Dynamical slowdown of polymers in laminar and random flow

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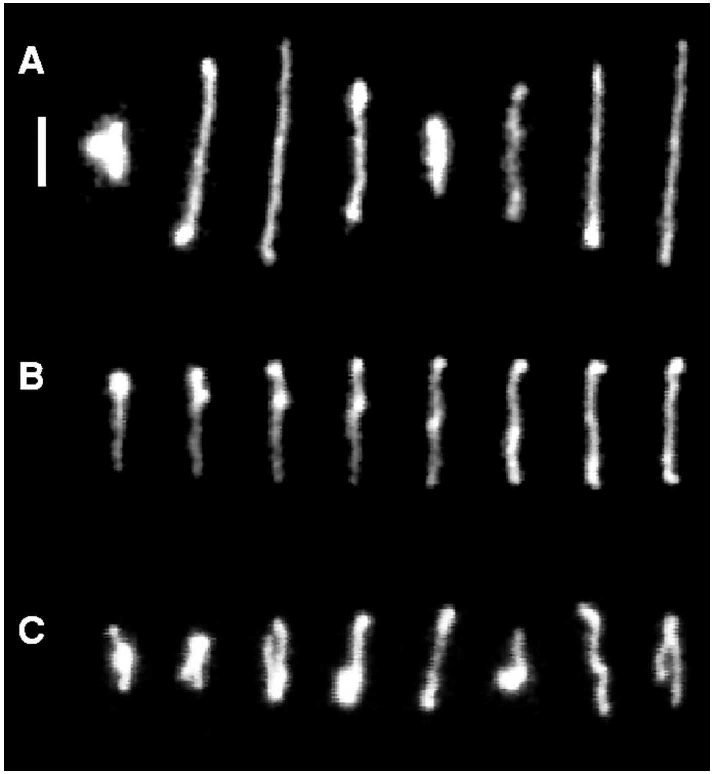
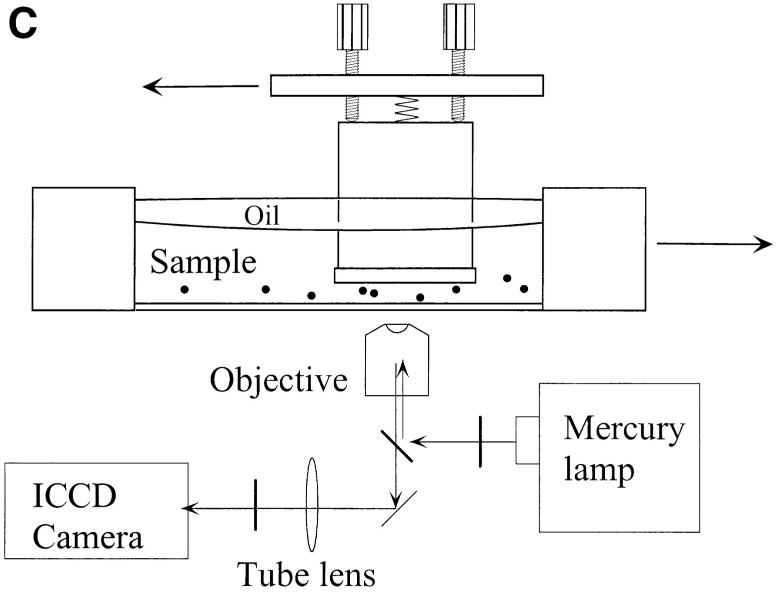
Single-polymer dynamics in flow



Smith, Babcock & Chu

Science (1999)



DNA

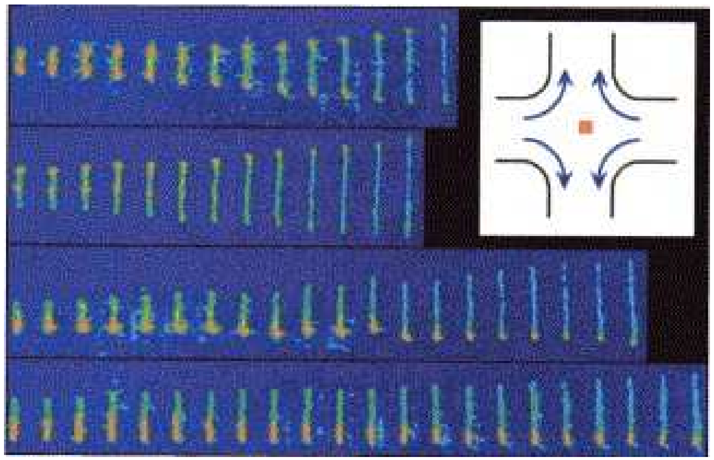
maximum extension

$L \approx 22 - 1300\ \mu m$

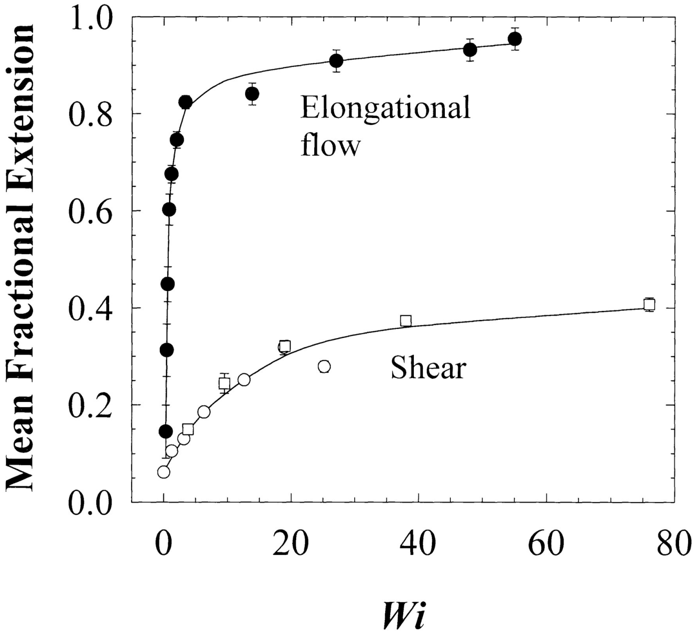
equilibrium extension

$R_0 \approx 1.8 - 15\ \mu m$

Coil-stretch transition – *extensional flow*



Perkins, Smith & Chu
Science (1997)



Smith, Babcock & Chu, *Science* (1999)

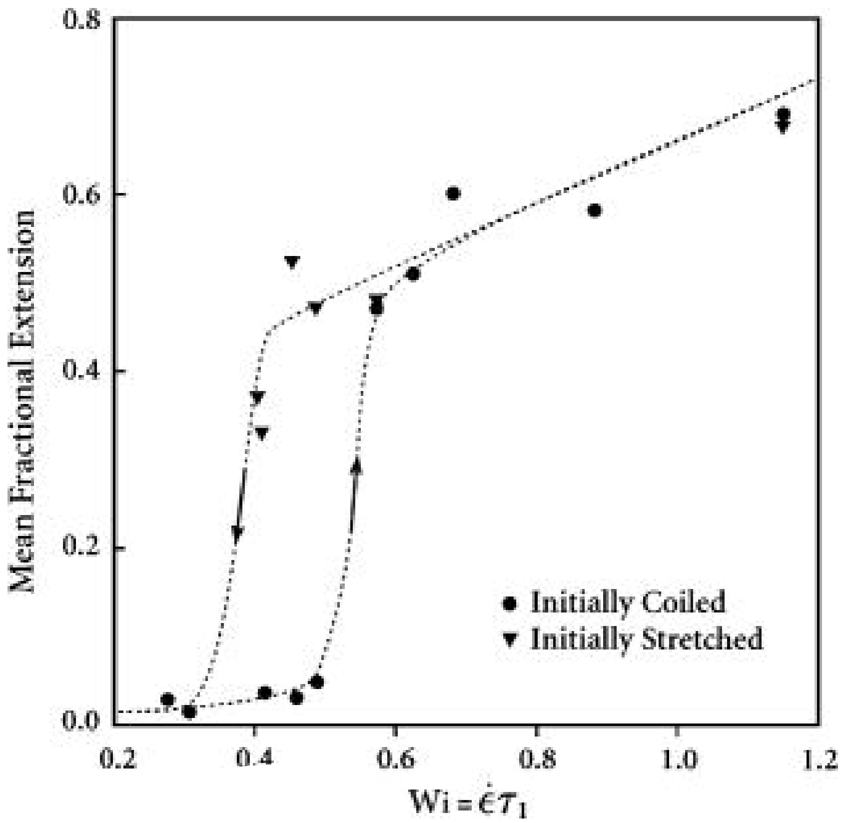
Weissenberg number

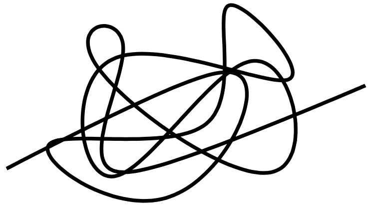
$$Wi = \gamma\tau$$

Conformation hysteresis – *extensional flow*

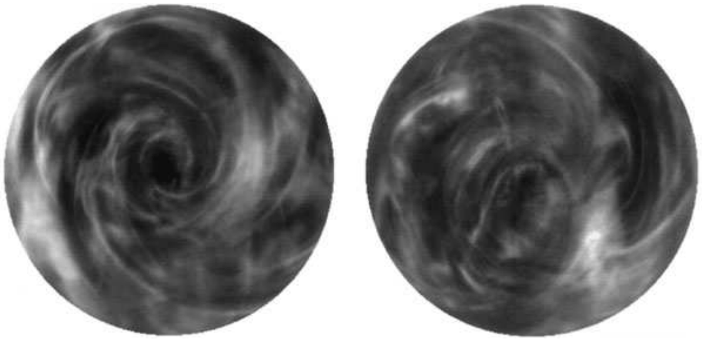
Schroeder et al., *Science* (2003)

Escherichia Coli DNA ($L = 1300\,\mu m$)

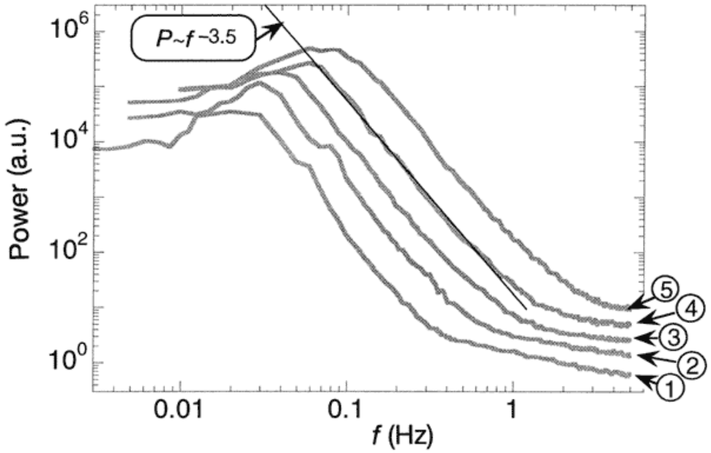




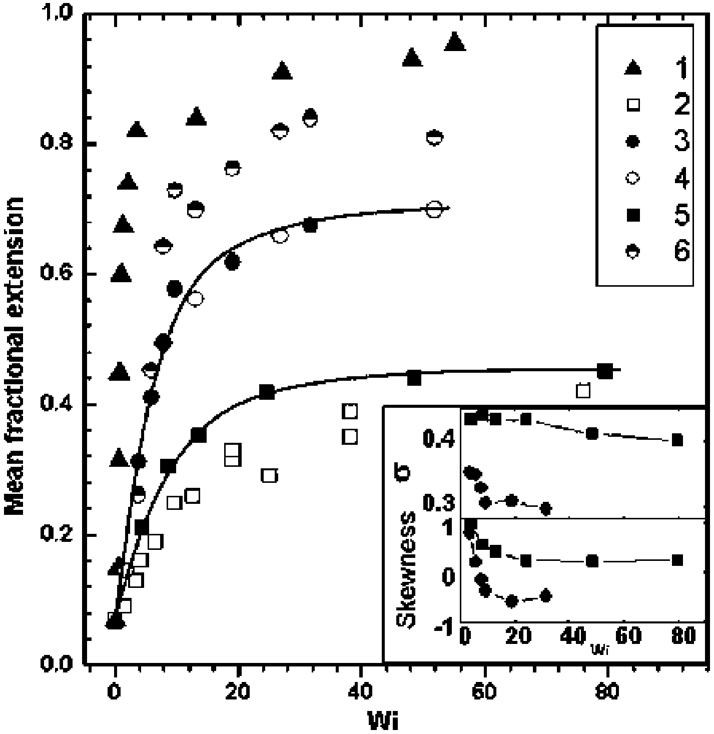
Coil-stretch transition – *random flow*



$Wi = 13$, $Re = 0.7$

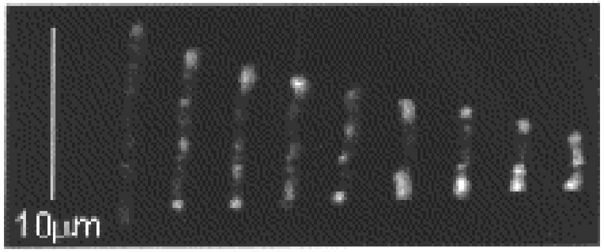


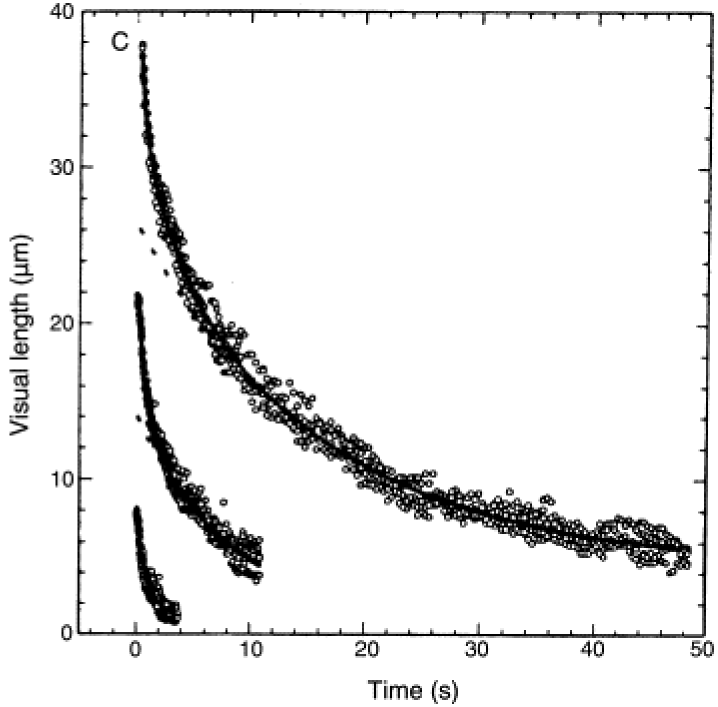
Groisman & Steinberg, *Nature* (2000)



Gerashchenko et al.
Europhys. Lett. (2005)

Experimental measure of the relaxation time

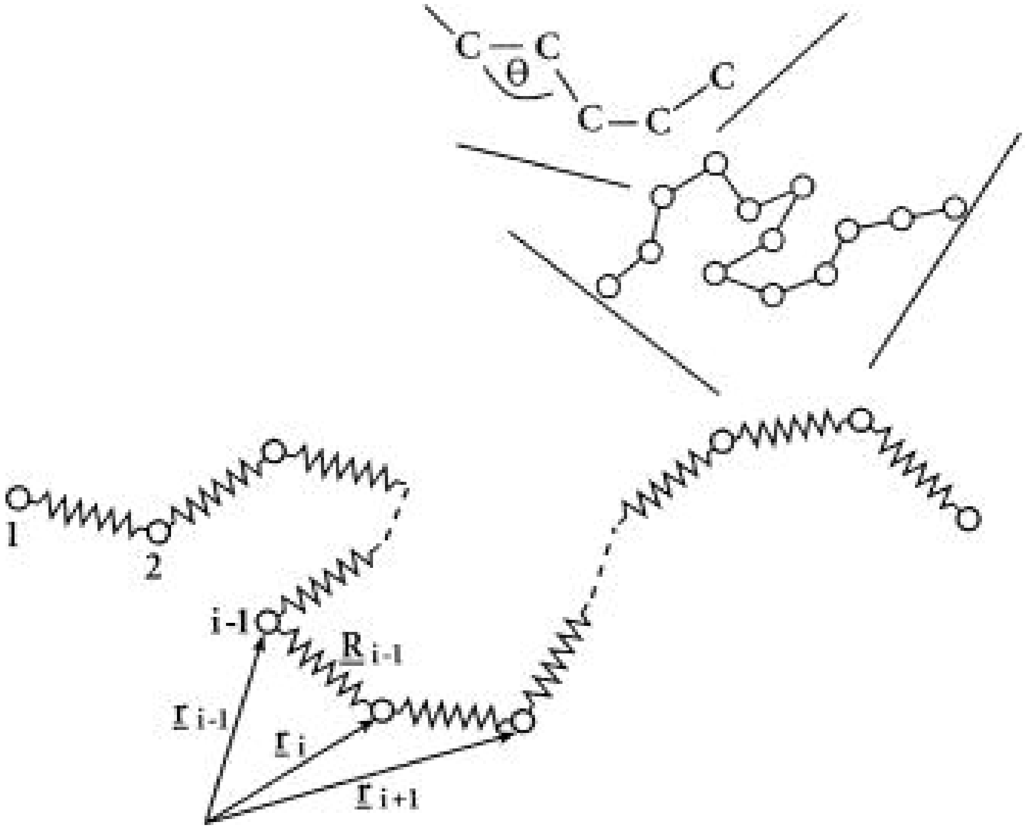




Perkins et al., *Science* (1994)

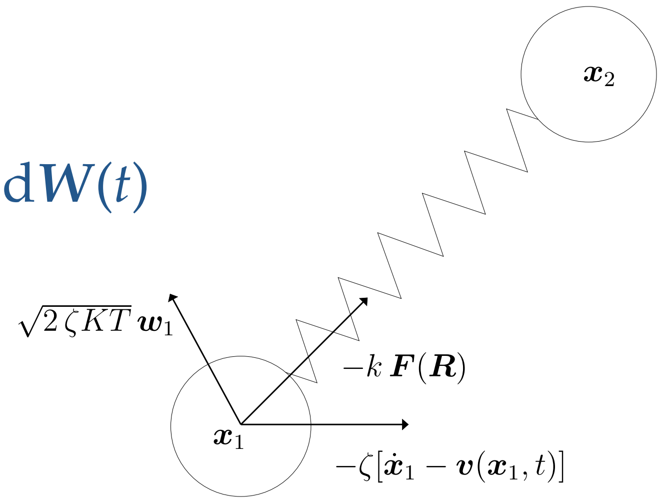
$$R(t) \approx R_{\text{in}} \exp(-t/\tau) + R_0$$

Rouse model



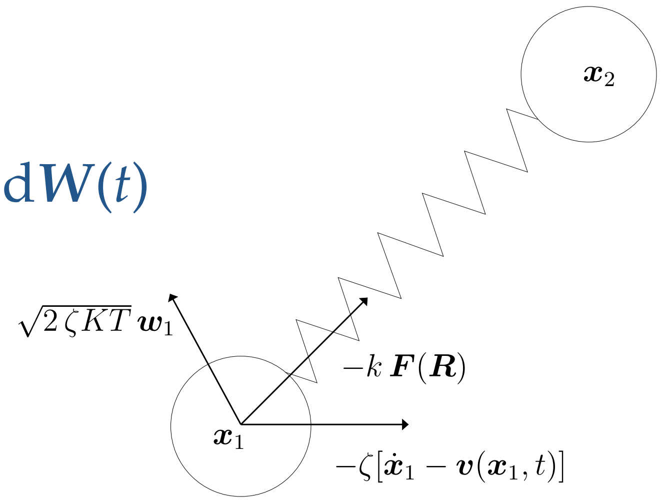
Dumbbell model

$$\mathrm{d}\boldsymbol{R} = \boldsymbol{R} \cdot \nabla \boldsymbol{v}(t)\,\mathrm{d}t - \frac{f(R)}{2\tau\nu(R)}\,\boldsymbol{R}\,\mathrm{d}t + \sqrt{\frac{R_0^2}{\tau\nu(R)}}\,\mathrm{d}\boldsymbol{W}(t)$$



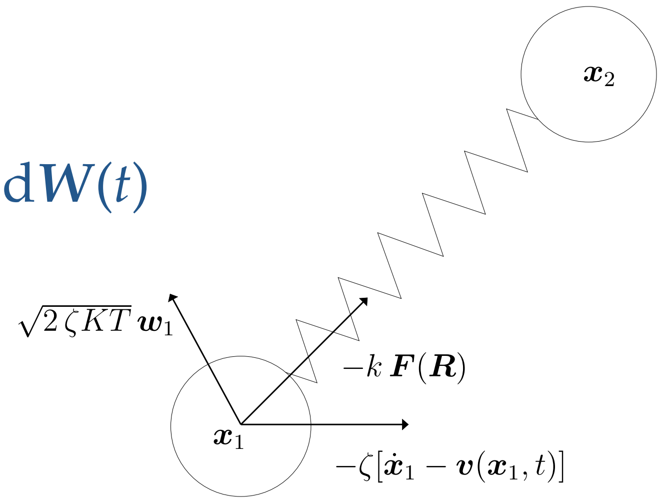
Dumbbell model

$$\mathrm{d}\boldsymbol{R} = \boldsymbol{R}\cdot\nabla v(t)\,\mathrm{d}t - \frac{f(R)}{2\tau\nu(R)}\,\boldsymbol{R}\,\mathrm{d}t + \sqrt{\frac{R_0^2}{\tau\nu(R)}}\,\mathrm{d}\boldsymbol{W}(t)$$



$$f(R) = \begin{cases} \dfrac{1}{1 - R^2/L^2} & \text{(FENE)} \\[2ex] \dfrac{2}{3} - \dfrac{L}{6R} + \dfrac{L}{6R(1 - R/L)^2} & \text{(WLC)} \end{cases}$$

Dumbbell model



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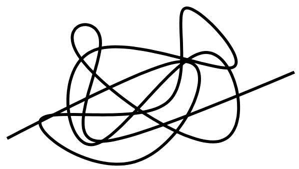
$$\tau \propto \zeta \qquad\qquad \nu(R) = 1 + \left(\frac{\zeta_s}{\zeta_c} - 1\right)\frac{R}{L}$$

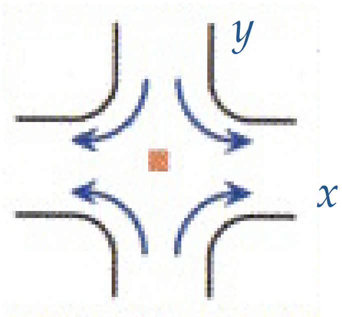


$$\zeta_c = \frac{3}{8}\sqrt{6\pi^3}R_0\eta$$

$$\zeta_s = \frac{2\pi L\eta}{\ln(L/\ell)}$$

Extensional flow

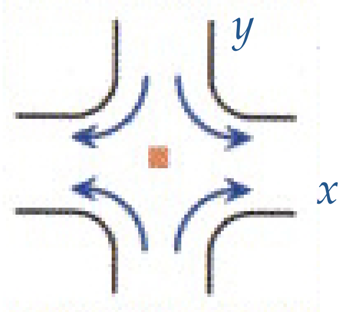
$$\boldsymbol{v}(x, y, z) = \gamma(x, -y, 0)$$



$$R \approx R_x$$

Extensional flow

$\boldsymbol{v}(x, y, z) = \gamma(x, -y, 0)$



$R \approx R_x$

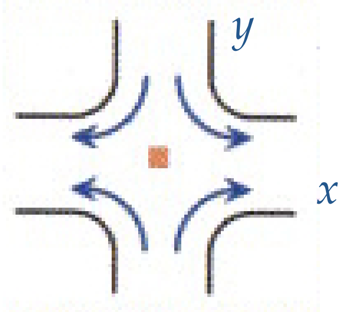
Fokker–Planck equation for $P(r, t')$ $\quad (t' = t/\tau,\ r = R/L)$

$$\partial_{t'} P = \mathscr{L}_{\mathrm{FP}} P \qquad \mathscr{L}_{\mathrm{FP}} \cdot = -\partial_r (D_1(r)\ \cdot) + \partial_r D_2(r) \partial_r\ \cdot$$

$$D_1 = Wi\, r - f(r) r / [2\nu(r)] \qquad D_2(r) = [2b\nu(r)]^{-1} \qquad b = L^2/R_0^2 \qquad Wi = \gamma\tau$$

Extensional flow

$$\boldsymbol{v}(x,y,z) = \gamma(x,-y,0)$$



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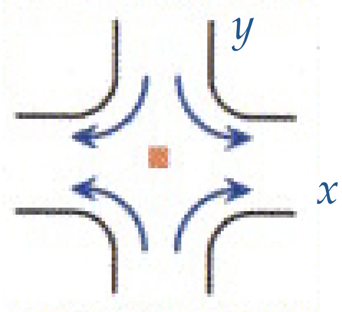
$$D_1 = Wi\, r - f(r) r/[2\nu(r)] \qquad D_2(r) = [2b\nu(r)]^{-1} \qquad b = L^2/R_0^2 \qquad Wi = \gamma\tau$$

Stationary distribution

$$P_{\rm st}(r) = N e^{-E(r)/KT} \qquad E(r) = -KT \int^r D_1(\rho)/D_2(\rho)\, \mathrm{d}\rho$$

Extensional flow

$$\boldsymbol{v}(x,y,z) = \gamma(x,-y,0)$$



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Stationary distribution

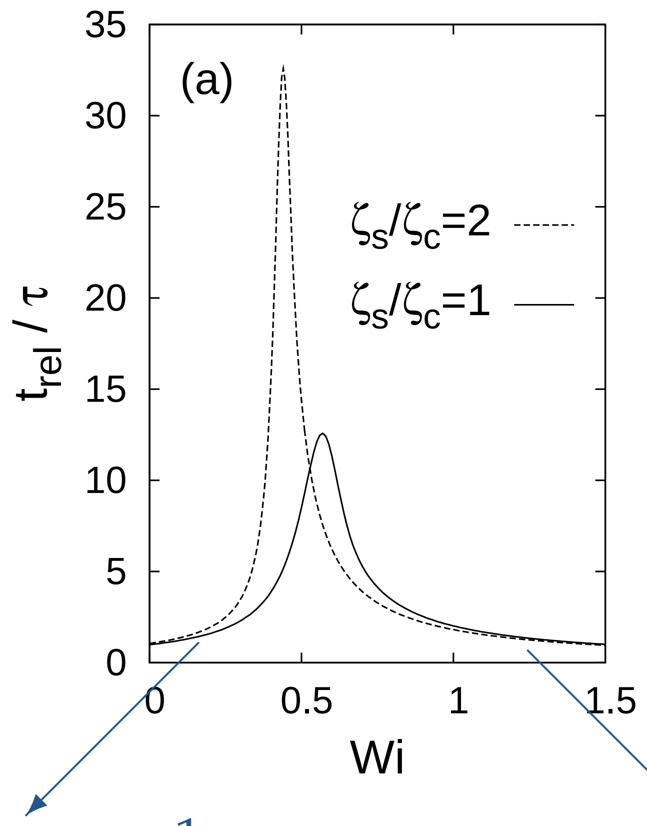
$$P_{\mathrm{st}}(r) = N e^{-E(r)/KT} \qquad E(r) = -KT \int^r D_1(\rho)/D_2(\rho)\, \mathrm{d}\rho$$

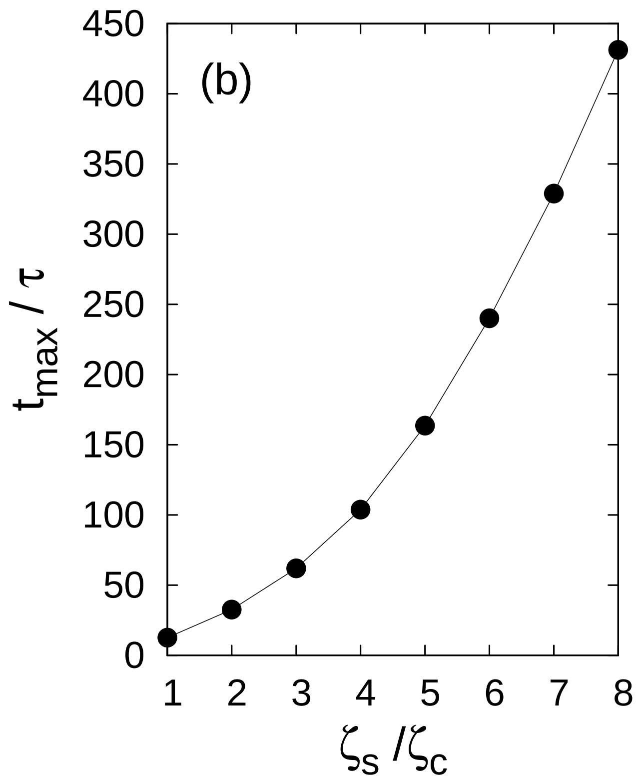
Relaxation time

$$P(r,t') = P_{\mathrm{st}}(r) + \sum_{n=1}^{\infty} a_n p_n(r)\, e^{-\sigma_n t'} \qquad (\sigma_n < \sigma_{n+1})$$

$$t_{\mathrm{rel}} \equiv \tau/\sigma_1$$

Extensional flow – *relaxation time*





$$t_{\mathrm{rel}}/\tau \sim \frac{1}{(1-2Wi)}$$

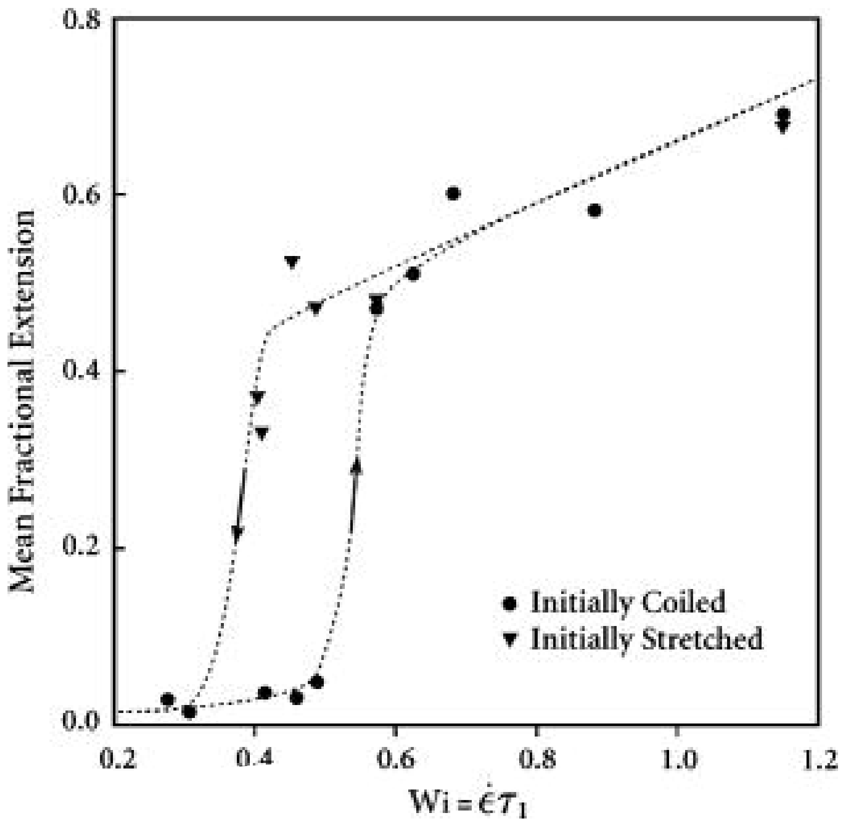
$$t_{\mathrm{rel}}/\tau \sim Wi^{-1}$$

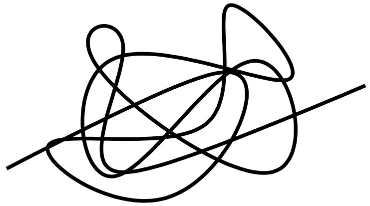
$$b = L^2/R_0^2 = 400$$

Extensional flow – *conformation hysteresis*

Schroeder et al., *Science* (2003)

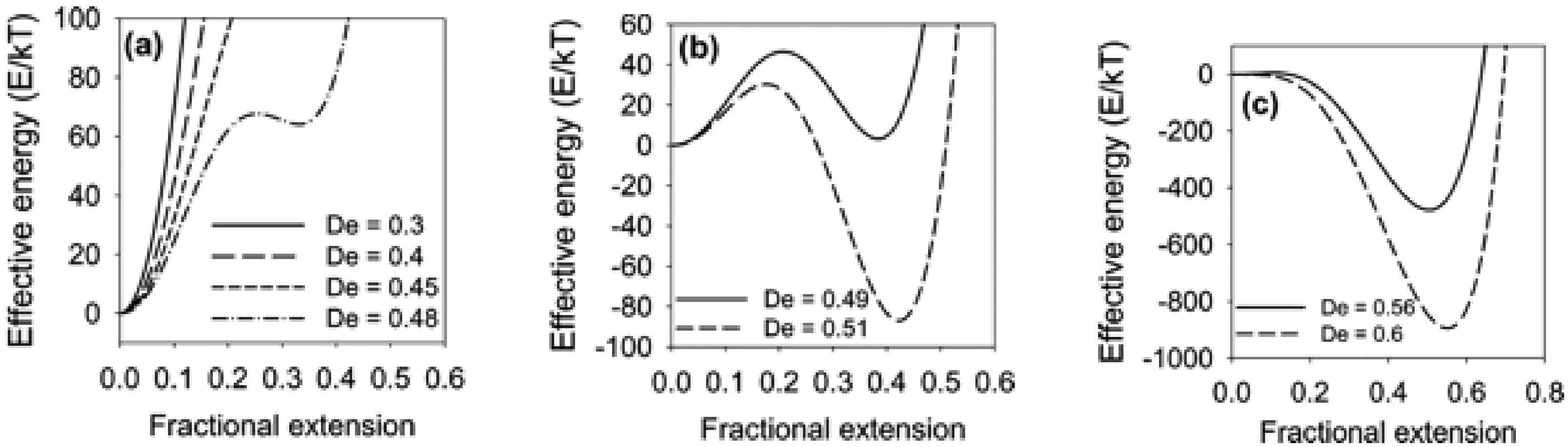
Escherichia Coli DNA ($L = 1300\,\mu m$)



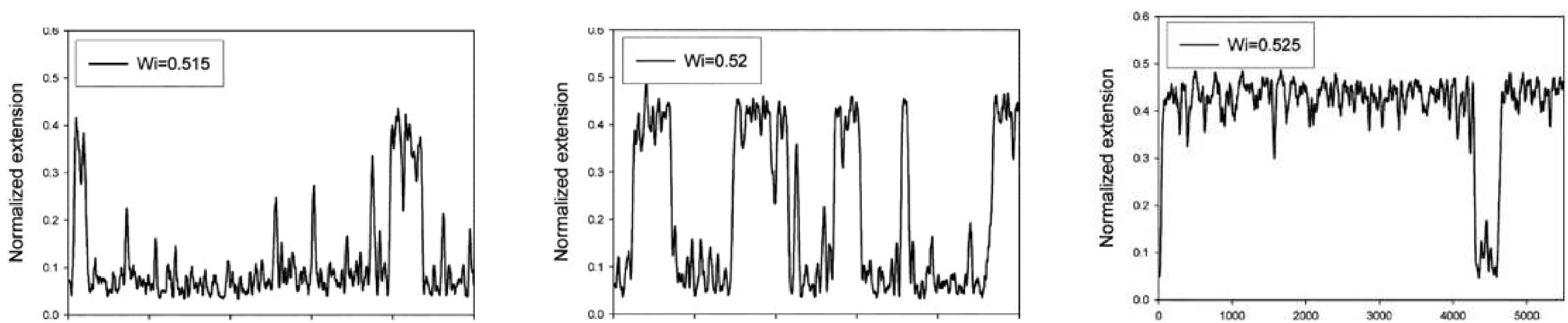


Extensional flow – *conformation hysteresis*

$$P_{\mathrm{st}}(r) = Ne^{-E(r)/KT}$$



Schroeder, Shaqfeh & Chu, *Macromolecules* (2004)



Hsieh & Larson, *J. Rheol.* (2005)

Smooth random flow

3D Batchelor-Kraichnan flow

$$\langle \partial_j v_i(t)\, \partial_k v_l(s) \rangle \;=\; \lambda\, C_{ijkl}\, \delta(t-s)$$

Fokker-Planck equation

$$\partial_{t'} P \;=\; -\partial_r (D_1(r) P) \;+\; \partial_r D_2(r) \partial_r P$$

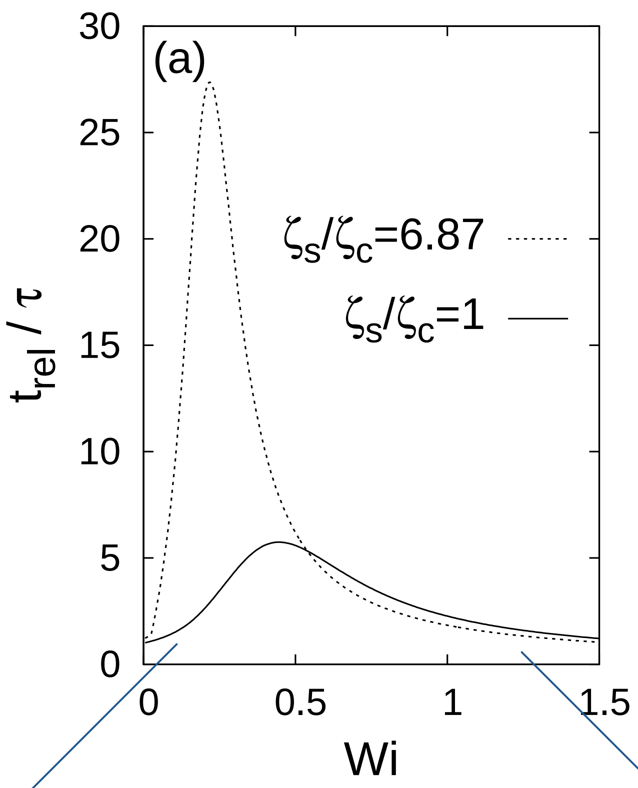
$$D_1(r) = \tfrac{2}{3} Wi\, r - f(r) r / [2\nu(r)] + [b\nu(r) r]^{-1} \qquad D_2(r) = [2b\nu(r)]^{-1} + \tfrac{1}{3} Wi\, r^2$$

$$Wi = \lambda\tau \qquad b = L^2/R_0^2$$

Relaxation time

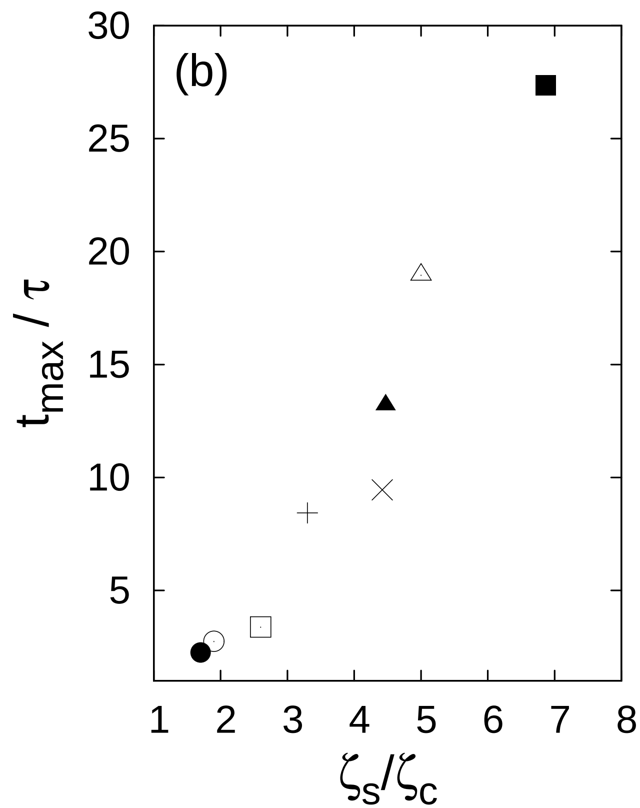
$$P(r,t') = P_{\rm st}(r) + \sum_{n=1}^{\infty} a_n p_n(r)\, e^{-\sigma_n t'}$$

Random flow – *relaxation time*



$t_{\rm rel}/\tau \sim (1 - \frac{5}{3}Wi)^{-1}$

$t_{\rm rel}/\tau \sim Wi^{-1}$



DNA (●, $b = 191.5$; ○, $b = 260$; □, $b = 565$; +, $b = 2250$)

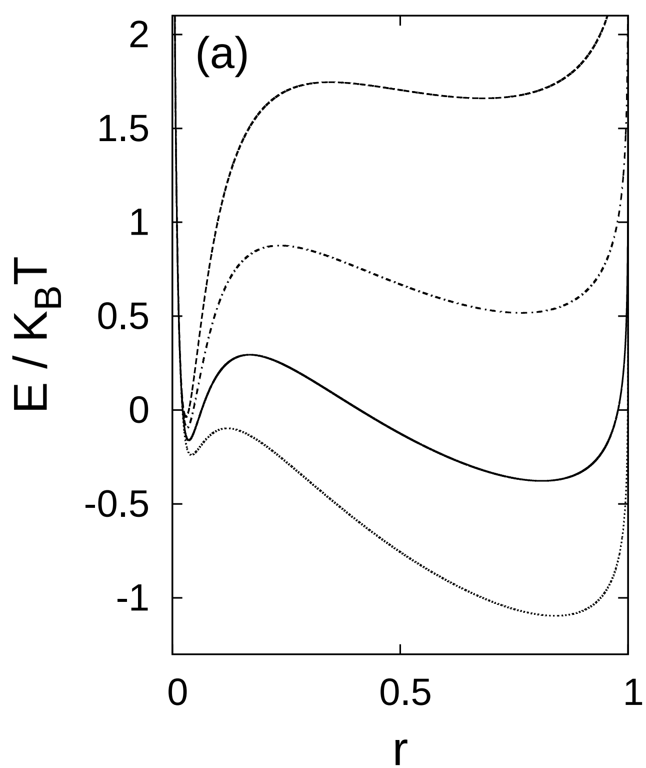
polystyrene (×, $b = 673$)

PEO (▲, $b = 1666$)

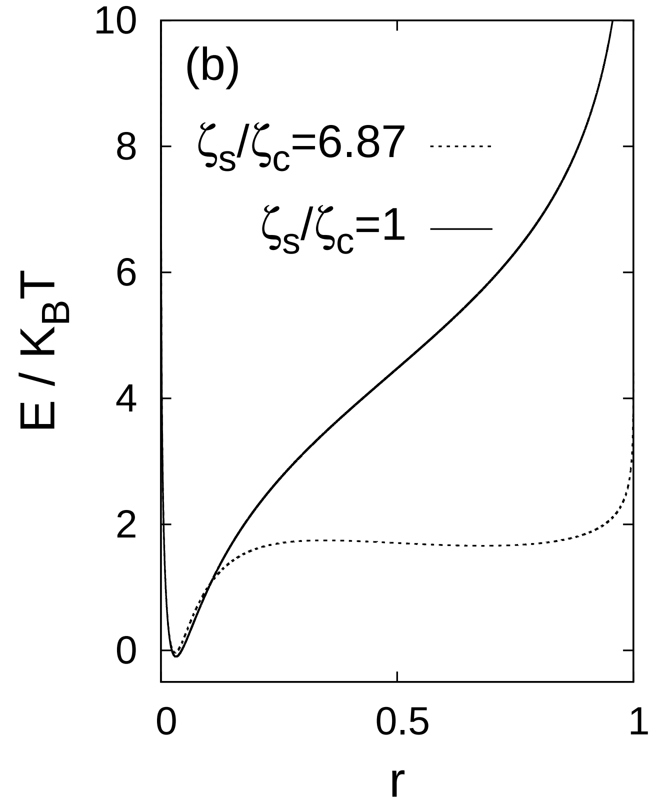
E. Coli DNA (△, $b = 9250$)

PAM (■, $b = 3953$,)

Random flow – *stationary pdf*



from top to bottom:
$Wi = 0.28, 0.33, 0.38, 0.43$



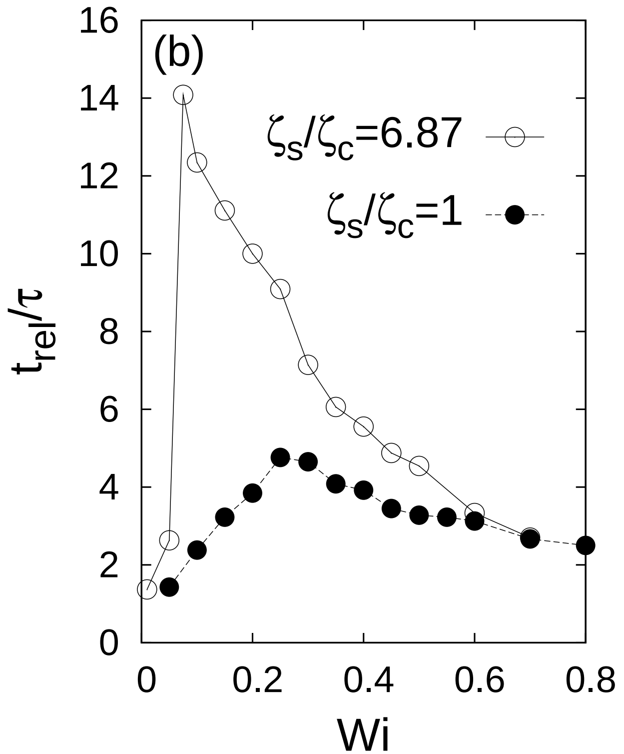
$Wi = 0.28$

polyacrylamide (PAM), $b = L^2/R_0^2 = 3953$, $\zeta_s/\zeta_c = 6.87$

Random flow – *BD simulations*

Brun–Koch–Lion flow (*Phys. Fluids*, 1997)

$$\langle \partial_j v_i(t)\,\partial_k v_l(0)\rangle \;=\; S_{ijkl}\,\exp\Big(-\frac{|t|}{T_S}\Big) + R_{ijkl}\,\exp\Big(-\frac{|t|}{T_R}\Big)$$



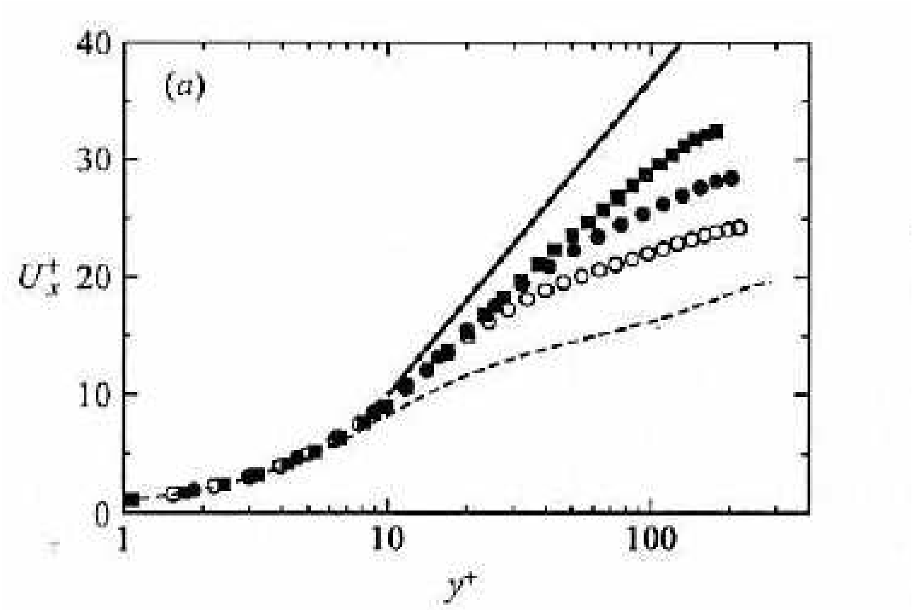
$$T_S = 2.3\tau_\eta, T_R = 7.2\tau_\eta$$

$$t_{\rm rel}^{-1} = -\lim_{t\to\infty} \ln[\langle r^2(t)\rangle - \langle r^2\rangle_{\rm st}]$$

Conclusions

- τ is not the relevant time scale for coil–stretch processes
- no hysteresis in smooth random flows
- the conformation-dependent drag is a basic ingredient of continuum models of polymer solutions

FENE-P model

- 

■ DR 60%
● DR 47%
○ DR 35%
- - Newtonian

Dubief et al., *J. Fluid Mech.* (2004)

$$C_{ij} \equiv \overline{R_i R_j}$$

$$\partial_t \boldsymbol{C} + \boldsymbol{v} \cdot \nabla \boldsymbol{C} = \boldsymbol{C} \cdot \nabla \boldsymbol{v} + (\nabla \boldsymbol{v})^{\mathrm{T}} \cdot \boldsymbol{C} - \frac{1}{\tau} [\hat{f}(\mathrm{Tr}\boldsymbol{C})\boldsymbol{C} - \boldsymbol{I}]$$