One- and Two-Particle Microrheology

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Outline

Sphere Through a Fluid

Given:

Navier-Stokes Equation:

$$\rho \frac{\partial \vec{v}}{\partial t} = -\vec{\nabla} p + \eta \nabla^2 \vec{v} - \rho \vec{v} \cdot \vec{\nabla} \vec{v} + \vec{f}$$

$$\downarrow$$

$$\vec{\nabla} p \approx \eta \nabla^2 \vec{v}$$

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$$\downarrow$$

$$\vec{\nabla} p \approx \eta \nabla^2 \vec{v}$$

BCs:

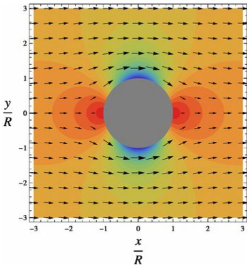
$$\begin{cases} v_r = 0 & \text{at } r = infty \\ v_\theta = 0 & \text{at } r = \infty \\ v_r = -V \cos\theta & \text{at } r = R \\ v_\theta = V \sin\theta & \text{at } r = R \end{cases}$$

Sphere Through a Fluid

Solution:

$$v_r = \frac{V}{2}\left[3\left(\frac{R}{r}\right) - \left(\frac{R}{r}\right)^3\right]\cos\theta$$

$$v_\theta = -\frac{V}{4}\left[3\left(\frac{R}{r}\right) + \left(\frac{R}{r}\right)^3\right]\sin\theta$$



Flow Past a Sphere

Given:

Can either reset BCs and solve again **or**

Flow Past a Sphere

Given:

Can either reset BCs and solve again **or** superimpose uniform flow (in spherical coordinates) on to our solution of a sphere moving through a fluid.

Solution:

$$v_r = -V\left[1 - \frac{3}{2}\left(\frac{R}{r}\right) + \frac{1}{2}\left(\frac{R}{r}\right)^3\right]\cos\theta$$

$$v_\theta = V\left[1 - \frac{3}{4}\left(\frac{R}{r}\right) + \frac{1}{4}\left(\frac{R}{r}\right)^3\right]\sin\theta$$

Drag

Technique:

Having found the velocity, one can get the pressure from the Navier-Stokes Eq. The force of the fluid on the sphere is then the integral of the pressure over the total surface area.

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Solution:

$$\vec{F} = 6\pi\eta R\vec{V}$$
$$= \xi\vec{V}$$

ξ is call the friction coefficient.

"Falling Ball" Rheology

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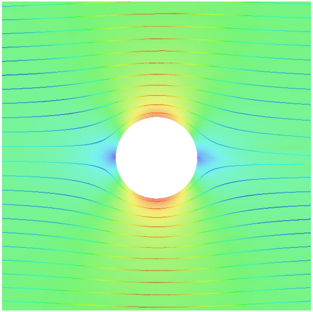
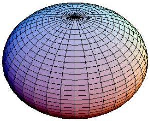
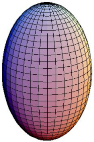
Give $\vec{F}$, R and measuring $\vec{V}$ one can determine η.



NonSpherical Objects





Orientation

Now the drag depends on the orientation suggesting

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$$F_i = 6\pi\eta R_{ij} V_j$$

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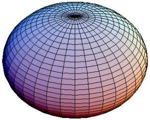

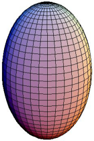

Orientation

Now the drag depends on the orientation suggesting

$$F_i = 6\pi\eta R_{ij} V_j$$

Translation Tensor

For a rigid body, $\hat{R}$ depends solely on the size and shape of the object. For a sphere, $R_{ij} = R\delta_{ij}$.

Friction

Grouping Translation Tensor and Viscosity

$$\hat{\xi} \equiv 6\pi\eta\hat{R}$$

Why was this a good idea?

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Why was this a good idea?
No-slip at spherical surface:

$$\vec{F} = 6\pi\eta R\vec{V}$$

Friction

Grouping Translation Tensor and Viscosity

$$\hat{\xi} \equiv 6\pi\eta\hat{R}$$

Why was this a good idea?
Perfect-slip at spherical surface:

$$\vec{F} = 4\pi\eta R\vec{V}$$

Friction

Grouping Translation Tensor and Viscosity

$$\hat{\xi} \equiv 6\pi\eta\hat{R}$$

Why was this a good idea?
General-slip at spherical surface:

$$\vec{F} = 6\pi\eta R\left(\frac{\beta R + 2\eta}{\beta R + 3\eta}\right)\vec{V}$$

Friction

Grouping Translation Tensor and Viscosity

$$\hat{\xi} \equiv 6\pi\eta\hat{R}$$

Why was this a good idea?
Spherical Liquid Droplet:

$$\vec{F} = 6\pi\eta R \left(\frac{\epsilon/R + 2\eta_o + 3\eta_i}{\epsilon/R + 3\eta_o + 3\eta_i} \right) \vec{V}$$

Friction

Grouping Translation Tensor and Viscosity

$$\hat{\xi} \equiv 6\pi\eta\hat{R}$$

Why was this a good idea?

Because the particle's interaction with the fluid requires definition

$$\vec{F} \equiv \hat{\xi}\vec{V}$$

$$\hat{\xi} = 6\pi k\eta\hat{R}$$

where k is any correction term to Stokes drag.

Elipsoids

Elipsoid (Return to translation tensor for a moment)

$$\hat{R} = \begin{pmatrix} R_{1,1} & R_{1,2} & R_{1,3} \\ R_{2,1} & R_{2,2} & R_{2,3} \\ R_{3,1} & R_{3,2} & R_{3,3} \end{pmatrix}$$

Can always rotate to principle moments (think moment of inertia tensor)

Elipsoids

Elipsoid (Return to translation tensor for a moment)

$$\hat{R} = \begin{pmatrix} R_1 & 0 & 0 \\ 0 & R_2 & 0 \\ 0 & 0 & R_3 \end{pmatrix}$$

R_i are the principle translation coefficients.

Elipsoids

Elipsoid (Return to translation tensor for a moment)

$$R_1 = \frac{8}{3}\frac{a^2 - b^2}{\left(2a^2 - b^2\right) S - 2a}$$

$$R_2 = R_3$$

$$= \frac{16}{3}\frac{a^2 - b^2}{\left(2a^2 - 3b^2\right) S - 2a}$$

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Prolate

For $a > b$:

$$S = 2\left(a^2 - b^2\right)^{-1/2}\ln\left[\frac{a + \left(a^2 - b^2\right)^{1/2}}{b}\right]$$

$a \gg b \rightarrow$ rod.

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$$= \frac{16}{3}\frac{a^2 - b^2}{\left(2a^2 - 3b^2\right) S - 2a}$$

Oblate

For $a < b$:

$$S = 2\left(a^2 - b^2\right)^{-1/2} \tan^{-1}\left[\frac{a + \left(a^2 - b^2\right)^{1/2}}{a}\right]$$

$b \gg a \rightarrow$ disk.

Mean Friction

Mean Translation Coefficient

$$\frac{1}{\langle R \rangle} = \frac{1}{3}\left(\frac{1}{R_1} + \frac{1}{R_2} + \frac{1}{R_3}\right)$$

which amounts to an equivalent radius.

Mean Friction Coefficient

$$\frac{1}{\langle \xi \rangle} = \frac{1}{3}\left(\frac{1}{\xi_1} + \frac{1}{\xi_2} + \frac{1}{\xi_3}\right)$$

where $1, 2, 3$ are the principle axes.

Perin Factor

Equivalent Sphere

The ratio of the mean translation coefficient to a sphere of the same volume is called the Perin Factor:

$$\mathcal{F} = \frac{\langle R \rangle}{R_{\text{sph}}} = \frac{\langle \xi \rangle}{\xi_{\text{sph}}}$$

Mobility

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$\hat{\mu}$'s relation to $\hat{\xi}$:

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$$\hat{\mu} \equiv \hat{\xi}^{-1}$$

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$$\frac{1}{\langle \xi \rangle} = \frac{1}{3}\left(\frac{1}{\xi_1} + \frac{1}{\xi_2} + \frac{1}{\xi_3}\right)$$

and $\mu = 1/\xi$ therefore ...

Mean Mobility

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and $\mu = 1/\xi$ therefore ...

Mean Mobility Coefficient

$$\langle \mu \rangle = \frac{1}{3}\left(\mu_1 + \mu_2 + \mu_3\right)$$

Oseen-Burgers Tensor

Components Notation

Consider the velocity of perturbed fluid due to the sphere's movement:

$$v_r = \frac{V}{2}\left[3\left(\frac{R}{r}\right) - \left(\frac{R}{r}\right)^3\right]\cos\theta$$

$$v_\theta = -\frac{V}{4}\left[3\left(\frac{R}{r}\right) + \left(\frac{R}{r}\right)^3\right]\sin\theta$$

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Vector Notation

$$\vec{v} = v_r\vec{e}_r + v_\theta\vec{e}_\theta$$

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Vector Notation

$$\vec{v} = \frac{V}{2}\left[3\left(\frac{R}{r}\right) - \left(\frac{R}{r}\right)^3\right]\cos\theta\,\vec{e}_r - \frac{V}{4}\left[3\left(\frac{R}{r}\right) + \left(\frac{R}{r}\right)^3\right]\sin\theta\,\vec{e}_\theta$$

Oseen-Burgers Tensor

Vector Notation

$$
\begin{aligned}
\vec{v} &= \frac{V}{2}\left[3\left(\frac{R}{r}\right) - \left(\frac{R}{r}\right)^3\right]\cos\theta\vec{e}_r - \frac{V}{4}\left[3\left(\frac{R}{r}\right) + \left(\frac{R}{r}\right)^3\right]\sin\theta\vec{e}_\theta \\
&= \frac{3V}{4}\left(\frac{R}{r}\right)\left[2\vec{e}_r\cos\theta - \vec{e}_\theta\sin\theta\right] - \frac{V}{4}\left(\frac{R}{r}\right)^3\left[2\vec{e}_r\cos\theta + \vec{e}_\theta\sin\theta\right] \\
&\approx \frac{3V}{4}\left(\frac{R}{r}\right)\left[2\vec{e}_r\cos\theta - \vec{e}_\theta\sin\theta\right] - \mathcal{O}\left(r^{-3}\right)
\end{aligned}
$$

Oseen-Burgers Tensor

Vector Notation

Since $\vec{e}_z = \vec{e}_r \cos\theta - \vec{e}_\theta \sin\theta$, the velocity to 1$^{\text{st}}$ order is

$$\vec{v} = \frac{3V}{4}\left(\frac{R}{r}\right)\left[2\vec{e}_r \cos\theta - \vec{e}_\theta \sin\theta\right]$$
$$= \frac{3V}{4}\left(\frac{R}{r}\right)\left[\vec{e}_z + \vec{e}_r \cos\theta\right]$$

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$$= \frac{3V}{4}\left(\frac{R}{r}\right)\left[\vec{e}_z + \vec{e}_r \cos\theta\right]$$

That's pretty.

Oseen-Burgers Tensor

Hydrodynamic Interaction

In terms of the drag force, $\vec{F} = 6\pi\eta RV\vec{e}_z$

Oseen-Burgers Tensor

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$$\begin{aligned}
\vec{v} &= \frac{3V}{4}\left(\frac{R}{r}\right)\left[\vec{e}_z + \vec{e}_r \cos\theta\right] \\
&= \frac{3}{4}\frac{\vec{F}}{6\pi\eta R\vec{e}_z}\left(\frac{R}{r}\right)\left[\vec{e}_z + \vec{e}_r \cos\theta\right] \\
&= \frac{1}{8\pi\eta r}\left[\frac{\vec{e}_z}{\vec{e}_z} + \frac{\vec{e}_r}{\vec{e}_z}\cos\theta\right]\vec{F} \\
&= \frac{1}{8\pi\eta r}\left[\hat{I} + \vec{e}_r\vec{e}_r\right]\vec{F}
\end{aligned}$$

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Hydrodynamic Interaction

In terms of the drag force, $\vec{F} = 6\pi\eta RV\vec{e}_z$

$$\vec{v} = \hat{\Omega}\vec{F}$$

where the Oseen-Burgers Tensor

$$\hat{\Omega} = \frac{1}{8\pi\eta r}\left[\hat{I} + \vec{e}_r\vec{e}_r\right]$$

describes the perturbation of fluid due to motion of a sphere.

Oseen-Burgers Tensor

Hydrodynamic Interaction

In terms of the drag force, $\vec{F} = 6\pi\eta R V \vec{e}_z$

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$$\hat{\Omega} = \frac{1}{8\pi\eta r}\left[\hat{I} + \vec{e}_r\vec{e}_r\right]$$

describes the perturbation of fluid due to motion of a sphere. Notice that it decays as r^{-1} with $\mathcal{O}\left(r^{-3}\right)$.

Compliance

Compliance (Often Called Response Function)

$$\vec{r} \equiv \hat{\alpha}\vec{F}$$

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So then:

$$\hat{\alpha} \stackrel{?}{=} \hat{\mu} \times t$$

This may seem silly but it turns out to be most useful.

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We'll come back to this in a moment after we generalize the Stokes Equation.

Extending "Falling Ball" Rheology to Finite Frequencies

Idea

The "falling ball" rheology is very passive and can be thought of as the zero-frequency limit of more active experiments

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$$\eta = \lim_{\omega \to 0} \frac{G''(\omega)}{\omega}$$

G'' is the loss modulus: $G''(\omega) = \omega\eta(\omega)$.

Reconsidering Compliance Tensor

Now the compliance doesn't seem so silly, does it?

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Now the compliance doesn't seem so silly, does it?

$$\alpha = \frac{\hat{\mu}}{\omega} = \frac{1}{\hat{\xi}\omega} = \frac{1}{6\pi\omega\eta(\omega) R} = \frac{1}{6\pi R G''}$$

Complex Compliance

Complication 1) Lose and Storage

Consider the relation between the compliance and the shear modulus for a viscous fluid:

$$\alpha = \frac{1}{6\pi R G''}$$

It stands to reason (and was previously discussed by Dr. Harden) then that for a viscoelastic fluid with $G^*(\omega) = G' + iG''$, the compliance is also complex: $\alpha^*(\omega) = \alpha' + i\alpha''$

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$$\alpha^*(\omega) = \frac{1}{6\pi R G^*(\omega)}$$

History Dependence

Complication 2) Memory

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- Friction Coefficient

$$F(t) = \int_{-\infty}^{t} \xi^{*}(t-\tau) V(\tau) d\tau$$
$$= (\xi^{*} * V)(t)$$

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- Friction Coefficient

$$\begin{aligned} F(t) &= \int_{-\infty}^{t} \xi^{*}(t-\tau)\, V(\tau)\, d\tau \\ &= (\xi^{*} * V)(t) \end{aligned}$$

- Mobility

$$\begin{aligned} V(t) &= \int_{-\infty}^{t} \mu^{*}(t-\tau)\, F(\tau)\, d\tau \\ &= (\mu^{*} * F)(t) \end{aligned}$$

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- Mobility

$$V(t) = \int_{-\infty}^{t} \mu^*(t-\tau) F(\tau)\, d\tau = (\mu^* * F)(t)$$

- Compliance

$$r(t) - r(0) = \int_{-\infty}^{t} \alpha^*(t-\tau) F(\tau)\, d\tau = (\alpha^* * F)(t)$$

Convolution

Because we’ll use it so often . . .

Convolution

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$$(f * g)(t) = \int_{-\infty}^{\infty} f(t-\tau)\, g(\tau)\, d\tau$$

Laplace Transform

Definition

$$\widetilde{f}(s) = \int_0^\infty e^{-st} f(t)\, dt$$

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$$\widetilde{(f * g)}(t) = \widetilde{f}(s)\, \widetilde{g}(s)$$

Linearity

$$f(\widetilde{t) + ig}(t) = \widetilde{f}(s) + i\widetilde{g}(s)$$

Derivative

$$\widetilde{f'(t)} = s\widetilde{f}(s) + f(0)$$

Fourier Transform

Definition

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Linearity

$$\overline{f(t) + ig(t)} = \overline{f}(\omega) + i\overline{g}(\omega)$$

Derivative

$$\overline{f'(t)} = i\omega \overline{f}(\omega)$$

Response Functions in Each Domain

Lag-Time, Fourier and Laplace Domains

- Friction Coefficient

$$F(t) = \left(\xi^{*} * V\right)(t)$$
$$\overline{F}(\omega) = \overline{\xi^{*}}(\omega)\, \overline{V}(\omega)$$
$$\tilde{F}(s) = \tilde{\xi}^{*}(s)\, \tilde{V}(s)$$

- Mobility

$$V(t) = \left(\mu^{*} * F\right)(t)$$
$$\overline{V}(\omega) = \overline{\mu^{*}}(\omega)\, \overline{F}(\omega)$$
$$\tilde{V}(s) = \tilde{\mu}^{*}(s)\, \tilde{F}(s)$$

- Compliance

$$\overline{\Delta r}(\omega) = \left(\alpha^{*} * F\right)(t)$$
$$\overline{\Delta r}(\omega) = \overline{\alpha^{*}}(\omega)\, \overline{F}(\omega)$$
$$\tilde{\Delta r}(s) = \tilde{\alpha}^{*}(s)\, \tilde{F}(s)$$

Response Functions in Each Domain

Lag-Time, Fourier and Laplace Domains

- Friction Coefficient

$$
\begin{aligned}
F(t) &= [\xi^* * V](t) \\
\overline{F}(\omega) &= \overline{\xi^*}(\omega)\,\overline{V}(\omega) \\
F(s) &= \xi^*(s)\,V(s)
\end{aligned}
$$

- Mobility

$$
\begin{aligned}
V(t) &= [\mu^* * F](t) \\
\overline{V}(\omega) &= \overline{\mu^*}(\omega)\,\overline{F}(\omega) \\
V(s) &= \mu^*(s)\,F(s)
\end{aligned}
$$

- Compliance

$$
\begin{aligned}
\overline{\Delta r}(\omega) &= [\alpha^* * F](t) \\
\overline{\Delta r}(\omega) &= \overline{\alpha^*}(\omega)\,\overline{F}(\omega) \\
\Delta r(s) &= \alpha^*(s)\,F(s)
\end{aligned}
$$

Nice

By working in the Laplace (or frequency) domain we recover relationships that look like the Stokes Equation

Generalized Einstein Equation

Langevin Equation

Viscoelastic materials have memory so their random walk can be more complicated:

$$m\dot{V} = F_{\text{rnd}} - \int_0^t \xi\left(t-\tau\right) V\left(\tau\right) d\tau$$

Generalized Einstein Equation

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Need to know entire history in time if you want to know it's current behaviour.

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In the Laplace Domain, the random walk of the Langevin Equation is much simpler.

Generalized Einstein Equation

Langevin Equation in Laplace Domain

$$m\dot{V} = F_{\text{rnd}} - \int_0^t \xi\left(t-\tau\right) V\left(\tau\right) d\tau$$

$$m\widetilde{\dot{V}} = \widetilde{F_{\text{rnd}}} - \widetilde{\left(\xi * V\right)}\left(t\right)$$

$$ms\widetilde{V} - mV\left(0\right) = \widetilde{F_{\text{rnd}}} - \widetilde{\xi}\widetilde{V}$$

$$\widetilde{V}\left(s\right) = \frac{mV\left(0\right) + \widetilde{F_{\text{rnd}}}\left(s\right)}{ms + \widetilde{\xi}\left(s\right)}$$

Generalized Einstein Equation

Langevin Equation in Laplace Domain

Recall, the velocity autocorrelation was the connection to the diffusion coefficient so multiply by $V(0)$ and average:

Generalized Einstein Equation

Langevin Equation in Laplace Domain

Recall, the velocity autocorrelation was the connection to the diffusion coefficient so multiply by $V(0)$ and average:

$$\left\langle V\left(0\right)\widetilde{V}\left(s\right)\right\rangle = \left\langle V\left(0\right)\frac{mV\left(0\right)+\widetilde{F_{\text{rnd}}}\left(s\right)}{ms+\widetilde{\xi}\left(s\right)}\right\rangle$$

Generalized Einstein Equation

Langevin Equation in Laplace Domain

Recall, the velocity autocorrelation was the connection to the diffusion coefficient so multiply by $V(0)$ and average:

$$\left\langle V(0)\,\widetilde{V}(s)\right\rangle = \frac{m\left\langle V^2(0)\right\rangle + \left\langle V(0)\,\widetilde{F_{\text{rnd}}}(s)\right\rangle}{ms + \widetilde{\xi}(s)}$$

Generalized Einstein Equation

Langevin Equation in Laplace Domain

Recall, the velocity autocorrelation was the connection to the diffusion coefficient so multiply by $V(0)$ and average:

$$\left\langle V(0)\,\widetilde{V}(s)\right\rangle = \frac{m\left\langle V^2(0)\right\rangle + \left\langle V(0)\,\widetilde{F_{\text{rnd}}}(s)\right\rangle}{ms + \widetilde{\xi}(s)}$$

1) Average of Random Noise

$$\left\langle V(0)\,\widetilde{F_{\text{rnd}}}(s)\right\rangle = 0$$

Generalized Einstein Equation

Langevin Equation in Laplace Domain

Recall, the velocity autocorrelation was the connection to the diffusion coefficient so multiply by $V(0)$ and average:

$$\left\langle V(0)\,\widetilde{V}(s)\right\rangle = \frac{m\left\langle V^2(0)\right\rangle + 0}{ms + \widetilde{\xi}(s)}$$

Generalized Einstein Equation

Langevin Equation in Laplace Domain

Recall, the velocity autocorrelation was the connection to the diffusion coefficient so multiply by $V(0)$ and average:

$$\left\langle V\left(0\right)\widetilde{V}\left(s\right)\right\rangle = \frac{m\left\langle V^{2}\left(0\right)\right\rangle + 0}{ms + \widetilde{\xi}\left(s\right)}$$

2) Equipartition Theorem

$$\frac{1}{2}m\left\langle V^{2}\left(0\right)\right\rangle = \frac{1}{2}k_{\mathrm{B}}T$$

Generalized Einstein Equation

Langevin Equation in Laplace Domain

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$$\widetilde{\xi} \gg ms$$

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Langevin Equation in Laplace Domain

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Generalized Einstein Equation

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$$\left\langle V(0)\,\widetilde{V}(s)\right\rangle = \frac{k_{\mathrm{B}}T}{\widetilde{\xi}(s)}$$

Replace Viscous with Viscoelastic

We've been doing it all lecture

$$\xi \to \xi^{*}$$

Generalized Einstein Equation

Langevin Equation in Laplace Domain

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$$\left\langle V\left(0\right)\widetilde{V}\left(s\right)\right\rangle = \frac{k_{\mathrm{B}}T}{\widetilde{\xi}^{*}\left(s\right)}$$

Generalized Einstein Equation

Identity

In Laplace Domain, it is true that the velocity autocorrelation and the mean square displacement are equivalent by the identy:

$$\left\langle V(0)\,\widetilde{V}(s)\right\rangle \equiv \frac{s^2}{2}\left\langle \Delta\widetilde{r}^2(s)\right\rangle$$

Notice, I wrote $\left\langle \Delta\widetilde{r}^2(s)\right\rangle \equiv \widetilde{\left\langle \Delta r^2\right\rangle}(s)$ just because it's prettier that way.

Generalized Einstein Equation

Hard Earned Results

$$\widetilde{\xi}^{*}(s) = \frac{2k_{\mathrm{B}}T}{s^{2}\left\langle \Delta\widetilde{r}^{2}(s)\right\rangle}$$

Generalized Einstein Equation

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$$\widetilde{\xi^*}(s) = \frac{2k_B T}{s^2 \left\langle \Delta \widetilde{r}^2(s) \right\rangle}$$

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$$\widetilde{G^*}(s) = \frac{k_B T}{\pi R s \left\langle \Delta \widetilde{r}^2(s) \right\rangle}$$

General Treatment

General Response Function

Last time David said:

General Treatment

General Response Function

Last time David said:

$$\mathcal{R}(t) = \frac{1}{k_{\mathrm{B}}T}\frac{\partial C_0}{\partial t}$$

where $\mathcal{R}$ is a response function (say α) and C_0 is the autocorrelation function (say $\left\langle \Delta q^2(t) \right\rangle$).

General Treatment

General Response Function

$$\mathcal{R}(t) = \frac{1}{k_{\mathrm{B}}T}\frac{\partial C_0}{\partial t}$$

General Laplace

$$\begin{aligned}\widetilde{\mathcal{R}} &= \frac{1}{k_{\mathrm{B}}T}\widetilde{\frac{\partial C_0}{\partial t}} \\ &= \frac{s}{k_{\mathrm{B}}T}\widetilde{C_0} \\ &= \frac{s}{k_{\mathrm{B}}T}\langle \widetilde{\Delta q^2(s)}\rangle \\ &= \frac{s}{k_{\mathrm{B}}T}\left\langle \Delta\widetilde{q}^2(s)\right\rangle\end{aligned}$$

General Treatment

General Response Function

$$\mathcal{R}(t) = \frac{1}{k_B T} \frac{\partial C_0}{\partial t}$$

General Fourier

$$\begin{aligned} \overline{\mathcal{R}} &= \frac{1}{k_B T} \overline{\frac{\partial C_0}{\partial t}} \\ &= \frac{i\omega}{k_B T} \overline{C_0} \end{aligned}$$

Fourier Transform of Autocorrelation

The Fourier transform of a cross correlation is

$$q \star p = \int_{-\infty}^{\infty} q^{CC}(\tau)\, p(t+\tau)\, d\tau$$
$$= \left(q^{cc}(-t) * p(t)\right)(t)$$
$$\overline{(q \star p)} = \overline{q}^{CC}\overline{p}$$

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Therefore, for autocorrelation we have

$$\begin{aligned} \overline{(q \star q)} &= \overline{q}^{CC}\overline{q} \\ &= |\overline{q}|^2 \end{aligned}$$

Return to response function ...

General Treatment

General Response Function

$$\mathcal{R}(t) = \frac{1}{k_B T}\frac{\partial C_0}{\partial t}$$

General Fourier

$$\begin{aligned}\overline{\mathcal{R}} &= \frac{1}{k_B T}\overline{\frac{\partial C_0}{\partial t}} \\ &= \frac{i\omega}{k_B T}\overline{C_0} \\ &= \frac{i\omega}{k_B T}\left|\overline{q}\right|^2\end{aligned}$$

Note: *be aware of upcoming statement about this solution.*

In the laboratory

Fourier Domain

We found the complex, viscoelastic response functions in Laplace space **but**

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$$\overline{\alpha''}(\omega) = \frac{\omega \left| \Delta \bar{r}^2 \right|}{2 k_\mathrm{B} T}$$

Important point:

In the laboratory

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Important point: only get loss compliance

In the laboratory

Kramers-Kronig

For any complex function, $f = f + ig$, there is an identity

$$g = -\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{f(w)}{w-\omega}dw$$

i.e. f and g are not independant.

In the laboratory

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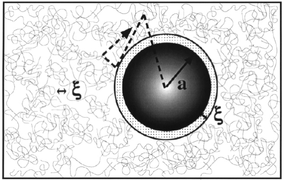
$$g = -\frac{1}{\pi}\mathcal{P}\int_{-\infty}^{\infty}\frac{f(w)}{w-\omega}dw$$

i.e. f and g are not independant.

Our case:

$$\begin{aligned}\alpha'(\omega) &= \frac{2}{\pi}\mathcal{P}\int_0^{\infty}\frac{w\alpha''(w)}{w^2-\omega^2}dw \\ &= \frac{2}{\pi}\int_0^{\infty}\cos(\omega t)\,dt\int_0^{\infty}\alpha''(w)\sin(wt)\,dw\end{aligned}$$

Local Medium



Local Medium

Probe particle is really existing in a bubble. Do the response functions see outside of the local region?

> *We imagine that each probe sphere is surrounded by a pocket of perturbed material with rheological properties diferent from those of the bulk.*

Local Medium

Homogeneous Media

For a homogeneous media, we went to great lengths to demonstrate

$$\hat{\Omega} \propto \frac{R}{r}$$

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Homogeneous Media

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Pocket Model

The hydrodynamic interaction still extends with the same scaling **but** it must displace some viscoelastic matrix at the interface between the bulk and the pocket.

Example

Levine and Lubensky

Assume that the media responds elastically to the perturbations due to the probe motion in viscous fluid:

$$0 = \lambda_1 \nabla^2 \vec{u} + (\lambda_1 + \lambda_2) \vec{\nabla} \vec{\nabla} \cdot \vec{u}$$

where $\vec{u}$ is the displacement of the media and λ_i are Lamè coefficients (for describing elasticity).

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Find:

$$\hat{\alpha} = \frac{\hat{I}}{6\pi\eta R} Z\left(\lambda_{1,\text{in}}, \lambda_{2,\text{in}}, \xi/R\right)$$

Example

Compliance Only Depends on Pocket Properties

The correction factor $Z\left(\lambda_{1,\text{in}}, \lambda_{2,\text{in}}, \xi/R\right)$ fails to capture storage information about bulk.

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Compliance Only Depends on Pocket Properties

The correction factor $Z\left(\lambda_{1,\text{in}}, \lambda_{2,\text{in}}, \xi/R\right)$ fails to capture storage information about bulk. FYI: **if** bulk can be treated as incompressible then Z takes the **relatively simple** form

$$Z = \frac{4\beta^6\kappa'^2 + 10\beta^3\kappa' - 9\beta^5\kappa'\kappa + 2\kappa\kappa'' + 3\beta\left[2 + \kappa - 3\kappa^2\right]}{2\left[\kappa'' - 2\beta^5\kappa'\right]}$$

where $\beta = 1 + \xi/R$, $\kappa = G^*_{\text{out}}/G^*_{\text{in}}$, $\kappa' = \kappa - 1$ and $\kappa'' = 3 + 2\kappa$.

Langevin Equation

1-Particle

$$m\dot{\vec{V}} = \vec{F}_{\text{rnd}} - \int_0^t \hat{\xi}^* (t-\tau) \vec{V}(\tau) \, d\tau$$

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2-Particles

$$m\dot{\vec{V}}_i = \vec{F}_{\text{rnd}} - \int_0^t \hat{\xi}^*_{ii} \left(t-\tau\right) \vec{V}_i\left(\tau\right) d\tau - \vec{F}_{ij}$$

Langevin Equation

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2-Particles

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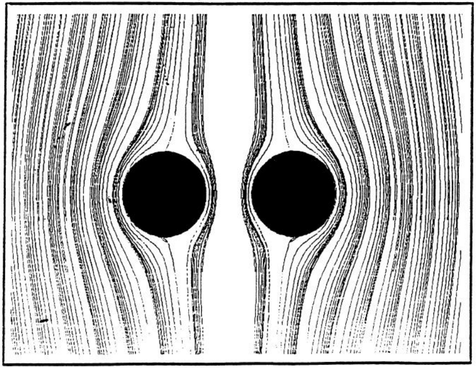
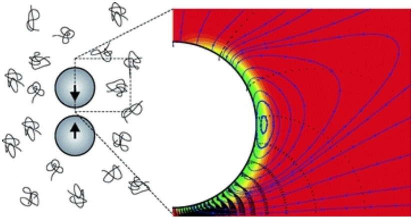
Force on particle i due to particle j - Viscous Fluid



Force on particle i due to particle j - Viscoelastic Fluid



Force on particle *i* due to particle *j*

Force from Second Particle

The second particle (j) acts on the first (i) because it's moving through the viscoelastic fluid.

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The second particle (j) acts on the first (i) because it's moving through the viscoelastic fluid. Sphere j has some velocity V_j which causes a drag

$$\vec{F}_j = \int_0^t \hat{\xi}_j^* \vec{V}_j dt$$

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Resulting in the force $\vec{F}_{ij}$ on particle i:

$$\begin{aligned}
\vec{F}_{ij} &= \hat{\Omega}^{-1}\vec{v} \\
&= \hat{\Omega}^{-1}\hat{\Omega}\vec{F}_j \\
&= \vec{F}_j \\
&= \int_0^t \hat{\xi}_j^* \vec{V}_j dt
\end{aligned}$$

Laplace Transform

Two-Particle Langevin Equation

$$
\begin{aligned}
m\dot{V}_i &= F_{\text{rnd}} - \int_0^t \xi_i^* \left(t-\tau\right) V_i\left(\tau\right) d\tau - F_{ij} \\
&= F_{\text{rnd}} - \int_0^t \xi_i^* \left(t-\tau\right) V_i\left(\tau\right) d\tau - \int_0^t \xi_j^* \left(t-\tau\right) V_j\left(\tau\right) d\tau \\
&= F_{\text{rnd}} - \int_0^t \xi_{ii}^* \left(t-\tau\right) V_i\left(\tau\right) d\tau - \int_0^t \xi_{ij}^* \left(t-\tau\right) V_j\left(\tau\right) d\tau \\
&= F_{\text{rnd}} - \left(\xi_{ii}^* * V_i\right)(t) - \left(\xi_{ij}^* * V_j\right)(t)
\end{aligned}
$$

Note: I turned the set friction tensors into an array of tensors.

Laplace Transform

Two-Particle Langevin Equation

$$ms\widetilde{V}_i - mV_i(0) = \widetilde{F_{\text{rnd}}} - \widetilde{\xi}^*_{ij}\widetilde{V}_j - \widetilde{\xi}^*_{ii}\widetilde{V}_i$$

Laplace Transform

Two-Particle Langevin Equation

$$ms\widetilde{V}_i - mV_i\left(0\right) = \widetilde{F_{\text{rnd}}} - \widetilde{\xi_{ij}^*}\widetilde{V}_j - \widetilde{\xi_{ii}^*}\widetilde{V}_i$$

We want to consider the distinct's interparticle part so on top of multiplying by $V_i(0)$ again, we also multiply by $\delta(R - r_{ij}) = \delta_{ij}$ before averaging

$$ms\widetilde{V}_i V_i\left(0\right)\delta_{ij} - mV_i^2\left(0\right)\delta_{ij} = \widetilde{F_{\text{rnd}}}V_i\left(0\right)\delta_{ij} - \widetilde{\xi_{ij}^*}\widetilde{V}_j V_i\left(0\right)\delta_{ij} - \widetilde{\xi_{ii}^*}\widetilde{V}_i V_i\left(0\right)\delta_{ij}$$

$$ms\left\langle \widetilde{V}_i V_i\left(0\right)\delta_{ij}\right\rangle - m\left\langle V_i^2\left(0\right)\delta_{ij}\right\rangle = \left\langle \widetilde{F_{\text{rnd}}}V_i\left(0\right)\delta_{ij}\right\rangle - \widetilde{\xi_{ij}^*}\left\langle \widetilde{V}_j V_i\left(0\right)\delta_{ij}\right\rangle - \widetilde{\xi_{ii}^*}\left\langle \widetilde{V}_i V_i\left(0\right)\delta_{ij}\right\rangle$$

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$$0 - k_B T = 0 - \widetilde{\xi}^*_{ij}\left\langle V_i(0)\,\widetilde{V}_j\delta_{ij}\right\rangle - 0$$

Response Functions

Correlation and Response Functions

$$\left\langle V_i(0)\,\widetilde{V}_j\delta_{ij}\right\rangle = \frac{k_B T}{\widetilde{\xi}^*_{ij}}$$

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Say $\hat{D} = \langle \Delta r_i(0)\,\Delta\widetilde{r}_j(s)\,\delta_{ij}\rangle$ is the **distinct** displacement tensor, (or the mobility correlation tensor)

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Say $\hat{D} = \langle \Delta r_i(0)\,\Delta\widetilde{r}_j(s)\,\delta_{ij}\rangle$ is the **distinct** displacement tensor, (or the mobility correlation tensor)

$$\begin{aligned} D_{ij} &= \frac{2k_\mathrm{B}T}{s^2\widetilde{\xi}^*_{ij}} \\ &= \frac{2k_\mathrm{B}T}{s^2}\widetilde{\mu}^*_{ij} \\ &= \frac{3k_\mathrm{B}T}{s}\widetilde{\alpha}^*_{ij} \end{aligned}$$

Complex Modulus

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But what about the complex modulus, $\widetilde{G}^* = 6\pi R\widetilde{\alpha}_{ij}$?

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$$D_{ij} = \frac{k_\mathrm{B}T}{2\pi sR\widetilde{G}^*_{ij}}$$

can not be permissible.

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Along the line connecting the particles the mobility correlation function is

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Non-Radial Components

$$D_{\theta\theta} = D(\phi\phi) = \frac{1}{2} D_{rr}$$

Comparison of Near and Far

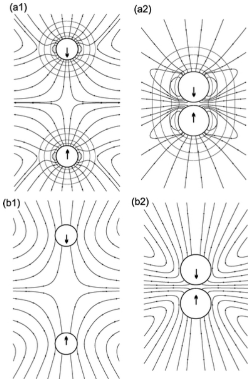
Far Field

The two particle method demands that the particles are far from each other.

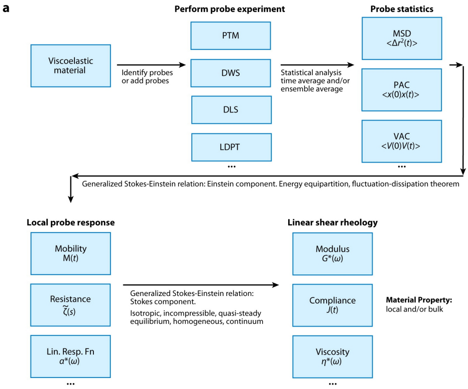
Comparison of Near and Far

Far Field

The two particle method demands that the particles are far from each other. If *local* environments overlap then measurements obviously won't represent the bulk properties.



Conclusion


a
Perform probe experiment
Probe statistics
Viscoelastic material
Identify probes or add probes
PTM
DWS
DLS
LDPT
...
Statistical analysis time average and/or ensemble average
MSD <Δr²(t)>
PAC <x(0)x(t)>
VAC <V(0)V(t)>
...
Generalized Stokes-Einstein relation: Einstein component. Energy equipartition, fluctuation-dissipation theorem
Local probe response
Mobility M(t)
Resistance ζ̃(s)
Lin. Resp. Fn α*(ω)
...
Generalized Stokes-Einstein relation: Stokes component.
Isotropic, incompressible, quasi-steady equilibrium, homogeneous, continuum
Linear shear rheology
Modulus G*(ω)
Compliance J(t)
Viscosity η*(ω)
...
Material Property: local and/or bulk

Thank you for your patience.