - Microscopic scale samples
  - Micrometer lengths

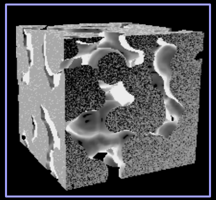


probes.com

Complex shear modulus $G^*(\omega)$

$$\sigma = G^* \varepsilon$$

- $G^*(\omega) = G'(\omega) + j\,G''(\omega)$
- Solid *vs.* fluid
- Resistance to deformation

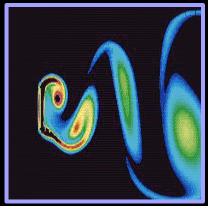


ciks.cbt.nist.gov

Storage modulus G'

Energy storage

Elasticity ~ Solid



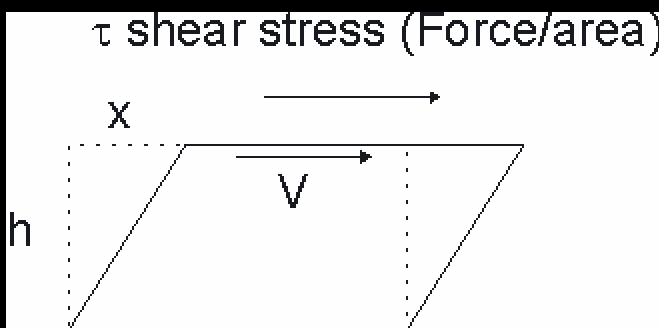
ma.man.ac.uk

Loss modulus G''

Energy dissipation

Viscosity ~ Fluid

Shear Modulus and Viscosity



Shear Modulus

$$\tilde{G} = \frac{\tilde{t}}{\tilde{x} / h}$$

Viscosity

$$\tilde{h} = \frac{\tilde{t}}{\tilde{v} / h} = \frac{\tilde{t}}{s\tilde{x} / h}$$

$$\tilde{G} = s\tilde{h}$$

Single Particle Tracking

Consider the thermal driven motion of a sphere in a complex fluid

Langevin Equation

$$m\dot{v}(t) = f(t) - \int_0^t x(t-t')v(t')dt'$$

Inertial force

Random thermal force

Memory function—
Material viscosity
Particle shape

Reminder of Laplace Transform

$$\tilde{f}(s) \equiv L[f(t)] \equiv \int_0^t f(t)e^{-st}dt$$

Definition of Laplace Transform
s is a complex number

$$L[f'(t)] = s\tilde{f}(s) - f(0)$$

Differentiation Rule

$$L(f * g(t)] = \tilde{f}(s)\tilde{g}(s)$$

Analog of Wiener-Khintchine Theorm

Laplace Transform of Langevin Equation

Laplace transforming Langevin's Equation:

$$L[m\dot{v}(t)] = L[f(t)] - L[\int_0^t x(t-t')v(t')dt']$$

$$ms\tilde{v}(s) - mv(0) = \tilde{f}(s) - \tilde{x}(s)\tilde{v}(s)$$

Collecting terms:

$$\tilde{v}(s) = \frac{\tilde{f}(s) + mv(0)}{ms + \tilde{x}(s)}$$

Langevin Equation in Frequency Domain

Laplace transform of Langevin Equation

$$\tilde{v}(s) = \frac{\tilde{f}(s) + mv(0)}{\tilde{x}(s) + ms}$$

Multiple by v(0),
taking a time average,
Ignoring inertial term

$$\tilde{G}(s) = \frac{kT}{pas < \Delta\tilde{r}^2(s) >}$$

Random force

$$< \tilde{f}(s)v(0) >= 0$$

Equipartition of energy

$$m < v(0)v(0) >= kT$$

Generalized Stokes Einstein

$$x(s) = 6pa\tilde{h}(s) \qquad \tilde{G}(s) = s\tilde{h}(s)$$

Definition and Laplace transform
of mean square displacement

$$< v(0)\tilde{v}(s) >= s^2 < \Delta\tilde{r}^2(s) > / 6$$

$$< \Delta\tilde{r}^2(s) >= L[< \sum_{i=1,2,3} (x_i(t) - x_i(0))^2 >]$$

Consider a simple example

$$<\Delta\tilde{r}^2(s)>\propto \frac{1}{s\tilde{G}(s)} \propto \frac{1}{s^2\tilde{h}(s)}$$

Purely viscose

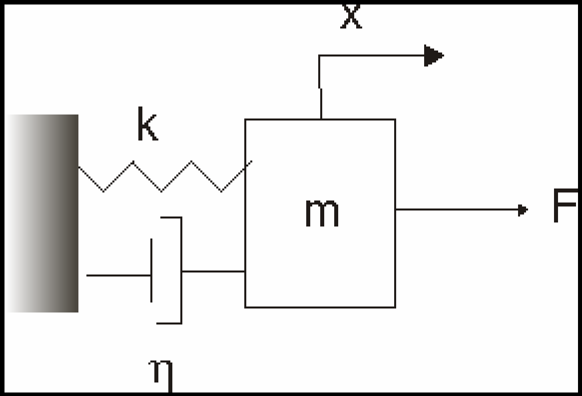
$$\tilde{h}(s) = h$$

$$<\Delta\tilde{r}^2(s)>\propto \frac{1}{s^2 h} \Rightarrow <\Delta r^2(t)>\propto \frac{t}{h}$$

Purely elastic

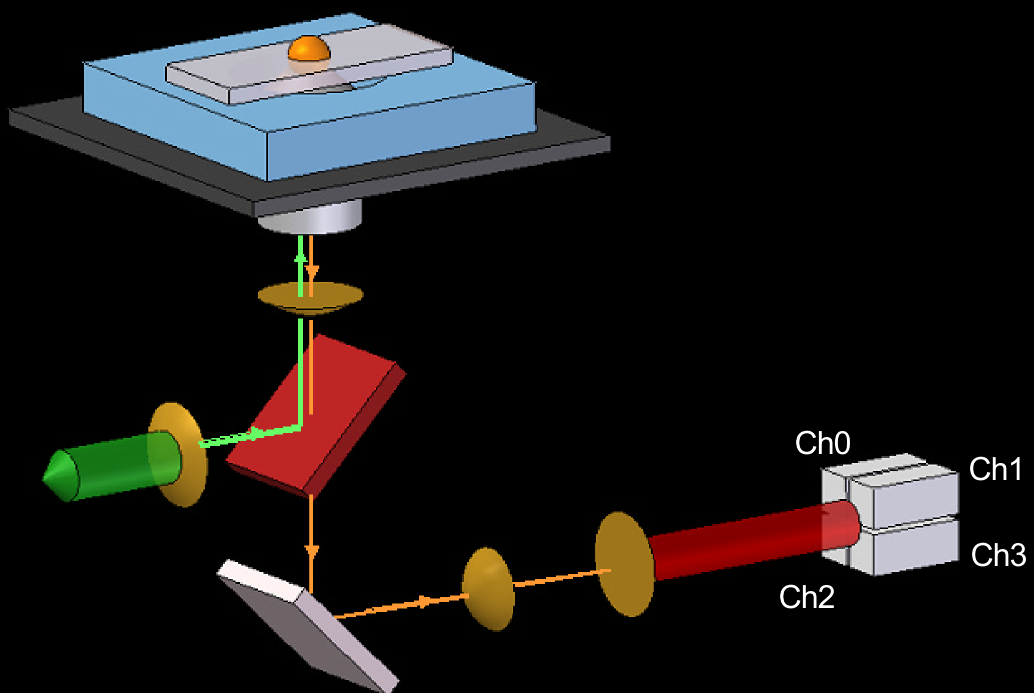
$$\tilde{h}(s) = \frac{1}{s}k \quad (F = k\int v dt)$$

$$<\Delta\tilde{r}^2(s)>\propto \frac{1}{sk} \Rightarrow <\Delta r^2(t)>\propto \frac{1}{k}$$

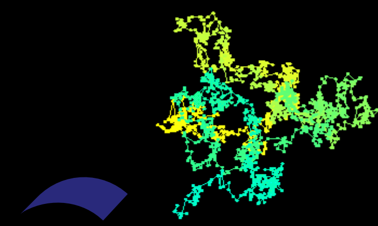


(2) Fluorescence Laser Tracking Microrheometer

- Approach: Monitoring the Brownian dynamics of particles embedded in a viscoelastic material to probe its frequency-dependent rheology



Yamada, Wirtz, Kuo, *Biophys. J.* 2000



trajectory

mean squared displacement

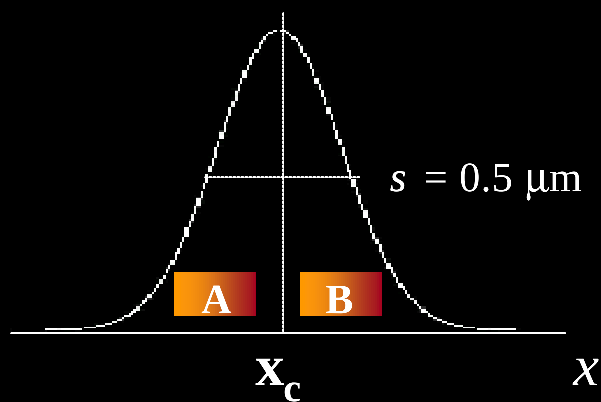
$$\left\langle \Delta R^2(\tau) \right\rangle = \left\langle \left(\vec{r}(t+\tau) - \vec{r}(t)\right)^2 \right\rangle$$

shear modulus

$$G^*(i\omega) = \frac{2k_BT}{3\pi \cdot a \cdot i\omega \cdot \left\langle \Delta\widetilde{R}^2(i\omega) \right\rangle}$$

(2) Nanometer Resolution for the Bead's Trajectory

- Collecting enough light from a fluorescent bead is critical

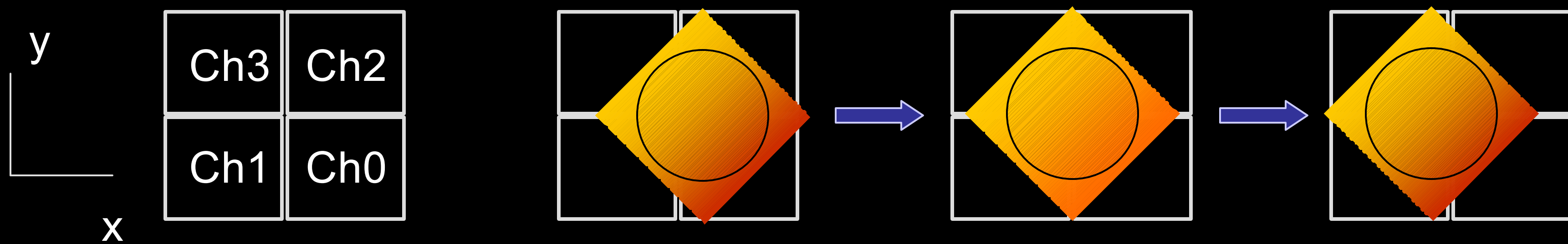


- $$p(x) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left[-\frac{(x-x_c)^2}{2\sigma^2}\right]$$
- $$\frac{N_A}{N_B} = \frac{p(A)}{p(B)} = \frac{\int_{-\infty}^{x_c} \exp\left(\frac{-x^2}{2\sigma^2}\right)dx}{\int_{x_c}^{+\infty} \exp\left(\frac{-x^2}{2\sigma^2}\right)dx}$$

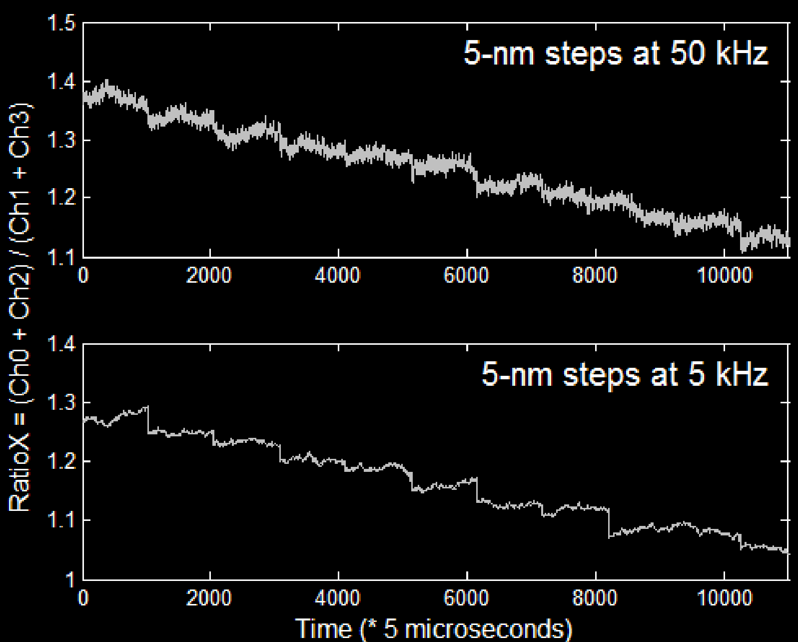
Photons detected per measurement	10^3	10^4	10^5	10^6
Uncertainty on $\frac{N_A}{N_B}$	0.033	0.010	0.003	0.001
Uncertainty on x_c (nm)	12	4	1.2	0.4

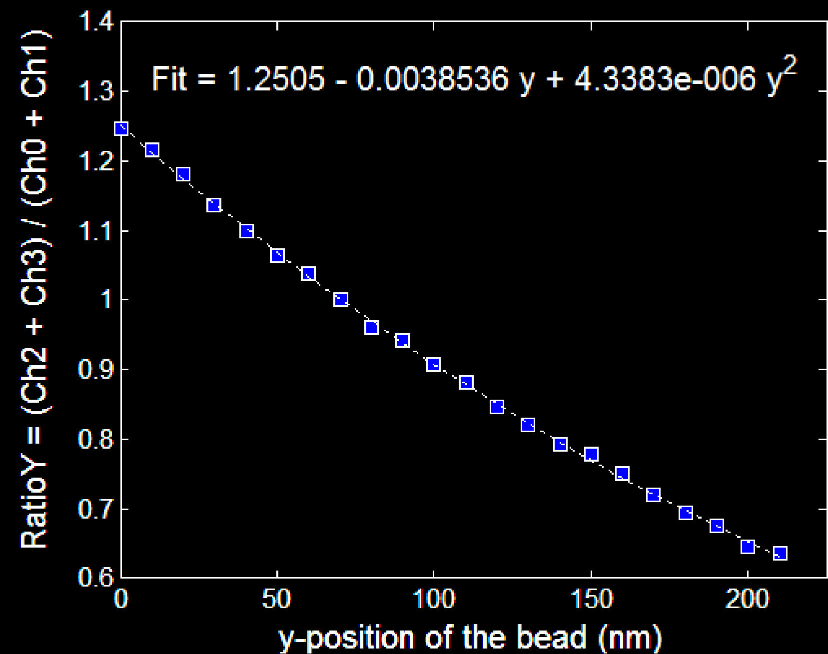
Nanometer resolution ↔ 10^4 photons per measurement

(2) Calibrating the FLTM

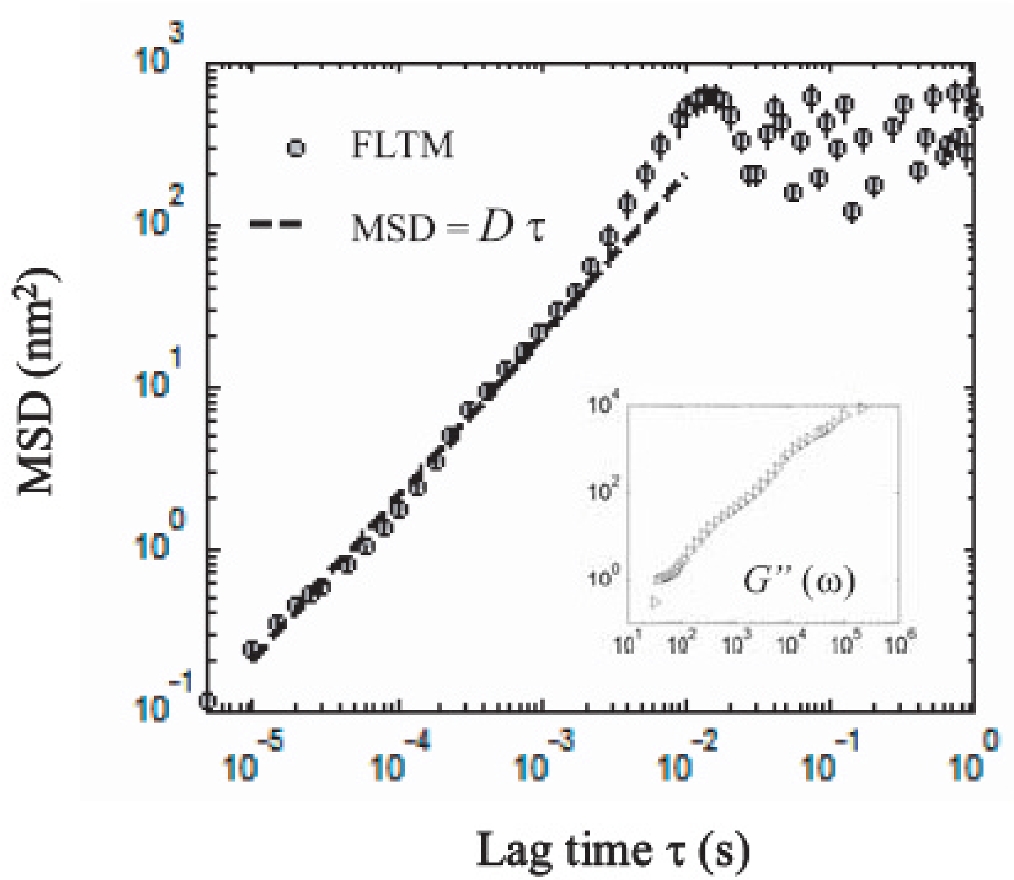


- 5-nm stepping at 5 or 50 kHz
- Curve fitting matches theory



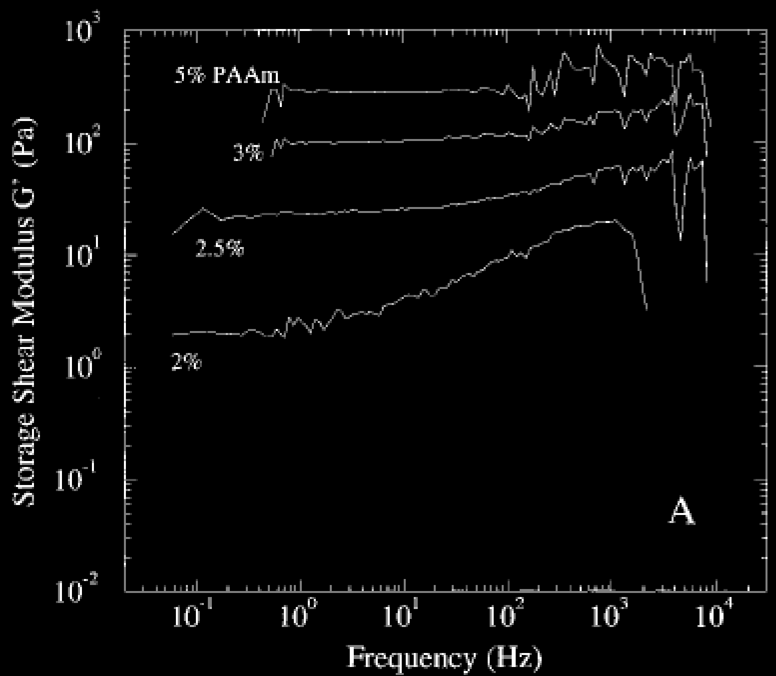


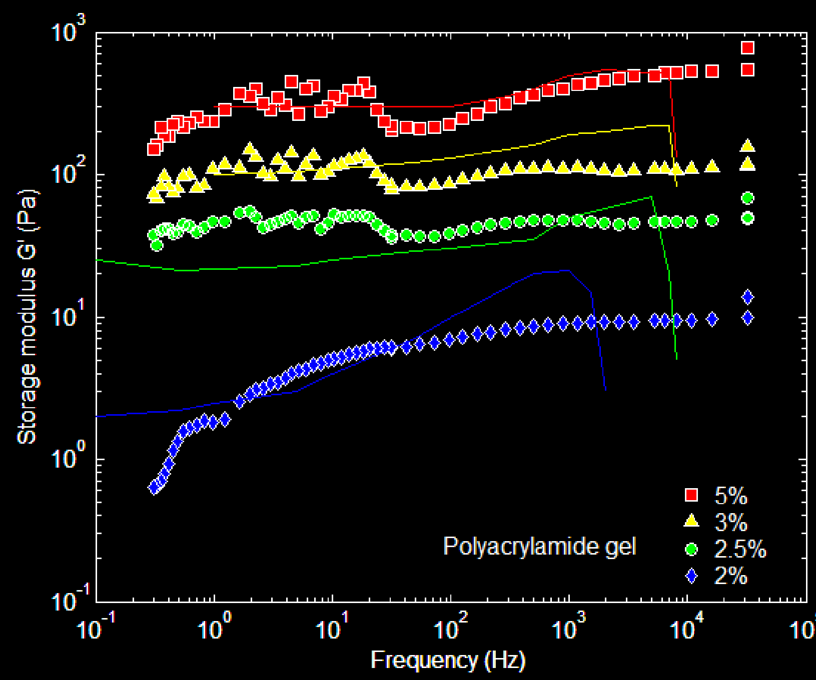
Diffusion in Glycerol



Characterizing the FLTM

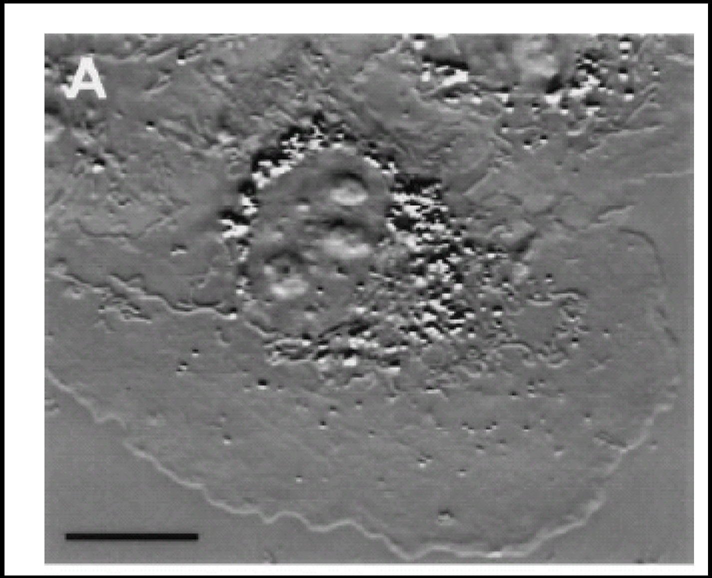
- Using polyacrylamide gels (w/v 2% to 5%) of known properties
- ✓ Good agreement with previously published data

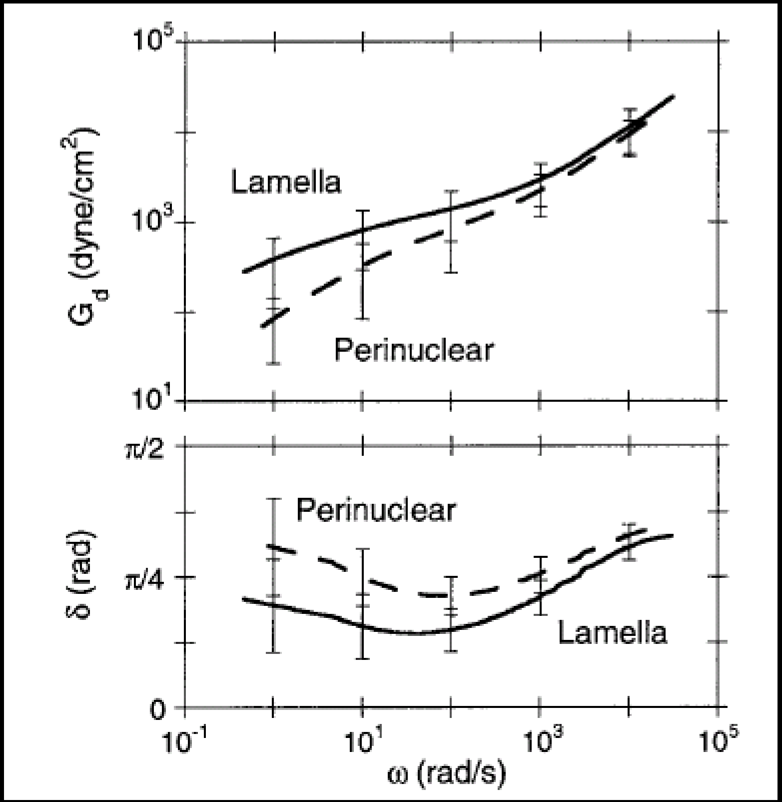




Schnurr B., Gittes F., MacKintosh F.C. & Schmidt C.F
Macromolecules (1997), **30**, p.7781-7792

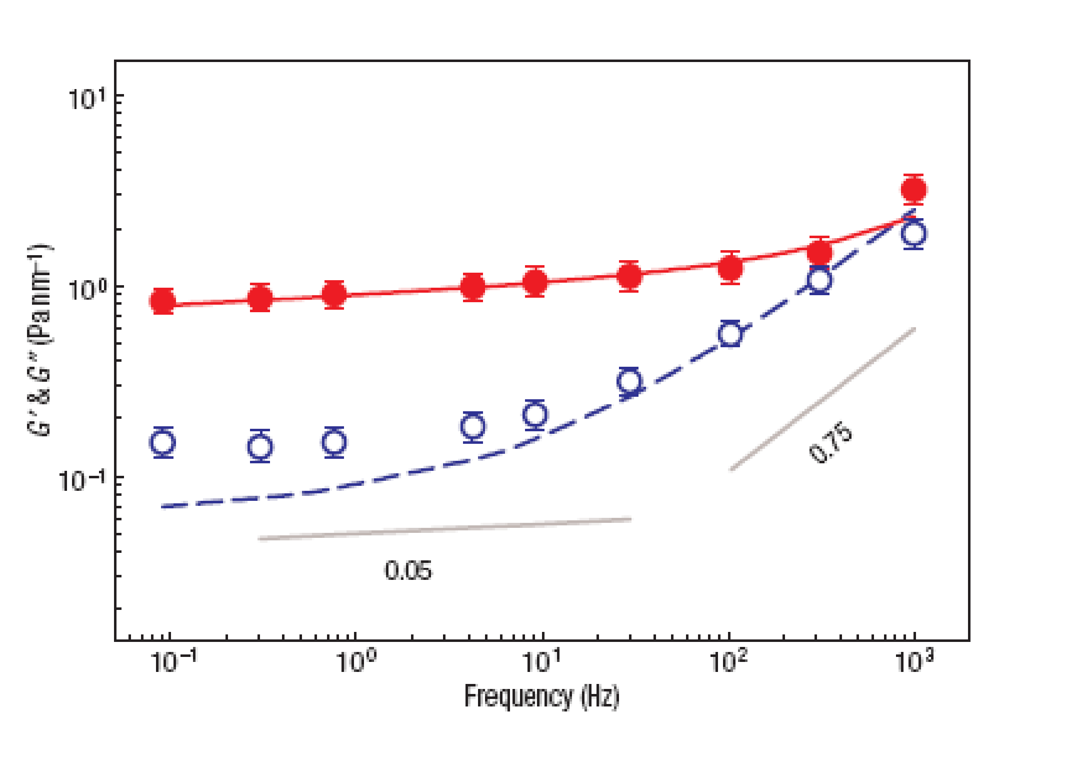
Single Particle Tracking Data





Yamada BJ 2000

Understanding Cell Mechanics



Deng, Nature Material, 2006