Fluctuating Hydrodynamics Approaches for Mesoscopic Modeling and Simulation Applications in Soft Materials and Fluidics

Theoretical Background and Applications

Summer School on Multiscale Modeling of Materials
Stanford University
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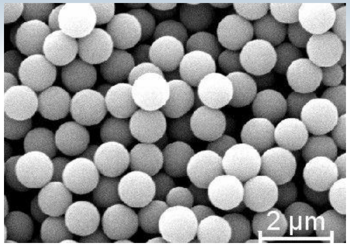
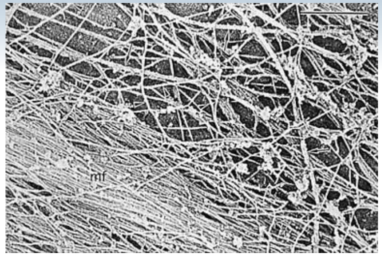
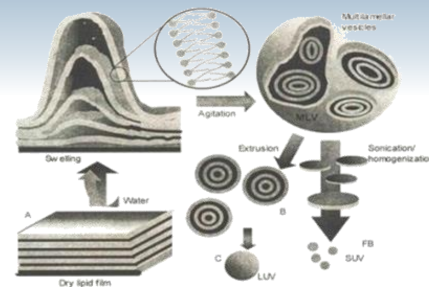
Soft Materials and Fluidics Simulations



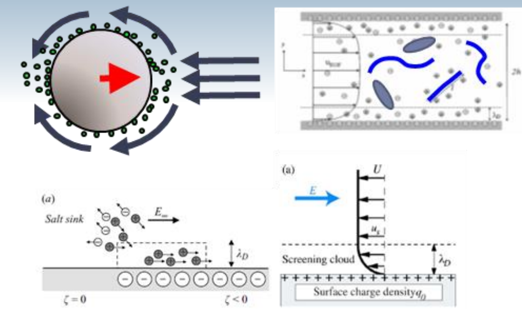
Colloids



Gels (Actin)



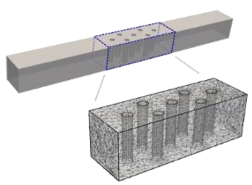
Membranes (lipids)



Fluidics

Soft Materials and Fluidics

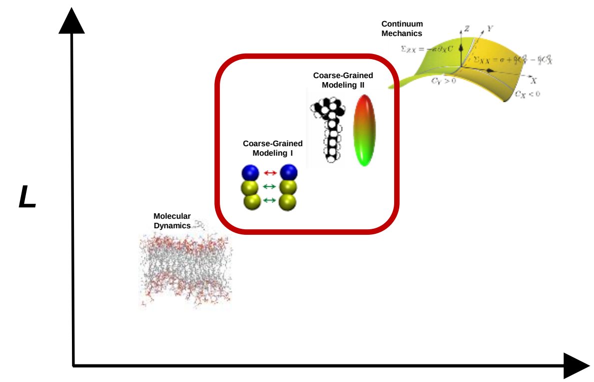
- Interactions on order of K_BT.
- Properties arise from balance of entropy-enthalpy.
- Solvent plays important role (interactions / responses).
- Phenomena span wide temporal-spatial scales.



Geometry / Confinement

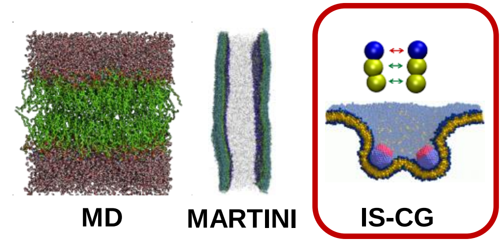
Approaches

- Atomistic Molecular Dynamics.
- Continuum Mechanics.
- Coarse-Grained Particle Models (solvated / implicit).



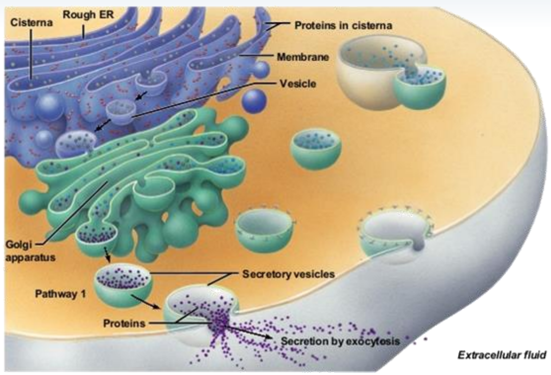
Simulation Methods / Thermostats

- NVE vs NVT ensembles.
- NVE → Velocity-Verlet (no thermostat).
- NVT → Berendsen, Nose-Hoover (artificial dynamics).
- NVT→ Langevin Dynamics (kinetics?).
- What about solvent mediated kinetics? What about other ensembles (NPT, $\dot{\gamma}$VT)?

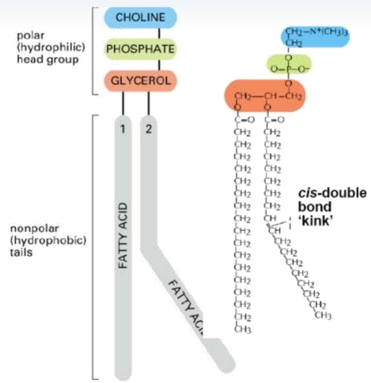


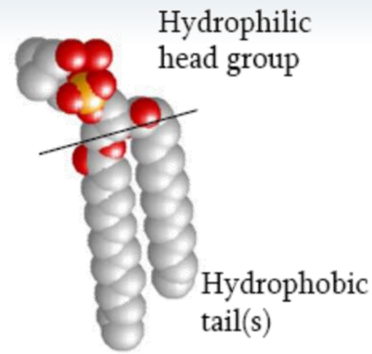
Lipid Bilayer Membranes : Amphiphilic Molecules

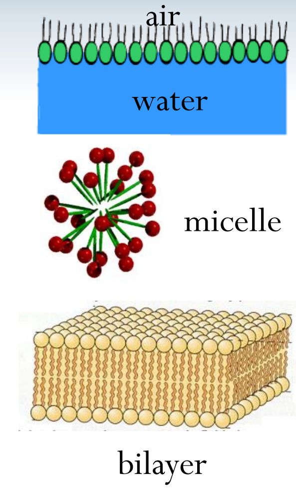
Cell Membranes



Phosphogylceride







Lipid Bilayer Membranes

- Cellular biology : membranes compartmentalize cell, dynamic structures, diverse functions.
- Fluid phase two layered structure (bilayer).
- Mechanics of a fluid-elastic sheet (in-plane flow, elastic response to bending).
- Phenomena span wide temporal-spatial scales.

Amphiphilic Molecules (Lipids)

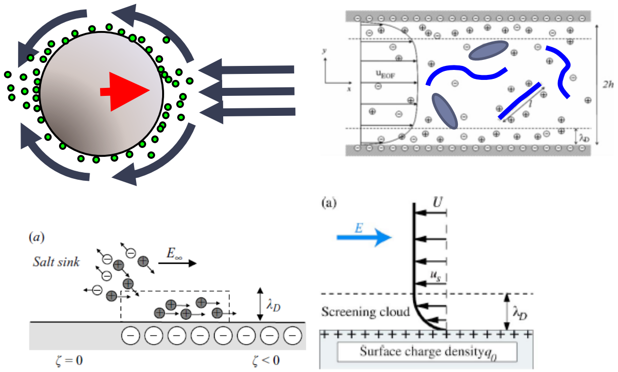
- Amphiphiles have a polar head (hydrophilic) and non-polar tail (hydrophobic).
- Solvent plays key role driving self-assembly (hydrophobic-hydrophilic effect).
- Phases fluid vs gel, micelle vs lamellar, size of polar vs non-polar part.

Partially Ordered Structures

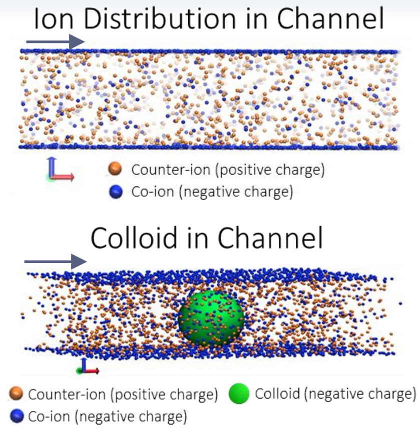
- Lyotropic liquid crystals (temperature and concentration determines phase).
- Smectic A and C phases (translational order in layers, orientation orthogonal/tilt in layer).
- Lamellar sheets most relevant to biology, but many other phases possible.

Fluidics Transport

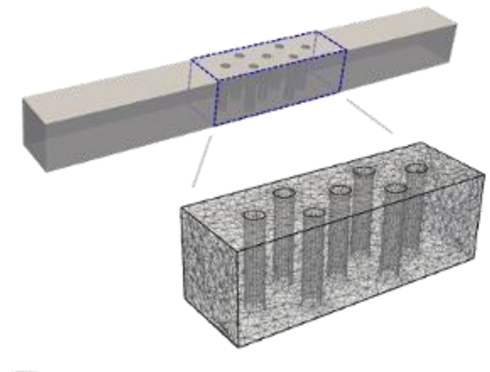
Fluidic Devices



Electrokinetics



Geometry / Confinement



Fluidic Devices

- Developed to miniaturize and automate many laboratory tests, diagnostics, characterization.
- Hydrodynamic transport at such scales must grapple with dissipation / friction.
- Electrokinetic effects utilized to drive flow.

Key Features

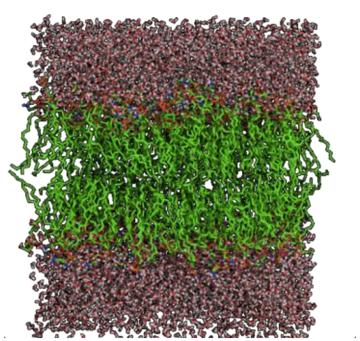
- Large surface area to volume.
- Ionic double-layers can be comparable to channel width.
- Brownian motion plays important role in ion distribution and analyte diffusion across channel.
- Hydrodynamic flow effected by close proximity to walls or other geometric features.
- Ionic concentrations often in regime with significant discrete correlations /density fluctuations.

Challenges

- Develop theory and methods beyond mean-field Poisson-Boltzmann theory.
- Methods capable of handling hydrodynamics, fluctuations, geometry/confinement.

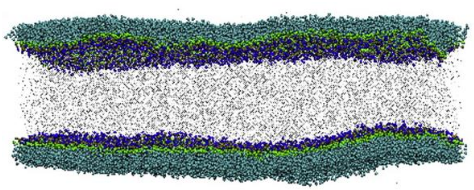
Modeling Approaches for Lipid Bilayer Membranes

Atomistic Molecular Dynamics



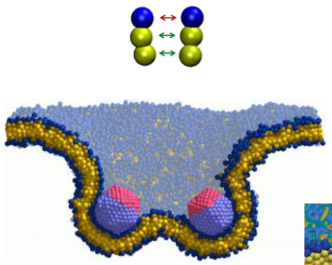
Grossfield 2013

Coarse-Grained **Explicit-Solvent**

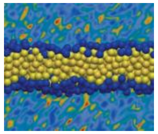


Duncan 2012

Coarse-Grained **Implicit-Solvent**

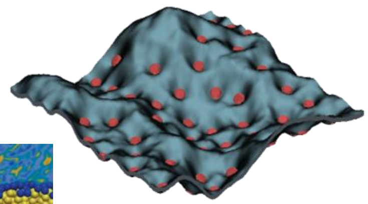


Deserno 2007



Atzberger 2013

Continuum Mechanics Hydrodynamics



Atzberger et al. 2009
Atzberger & Sigurdsson 2013

Atomistic Molecular Dynamics

- Representation of solvent fluid molecules and lipids.
- Atomic detail of molecules.
- Limited length and time-scales.

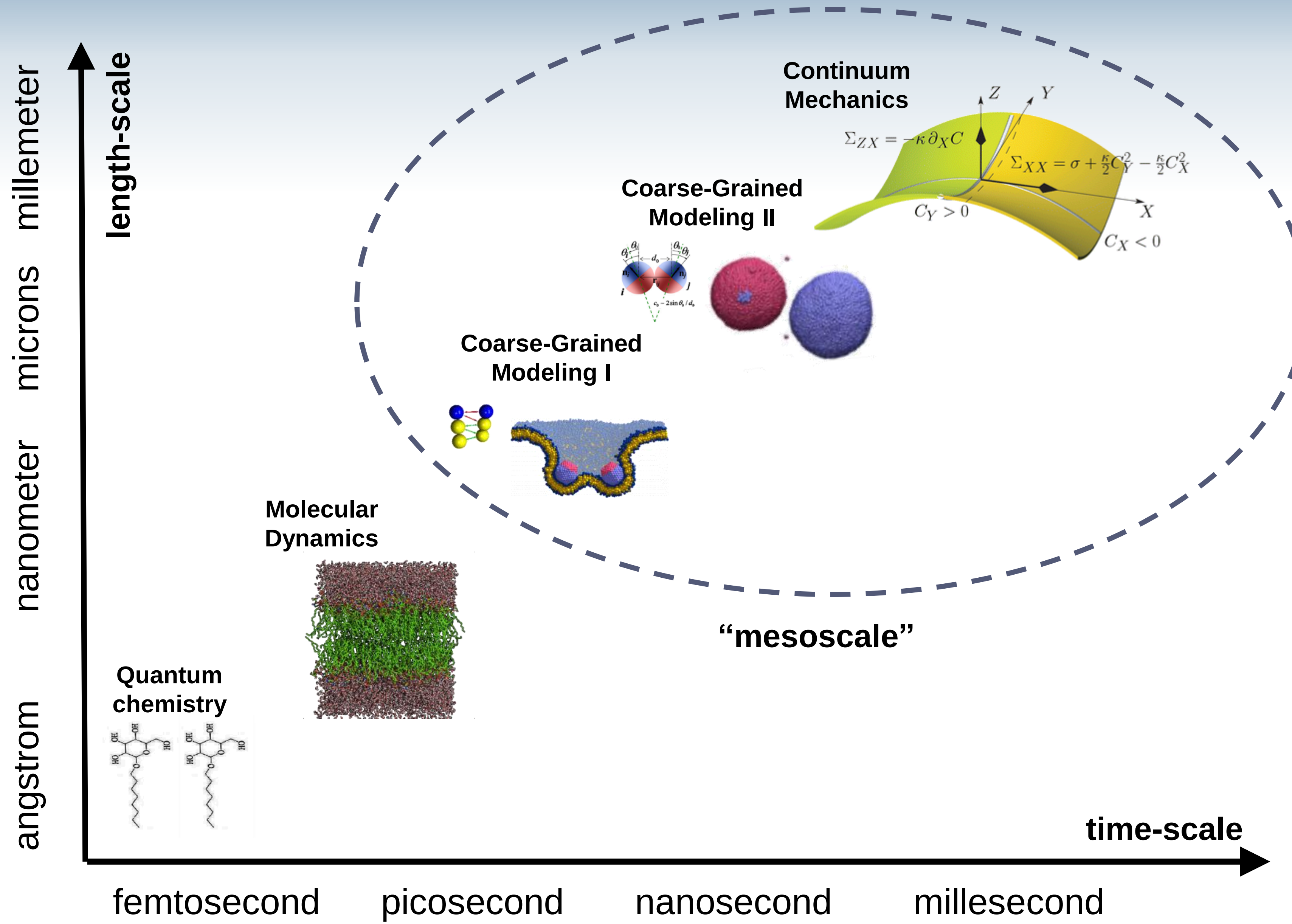
Explicit-Solvent Coarse-Grained (ES-CG)

- Atoms grouped/represented by coarse-grained units.
- Effective free-energy of interaction used on remaining degrees of freedom (DOF).
- Reduces entropy of the system (caution).
- Smooths energy landscape with often less stiff dynamics.
- Explicit-solvent is expensive, still requires resolving molecules of the bulk.

Implicit-Solvent Coarse-Grained (IS-CG)

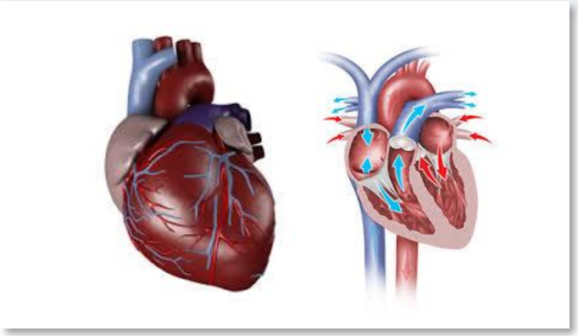
- Atoms grouped/represented by coarse-grained units.
- Effective free-energy of interaction used on remaining degrees of freedom (DOF).
- Used widely for equilibrium studies, however, dynamics augmented by missing solvent effects.
- To extend for kinetic studies, need thermostats to account for correlation contributions of solvent in IS-CG.

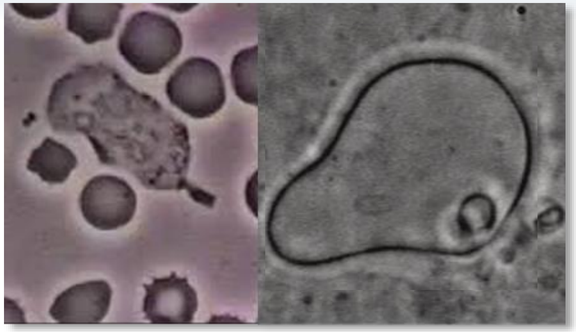
Model Resolution



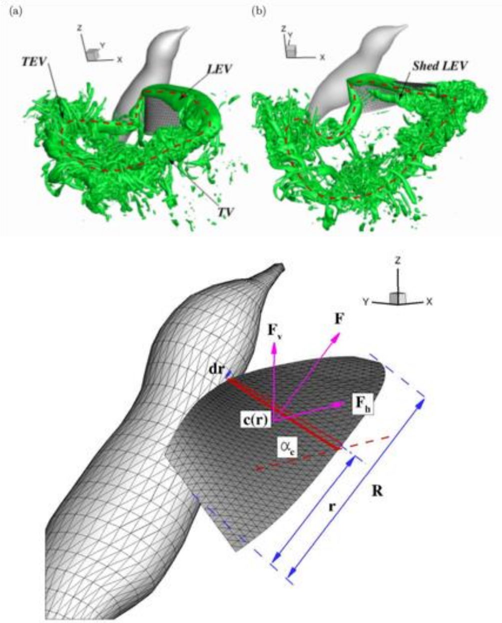
Solvent Hydrodynamics
CFD Fluid-Structure Interactions

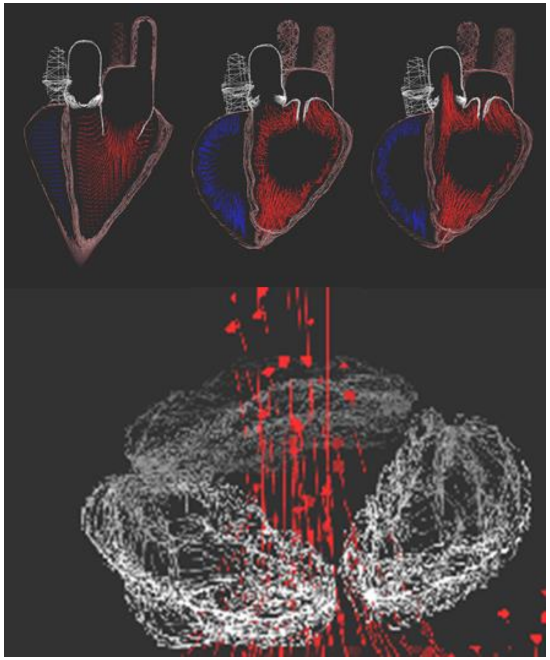
Fluid-Structure Interactions

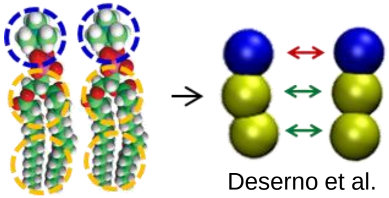




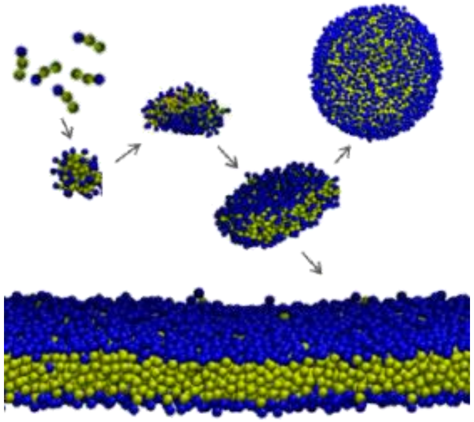
David Rogers







Deserno et al.

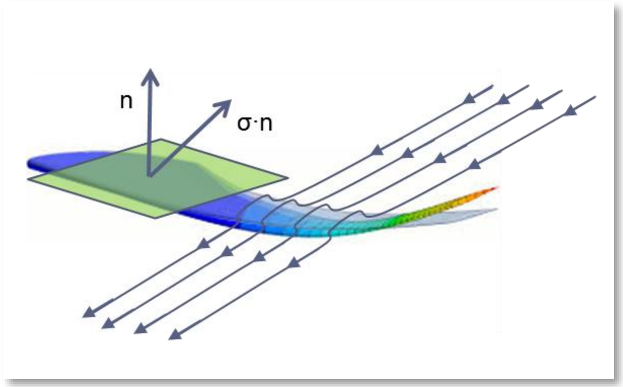


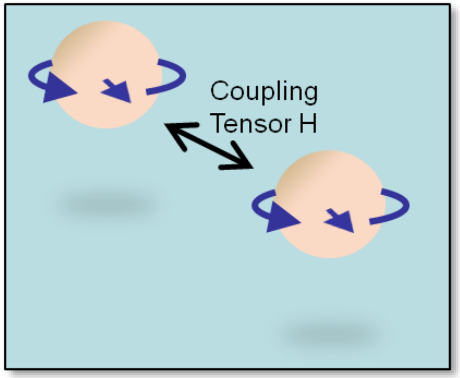
Song, J., Luo, H., Hedrick, T.L.

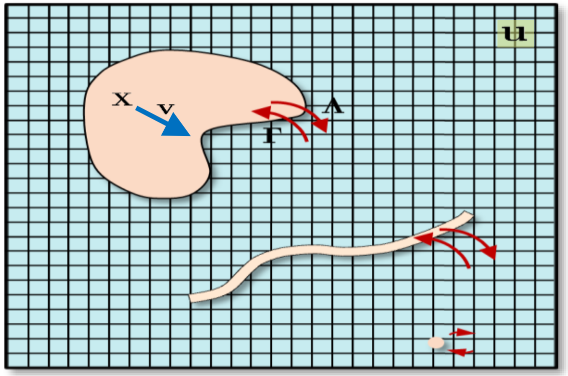
Peskin,C and McQueen, D. et al.

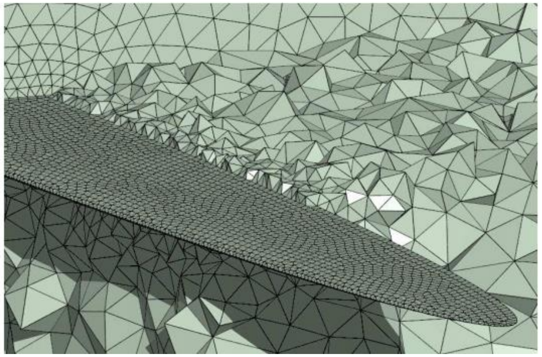
Atzberger, P.,Sigurdsson, J. et al.

CFD : Approaches



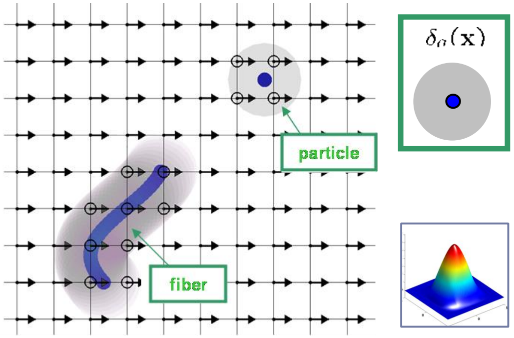






J. Peraire and P.-O. Persson

Brady et al., G. Gompper et al.



Atzberger, Peskin, Kramer

Thermostats

Berendson, Nose-Hoover

particle momentum

particle momentum

heat bath

Langevin

particle momentum

X (sink)

thermal fluctuations (stochastic)

$$
\begin{aligned}
m\frac{d\mathbf{V}}{dt} &= -\gamma\mathbf{V} - \nabla\Phi(\mathbf{X}) + \sqrt{2k_BT\gamma}\frac{d\mathbf{B}_t}{dt} \\
\frac{d\mathbf{X}}{dt} &= \mathbf{V}.
\end{aligned}
$$

missing correlations through solvent!

Fluctuating Hydrodynamics

particle momentum

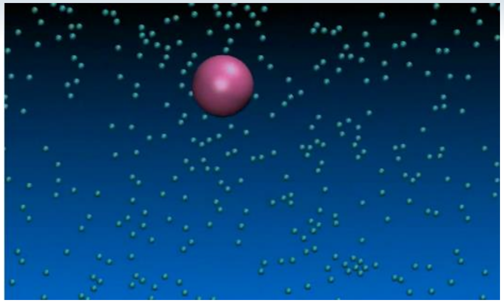
solvent (fluid modes)

solvent (fluid fluctuations)

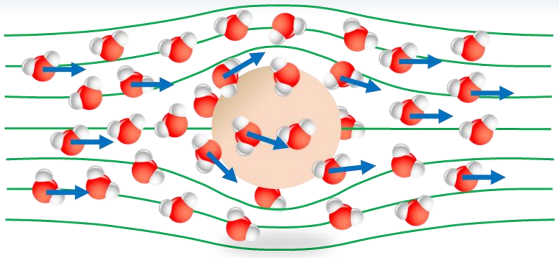
$$
\begin{aligned}
\frac{d\mathbf{X}}{dt} &= \mathbf{v} \\
m\frac{d\mathbf{v}}{dt} &= -\Upsilon\left(\mathbf{v} - \Gamma\mathbf{u}\right) - \nabla_{\mathbf{X}}\Phi[\mathbf{X}] + \zeta + \mathbf{F}_{\mathrm{thm}}
\end{aligned}
$$

lateral momentum transfer : correlations

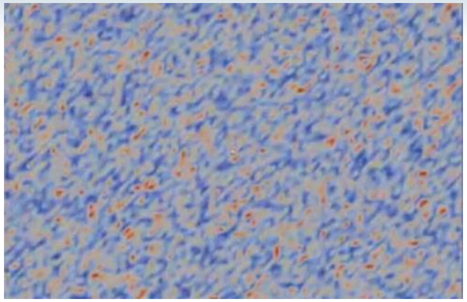
Fluctuating Hydrodynamics



Brownian Motion: Molecular Collisions



Hydrodynamics + Fluctuations



Continuum Gaussian Random Field

Landau-Lifschitz fluctuating hydrodynamics

$$\rho\left(\frac{\partial \mathbf{u}(\mathbf{x},t)}{\partial t} + \mathbf{u}(\mathbf{x},t)\cdot\nabla\mathbf{u}(\mathbf{x},t)\right) = \mu\Delta\mathbf{u}(\mathbf{x},t) - \nabla p(\mathbf{x},t) + \nabla\cdot\mathbf{\Sigma}(\mathbf{x},t).$$

$$\nabla\cdot\mathbf{u}(\mathbf{x},t) = 0.$$

$$\langle \Sigma_{ij}(\mathbf{x},t)\Sigma_{kl}(\mathbf{y},s)\rangle = 2\mu k_B T\left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}\right)\delta(\mathbf{x}-\mathbf{y})\delta(t-s).$$

- Spontaneous momentum transfer from molecular-level interactions.
- Thermal fluctuations captured through random stress Σ.
- Mathematically, equations present challenges since δ-correlation in space-time.
- Fluid-structure interactions?

Immersed Boundary Method

Fluid dynamics

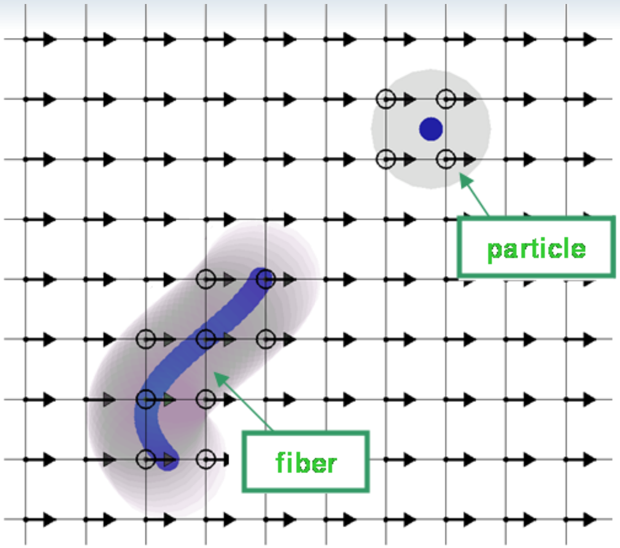
$$\rho \frac{D\mathbf{u}(\mathbf{x},t)}{Dt} = \mu \Delta \mathbf{u}(\mathbf{x},t) - \nabla p(\mathbf{x},t) + \mathbf{F}_{\mathrm{prt}}(\mathbf{x},t) + \mathbf{F}_{\mathrm{thm}}(\mathbf{x},t).$$

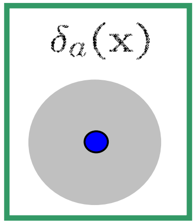
$$\nabla \cdot \mathbf{u}(\mathbf{x},t) = 0.$$

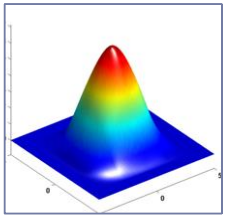
Structure dynamics

$$\frac{d\mathbf{X}^{[j]}(t)}{dt} = \int \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))\mathbf{u}(\mathbf{x},t)d\mathbf{x}$$

$$\mathbf{F}_{\mathrm{ptr}}(\mathbf{x},t) = \sum_{j=1}^{M} \mathbf{F}^{[j]} \delta_a\left(\mathbf{x} - \mathbf{X}^{[j]}(t)\right)$$

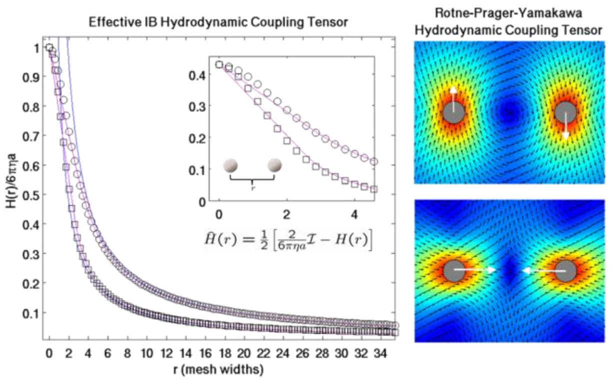






Features:

- Allows conventional discretizations for fluid domain (FV, FFTs).
- Particles, fibers, membranes, and bodies possible.
- Thermal fluctuations: F_{thm} = ?



Atzberger, Peskin, Kramer 2007

Stochastic Immersed Boundary Method

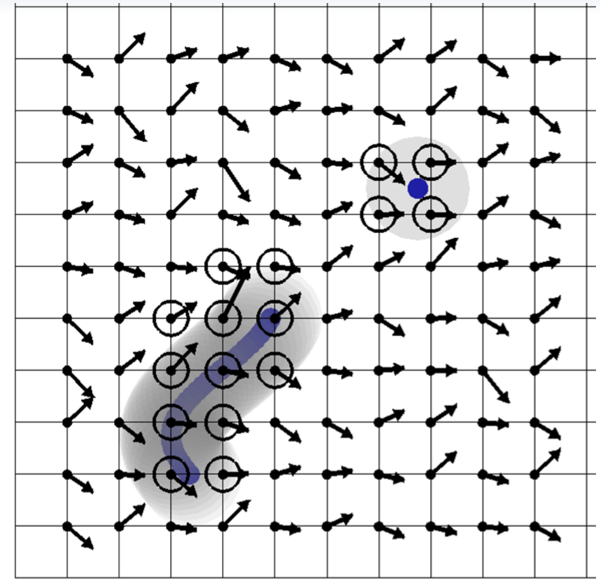
Fluid-structure equations

$$\rho \frac{D\mathbf{u}(\mathbf{x},t)}{Dt} = \mu \Delta \mathbf{u}(\mathbf{x},t) - \nabla p(\mathbf{x},t) + \mathbf{F}_{\mathrm{prt}}(\mathbf{x},t) + \mathbf{F}_{\mathrm{thm}}(\mathbf{x},t).$$

$$\nabla \cdot \mathbf{u}(\mathbf{x},t) = 0.$$

$$\frac{d\mathbf{X}^{[j]}(t)}{dt} = \int \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))\mathbf{u}(\mathbf{x},t)d\mathbf{x}$$

$$\mathbf{F}_{\mathrm{ptr}}(\mathbf{x},t) = \sum_{j=1}^{M} \mathbf{F}^{[j]}\delta_a\left(\mathbf{x} - \mathbf{X}^{[j]}(t)\right)$$

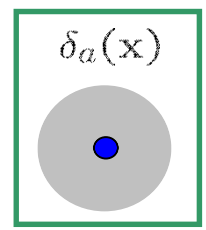


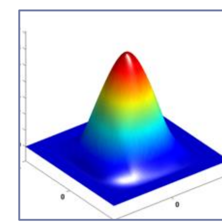
Thermal fluctuations

$$\mathbf{F}_{\mathrm{thm}}(\mathbf{x},t) = \mathbf{F}_{\mathrm{drift}}(\mathbf{x},t) + \mathbf{F}_{\mathrm{stoch}}(\mathbf{x},t)$$

$$\mathbf{F}_{\mathrm{drift}} = -k_B T \sum_{j=1}^{M} \nabla_{\mathbf{X}^{[j]}} \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))$$

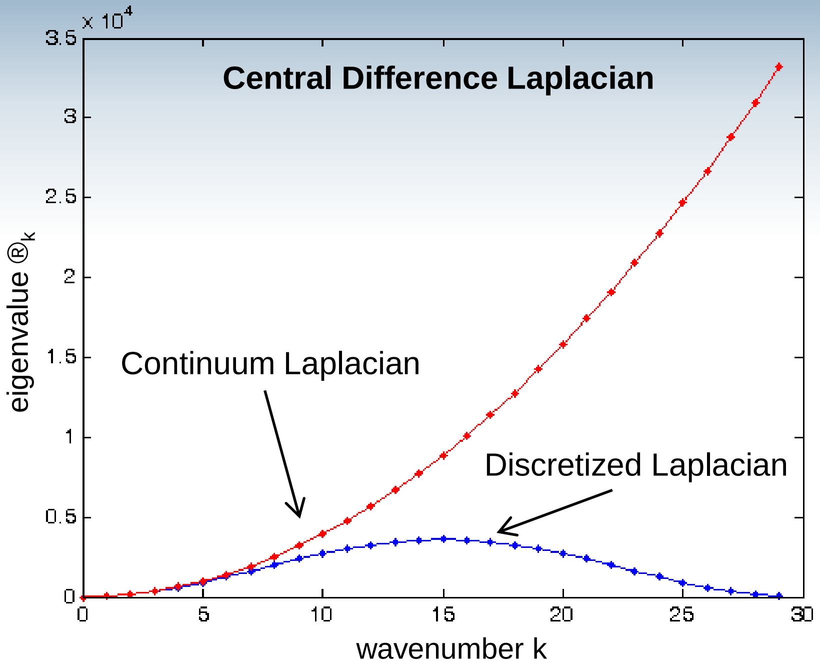
$$\left\langle \mathbf{F}_{\mathrm{stoch}}(\mathbf{x},t)\mathbf{F}^T_{\mathrm{stoch}}(\mathbf{y},s)\right\rangle = -2k_B T \mu \Delta \delta(\mathbf{x}-\mathbf{y})\delta(t-s)$$

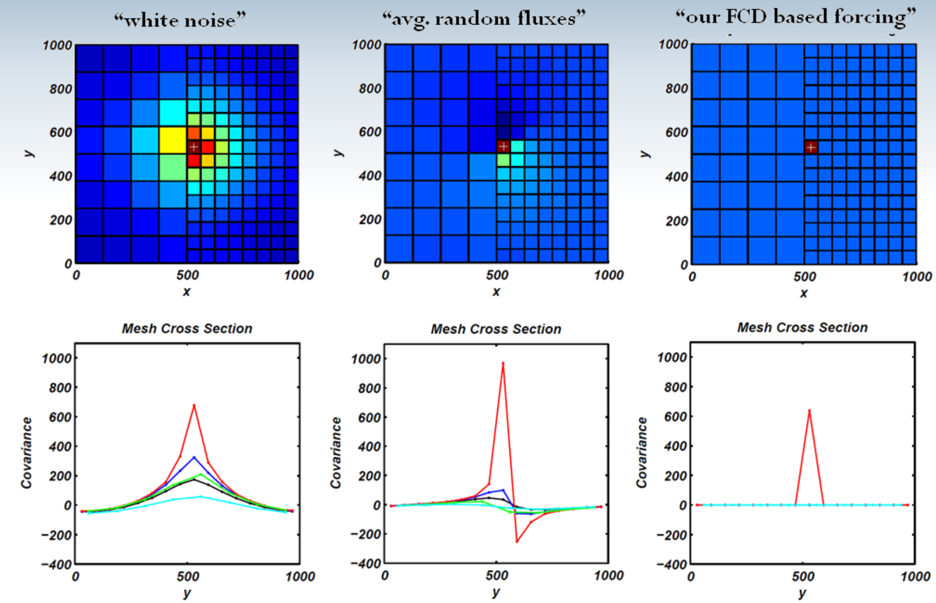




Atzberger, Peskin, Kramer 2007

Numerical Approximation





Dissipation rates are different for continuum and discrete system

- Must approximate differently thermal fluctuations in design of numerical methods.

- Mathematical formulation (Atzberger, Kramer, Peskin 2007, Atzberger 2011):
  - Fluctuation-dissipation balance (ito calculus, nyquist relations).
  - Invariance of Gibbs-Boltzmann (kolomogorov pde's, detailed balance)

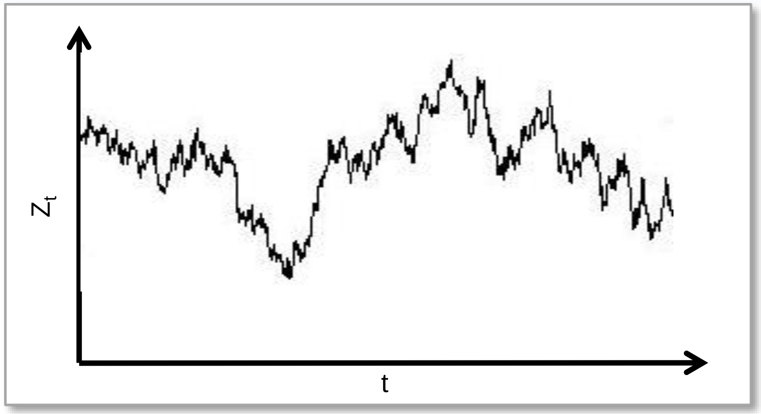
Fluctuation-dissipation balance condition

$$\langle \mathbf{f}_{\mathrm{thm}}(s)\mathbf{f}_{\mathrm{thm}}^{T}(t)\rangle = -\left(LC + CL^{T}\right)\delta(t-s)$$

$$L\mathbf{u} \leftarrow \mu\Delta\mathbf{u} \qquad C_{ij} \leftarrow \frac{k_B T}{\rho\Delta x^3}\delta_{ij}$$

Numerical Stiffness

Time-scales	
Fluid Modes	Particle Diffusion
$\tau_\lambda = \frac{\rho}{4\pi^2\mu}\lambda^2$	$\tau_{\text{diff}}(a) \approx \frac{a^2}{D_a}$
λ = 10nm : $\tau = 10^{-3}$ns	$\tau_{\text{diff}}(1\text{nm}) \approx 10^0\text{ns}$
λ = 1000nm : τ = 10ns	$\tau_{\text{diff}}(10\text{nm}) \approx 10^3\text{ns}$



Sources of stiffness

- Fluid-structure have stochastic trajectories.
- Thermal fluctuations excite all fluid modes.
- Length-scales of microstructure involve fluid dynamics at small Re << 1.
- Equilibration relaxation time-scales of system.
- Elasticity of microstructures.

Two approaches

- Develop stiff stochastic time-step integrators.
- Perturbation analysis of SPDEs : reduced descriptions.

Stiff Time-step Integrator

Fluid equations

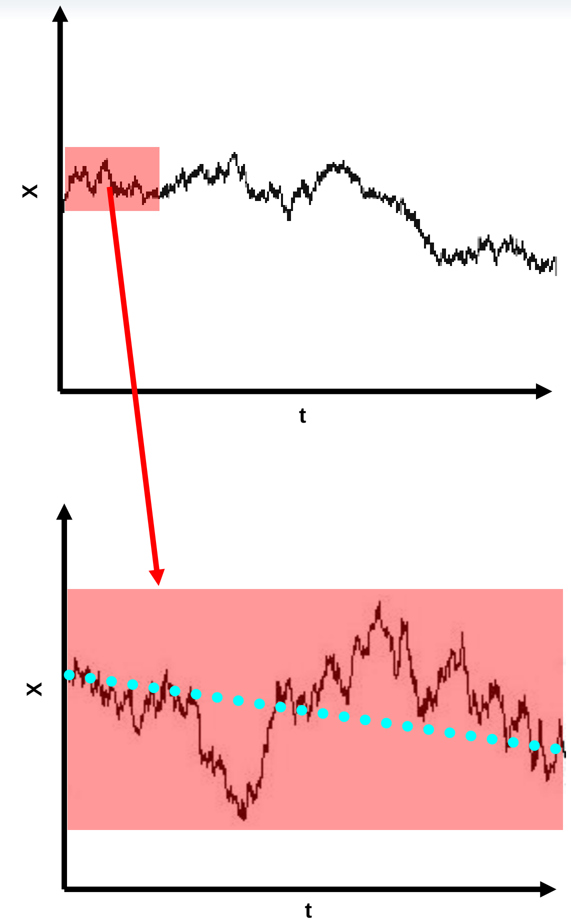
$$
\begin{aligned}
d\mathbf{u} &= \mathcal{L}\mathbf{u}\,dt && \text{(viscous damping)} \\
&+ \rho^{-1}\mathbf{F}_{\mathrm{prt}}\,dt && \text{(particle force)} \\
&+ Q\,d\mathbf{B}_t && \text{(thermal force)}
\end{aligned}
$$

$$\nabla \cdot \mathbf{u} = 0 \qquad \text{(incompressibility)}$$

Structure equations

$$\frac{d\mathbf{X}^{[j]}(t)}{dt} = \int \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))\,\mathbf{u}(\mathbf{x},t)\,d\mathbf{x}$$

$$\mathbf{F}_{\mathrm{prt}}(\mathbf{x},t) = \sum_{j=1}^{M} -\nabla_{\mathbf{X}^{[j]}} V(\{\mathbf{X}(t)\})\,\delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))$$



Integration by exponential factor (ito calculus)

$$= e^{t\mathcal{L}}\mathbf{u}(0) + \int_0^t e^{(t-s)\mathcal{L}}\rho^{-1}\mathbf{F}_{\mathrm{prt}}(s)\,ds + \int_0^t e^{(t-s)\mathcal{L}} Q\,d\mathbf{B}_s = e^{t\mathcal{L}}\mathbf{u}(0) + \bar{\mathbf{I}}_{\mathrm{prt}} + \bar{\mathbf{I}}_{\mathrm{thm}}$$

Stiff Time-step Integrator

Integration by exponential factor (ito calculus)

$$\mathbf{u}(t) = e^{t\mathcal{L}}\mathbf{u}(0) + \int_0^t e^{(t-s)\mathcal{L}}\rho^{-1}\mathbf{F}_{\mathrm{prt}}(s)ds + \int_0^t e^{(t-s)\mathcal{L}}Qd\mathbf{B}_s = e^{t\mathcal{L}}\mathbf{u}(0) + \bar{\mathbf{I}}_{\mathrm{prt}} + \bar{\mathbf{I}}_{\mathrm{thm}}$$

Particle force

$$\mathbf{I}_{\mathrm{prt}}(t) := \int_0^t e^{(t-s)\mathcal{L}}\rho^{-1}\mathbf{F}_{\mathrm{prt}}(s)ds$$

Approximate by constant force

$$\mathbf{I}_{\mathrm{prt}}(t) \approx -\rho^{-1}\mathcal{L}^{-1}\left[\mathcal{I} - e^{t\mathcal{L}}\right]\mathbf{F}_{\mathrm{prt}}(0)$$

Thermal fluctuations

$$\mathbf{I}_{\mathrm{thm}}(t) := \int_0^t e^{(t-s)\mathcal{L}}Qd\mathbf{B}_s$$

Ito calculus yields Gaussian with

$$\langle \mathbf{I}_{\mathrm{thm}}(t)\rangle = 0$$

$$\langle \mathbf{I}_{\mathrm{thm}}(t)\mathbf{I}_{\mathrm{thm}}(t)^T\rangle = \int_0^t e^{(t-s)\mathcal{L}}QQ^T e^{(t-s)\mathcal{L}^T}ds := \Lambda(t)$$

$$\Lambda_{\mathbf{k},\mathbf{k}}(t) = -\frac{1}{2\alpha_{\mathbf{k}}}\left[1 - e^{-2\alpha_{\mathbf{k}}\Delta t}\right]Q^2_{\mathbf{k},\mathbf{k}}$$

Stiff Time-step Integrator

Integration by exponential factor (ito calculus)

$$\mathbf{u}(t) \;=\; e^{t\mathcal{L}}\mathbf{u}(0) + \int_0^t e^{(t-s)\mathcal{L}}\rho^{-1}\mathbf{F}_{\text{prt}}(s)ds + \int_0^t e^{(t-s)\mathcal{L}}Q d\mathbf{B}_s \;=\; e^{t\mathcal{L}}\mathbf{u}(0) + \bar{\mathbf{I}}_{\text{prt}} + \bar{\mathbf{I}}_{\text{thm}}$$

$$\mathbf{I}_{\text{prt}}(t) \approx -\rho^{-1}\mathcal{L}^{-1}\left[\mathcal{I} - e^{t\mathcal{L}}\right]\mathbf{F}_{\text{prt}}(0)$$

$$\Lambda_{\mathbf{k},\mathbf{k}}(t) = -\frac{1}{2\alpha_{\mathbf{k}}}\left[1 - e^{-2\alpha_{\mathbf{k}}\Delta t}\right]Q^2_{\mathbf{k},\mathbf{k}}$$

Fluid Integrator

$$\mathbf{u}^{n+1} \;=\; e^{\Delta t\mathcal{L}}\mathbf{u}^n + \mathcal{L}^{-1}\left[\mathcal{I} - e^{\Delta t\mathcal{L}}\right]\rho^{-1}\mathbf{F}^n_{\text{prt}} + \Gamma\xi^n$$

ξ is Gaussian with

$\langle\xi\rangle = 0$, $\langle\xi\xi^T\rangle = \mathcal{I}$

$\Lambda = \Gamma\Gamma^T$

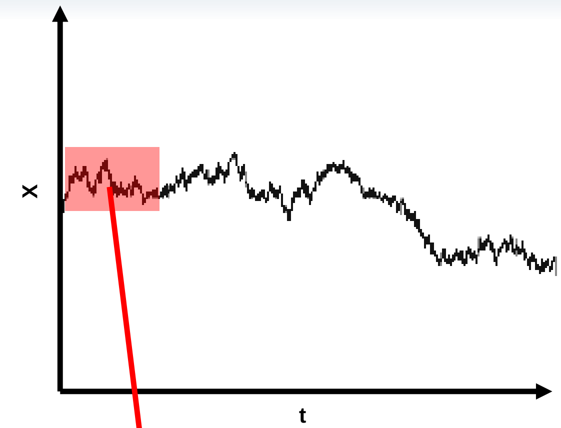
- unconditionally stable.
- accuracy depends only on structure force approximation (otherwise exact).
- requires prior knowledge of Γ.
- method viable only if efficient to compute $e^{\Delta t\mathcal{L}}$.
- viable for uniform meshes (FFTs).

Stiff Time-step Integrator

Fluid equations

$$\begin{aligned} d\mathbf{u} &= \mathcal{L}\mathbf{u}dt && \text{(viscous damping)} \\ &+ \rho^{-1}\mathbf{F}_{\text{prt}}dt && \text{(particle force)} \\ &+ Qd\mathbf{B}_t && \text{(thermal force)} \end{aligned}$$

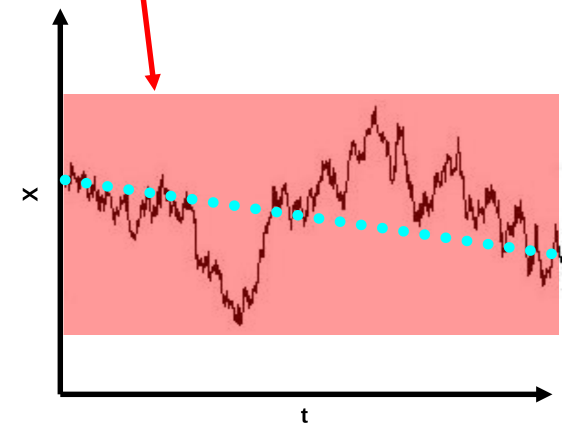
$$\nabla \cdot \mathbf{u} = 0 \qquad \text{(incompressibility)}$$



Structure equations

$$\frac{d\mathbf{X}^{[j]}(t)}{dt} = \int \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))\mathbf{u}(\mathbf{x},t)d\mathbf{x}$$

$$\mathbf{F}_{\text{prt}}(\mathbf{x},t) = \sum_{j=1}^{M} -\nabla_{\mathbf{X}^{[j]}} V(\{\mathbf{X}(t)\})\delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))$$



Integrate structure dynamics (ito calculus)

$$\mathbf{X}^{[j]}(t) = \mathbf{X}^{[j]}(0) + \int_0^t \int \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(s))\mathbf{u}(\mathbf{x},s)d\mathbf{x}ds \approx \mathbf{X}^{[j]}(0) + \int \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(0)) \int_0^t \mathbf{u}(\mathbf{x},s)dsd\mathbf{x}$$

Stiff Time-step Integrator

Integrate structure dynamics (ito calculus)

$$\mathbf{X}^{[j]}(t) \approx \mathbf{X}^{[j]}(0) + \int \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(0)) \int_0^t \mathbf{u}(\mathbf{x}, s)\, ds\, d\mathbf{x}$$

$$\longrightarrow \mathbf{X}^{[j],n+1} = \mathbf{X}^{[j],n} + \int \delta_a(\mathbf{x} - \mathbf{X}^{[j],n}) \mathbf{I}_{\text{vel}}(\mathbf{x}, \Delta t)\, d\mathbf{x}$$

$$\mathbf{I}_{\text{vel}}(t) := \int_0^t \mathbf{u}(s)\, ds$$

Integrated fluctuating fluid velocity

$\mathbf{I}_{\text{vel}}(t)$ is a Gaussian with

$$\bar{\mathbf{I}}_{\text{vel}} := \langle \mathbf{I}_{\text{vel}}(t) \rangle = \int_0^t \langle \mathbf{u}(s) \rangle ds = -\mathcal{L}^{-1}\left[\mathcal{I} - e^{t\mathcal{L}}\right] \mathbf{u}(0) + -\mathcal{L}^{-1}\left[t + \mathcal{L}^{-1}\left[\mathcal{I} - e^{t\mathcal{L}}\right]\right] \mathbf{F}_{\text{prt}}(0)$$

$$\Phi := \left\langle \left(\mathbf{I}_{\text{vel}}(t) - \bar{\mathbf{I}}_{\text{vel}}(t)\right)\left(\mathbf{I}_{\text{vel}}^T(t) - \bar{\mathbf{I}}_{\text{vel}}^T(t)\right)\right\rangle = \int_0^t \int_0^t e^{r\mathcal{L}} C e^{s\mathcal{L}^T} dr ds + \int_0^t \int_0^t \int_0^{s \wedge r} e^{(r-w)\mathcal{L}} Q Q^T e^{(s-w)\mathcal{L}^T} dw dr ds$$

$\mathbf{I}_{\text{vel}}(t)$ is correlated with $\mathbf{I}_{\text{thm}}(t)$

$$W := \left\langle \left(\mathbf{I}_{\text{vel}}(t) - \bar{\mathbf{I}}_{\text{vel}}(t)\right) \mathbf{I}_{\text{thm}}^T(t)\right\rangle = \mathcal{L}^{-1} \int_0^t e^{(t-w)\mathcal{L}} Q Q^T e^{(t-w)\mathcal{L}^T} dw + \mathcal{L}^{-1} Q Q^T \mathcal{L}^{-T}\left[\mathcal{I} - e^{t\mathcal{L}^T}\right]$$

Structure Integrator

$$\mathbf{X}^{[j],n+1} = \mathbf{X}^{[j],n} + \int \delta_a(\mathbf{x} - \mathbf{X}^{[j],n}) \mathbf{I}_{\text{vel}}(\mathbf{x}, \Delta t)\, d\mathbf{x}$$

- stability depends now on structure forces.
- accuracy depends on
  - fluid sampling approximation X(t) ~ X(0) and structure force approximation.
- method viable only if efficient to compute exponentials.
- viable for uniform meshes (FFTs).

Summary : Stiff Integrator

Fluid Integrator

$$\mathbf{u}^{n+1} = e^{\Delta t \mathcal{L}} \mathbf{u}^n + \mathcal{L}^{-1} \left[\mathcal{I} - e^{\Delta t \mathcal{L}} \right] \rho^{-1} \mathbf{F}^n_{\text{prt}} + \Gamma \xi^n$$

ξ is Gaussian with

$$\langle \xi \rangle = 0, \quad \langle \xi \xi^T \rangle = \mathcal{I}$$

$$\Lambda = \Gamma \Gamma^T$$

Structure Integrator

$$\mathbf{X}^{[j],n+1} = \mathbf{X}^{[j],n} + \int \delta_a(\mathbf{x} - \mathbf{X}^{[j],n}) \mathbf{I}_{\text{vel}}(\mathbf{x}, \Delta t) d\mathbf{x}$$

$\mathbf{I}_{\text{vel}}(t)$ is a Gaussian with

$$\bar{\mathbf{I}}_{\text{vel}} := \langle \mathbf{I}_{\text{vel}}(t) \rangle = \int_0^t \langle \mathbf{u}(s) \rangle ds = -\mathcal{L}^{-1} \left[\mathcal{I} - e^{t\mathcal{L}} \right] \mathbf{u}(0) + -\mathcal{L}^{-1} \left[t + \mathcal{L}^{-1} \left[\mathcal{I} - e^{t\mathcal{L}} \right] \right] \mathbf{F}_{\text{prt}}(0)$$

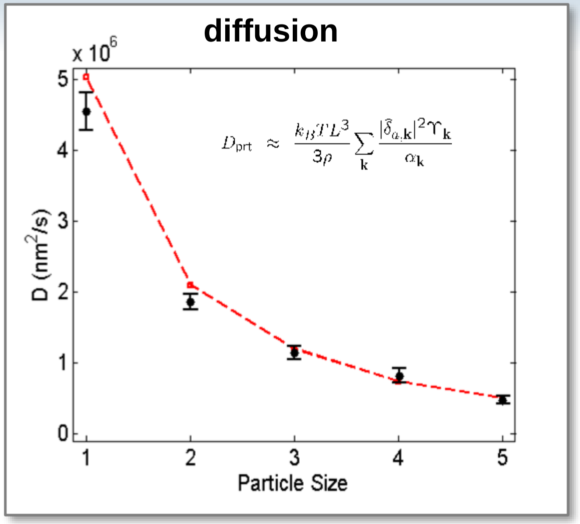
$$\Phi := \langle \left(\mathbf{I}_{\text{vel}}(t) - \bar{\mathbf{I}}_{\text{vel}}(t) \right) \left(\mathbf{I}^T_{\text{vel}}(t) - \bar{\mathbf{I}}^T_{\text{vel}}(t) \right) \rangle = \int_0^t \int_0^t e^{r\mathcal{L}} C e^{s\mathcal{L}^T} dr ds + \int_0^t \int_0^t \int_0^{s \wedge r} e^{(r-w)\mathcal{L}} Q Q^T e^{(s-w)\mathcal{L}^T} dw dr ds$$

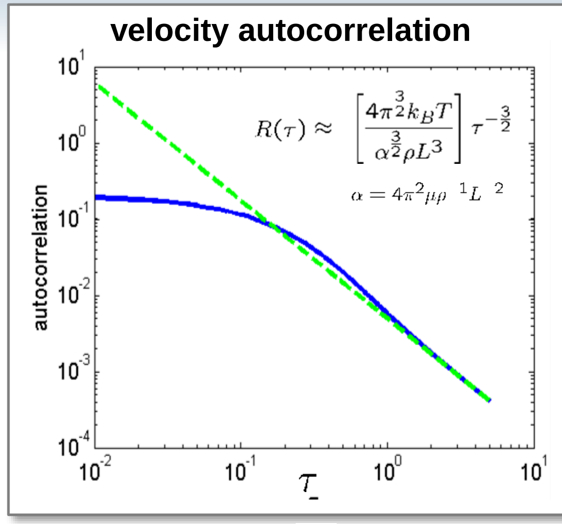
$\mathbf{I}_{\text{vel}}(t)$ is correlated with $\mathbf{I}_{\text{thm}}(t)$

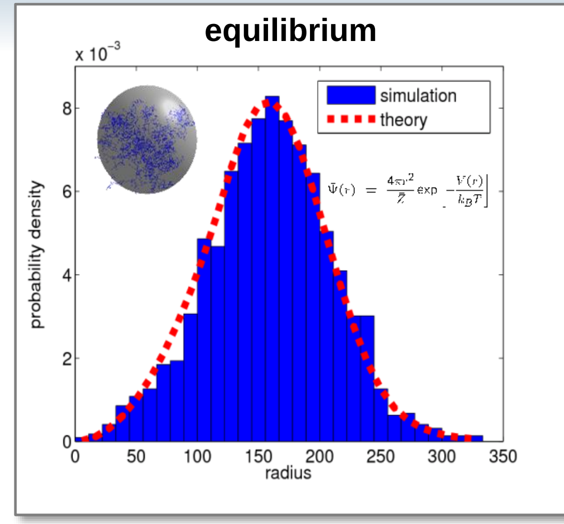
$$W := \langle \left(\mathbf{I}_{\text{vel}}(t) - \bar{\mathbf{I}}_{\text{vel}}(t) \right) \mathbf{I}^T_{\text{thm}}(t) \rangle = \mathcal{L}^{-1} \int_0^t e^{(t-w)\mathcal{L}} Q Q^T e^{(t-w)\mathcal{L}^T} dw + \mathcal{L}^{-1} Q Q^T \mathcal{L}^{-T} \left[\mathcal{I} - e^{t\mathcal{L}^T} \right]$$

- method viable only if efficient to compute exponentials.
- viable for uniform meshes (FFTs).
- under-resolves fluid mode dynamics and fluctuations.
- time-step limited by structure's motions.

Validation of Numerical Methods

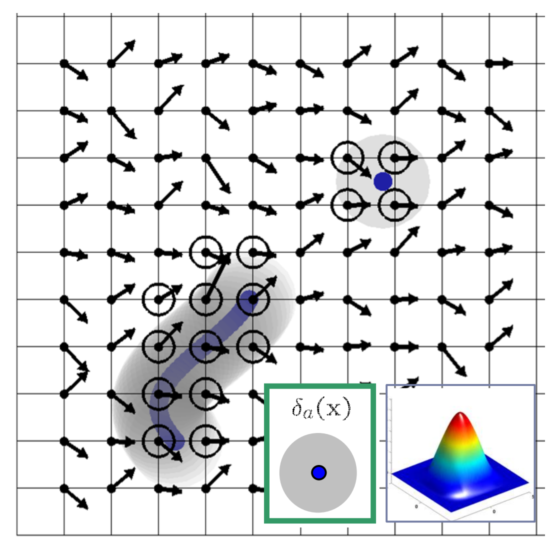






Validation

- Diffusivity of under-resolved particles correct.
- Velocity auto-correlation has $t^{-3/2}$ tail (Adler & Wainright 1950),
- Auto-correlation persists from hydrodynamic “memory.”
- Equilibrium configurations have Gibbs-Boltzmann statistics.
- Can ideas be extended to other coupling types and regimes?



Generalization : Stochastic Eulerian Lagrangian Methods

Fluid

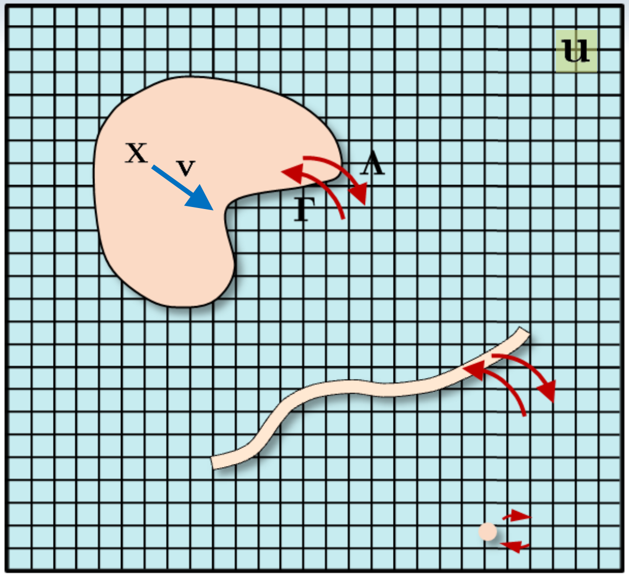
$$\rho \frac{\partial \mathbf{u}}{\partial t} = \mathcal{L}\mathbf{u} + \Lambda[\Upsilon(\mathbf{v} - \Gamma\mathbf{u})] + \lambda + \mathbf{f}_{\mathrm{thm}}$$
$$\nabla \cdot \mathbf{u} = 0$$

Structure

$$\frac{d\mathbf{X}}{dt} = \mathbf{v}$$
$$m\frac{d\mathbf{v}}{dt} = -\Upsilon\left(\mathbf{v} - \Gamma\mathbf{u}\right) - \nabla_{\mathbf{X}}\Phi[\mathbf{X}] + \zeta + \mathbf{F}_{\mathrm{thm}}$$

Thermal fluctuations

$$\langle \mathbf{f}_{\mathrm{thm}}(s)\mathbf{f}_{\mathrm{thm}}^T(t)\rangle = -\left(2k_BT\right)\left(\mathcal{L} - \Lambda\Upsilon\Gamma\right)\delta(t-s)$$
$$\langle \mathbf{F}_{\mathrm{thm}}(s)\mathbf{F}_{\mathrm{thm}}^T(t)\rangle = \left(2k_BT\right)\Upsilon\,\delta(t-s)$$
$$\langle \mathbf{f}_{\mathrm{thm}}(s)\mathbf{F}_{\mathrm{thm}}^T(t)\rangle = -\left(2k_BT\right)\Lambda\Upsilon\,\delta(t-s).$$



Operators:

- $\mathcal{L} \longrightarrow$ Fluid dissipation (viscosity).
- $\Upsilon \longrightarrow$ Structure "slip" relative to local flow field.
- $\Gamma \longrightarrow$ Kinematic particle velocity for given flow.
- $\Lambda \longrightarrow$ Induced fluid force density from particle.

Notation:

- $\mathbf{u} = \mathbf{u}(\mathbf{x}, t) \longrightarrow$ Fluid velocity.
- $\mathbf{X} = \mathbf{X}(\mathbf{q}, t) \longrightarrow$ Structure configuration
- $\mathbf{v} = \mathbf{v}(\mathbf{q}, t) \longrightarrow$ Structure velocity.

Atzberger 2011

Coupling Operators

Conservation of momentum

$$\int_\Omega (\Lambda \mathbf{F})(\mathbf{x}) d\mathbf{x} = \int_S \mathbf{F}(\mathbf{q}) d\mathbf{q}$$

→ "integrates to one."

Conservation of energy

(overdamped limit)

$$E[\mathbf{u}, \mathbf{X}] = \frac{1}{2} \int \rho |\mathbf{u}(\mathbf{y})|^2 d\mathbf{y} + \Phi(\mathbf{X})$$

Adjoint condition

$$\int_S (\Gamma \mathbf{u})(\mathbf{q}) \cdot \mathbf{F}(\mathbf{q}) d\mathbf{q} = \int_\Omega \mathbf{u}(\mathbf{x}) \cdot (\Lambda \mathbf{F})(\mathbf{x}) d\mathbf{x}$$

→ $\langle \Gamma \mathbf{u}, \mathbf{F} \rangle = \langle \mathbf{u}, \Lambda \mathbf{F} \rangle$ ⟶ "$\Gamma = \Lambda^T$"

- Energy conserved → coupling operators are adjoints!
- Useful for deriving coupling operators.

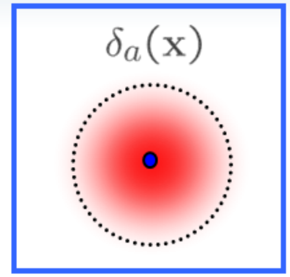
Immersed Boundary Method

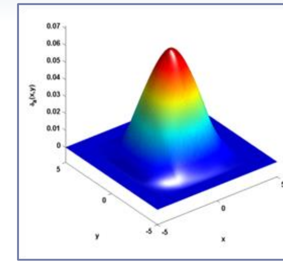
Coupling operators

$$\Gamma[u] = \int \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))\mathbf{u}(\mathbf{x}, t)d\mathbf{x}$$

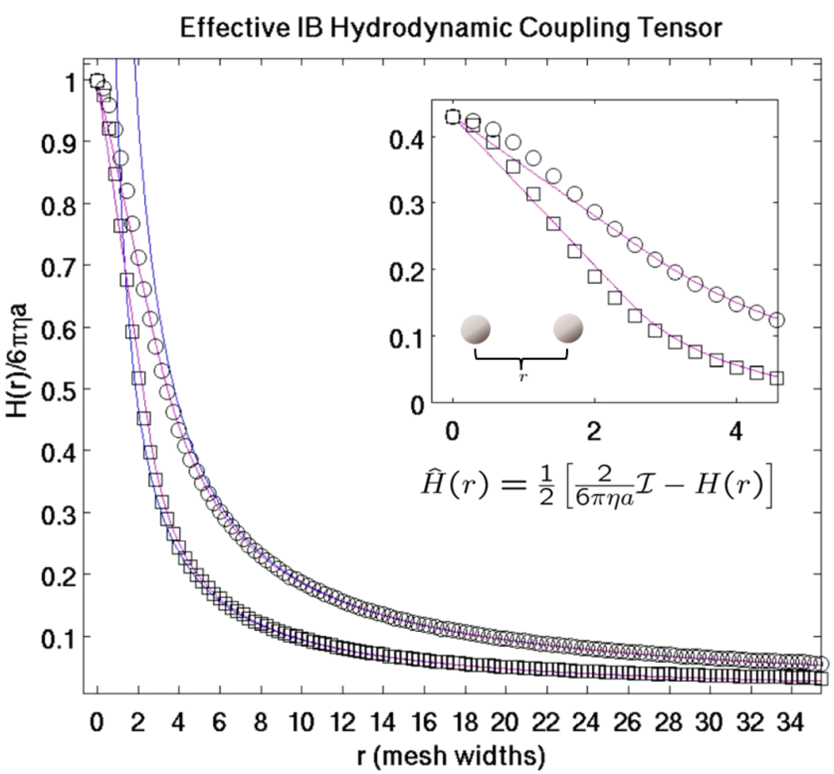
$$\Lambda[F] = \delta_a(\mathbf{x} - \mathbf{X}^{[j]}(t))\mathbf{F}$$

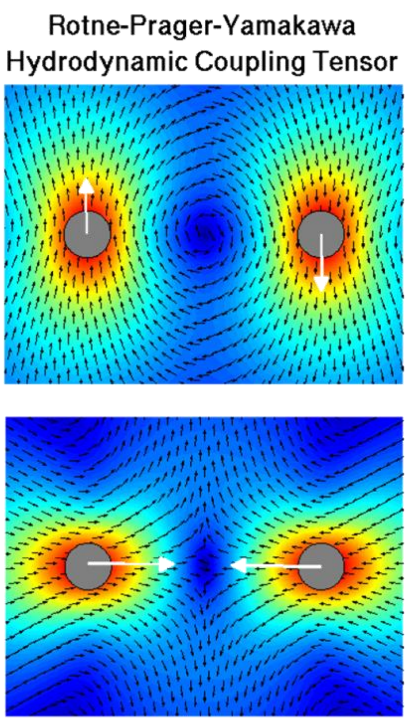
$$\text{“}\Gamma = \Lambda^T\text{”}$$



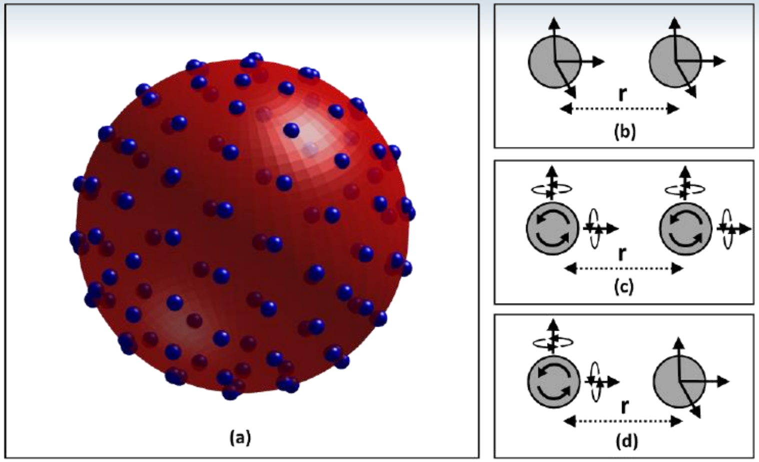


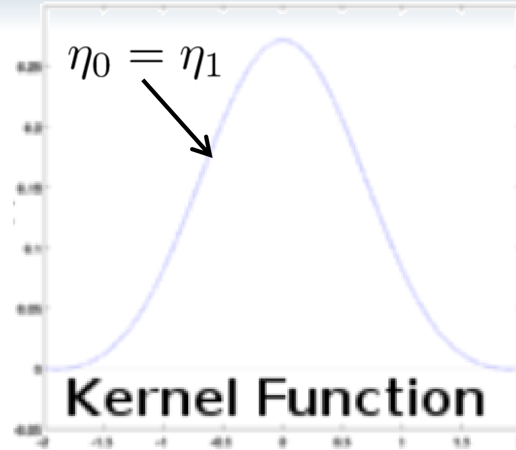
Peskin delta-function





Coupling Operators based on Faxen Relations





Faxen Kinematic Relations→ Γ :

$$\Gamma_0 \mathbf{u} = \sum_{\mathbf{m}} \langle\, \eta_0(\mathbf{y_m} - (\mathbf{X}_{cm} + \mathbf{z}))\; \mathbf{u_m} \,\rangle_{\tilde{\mathcal{S}},|\mathbf{z}|=R} \Delta x_{\mathbf{m}}^3$$

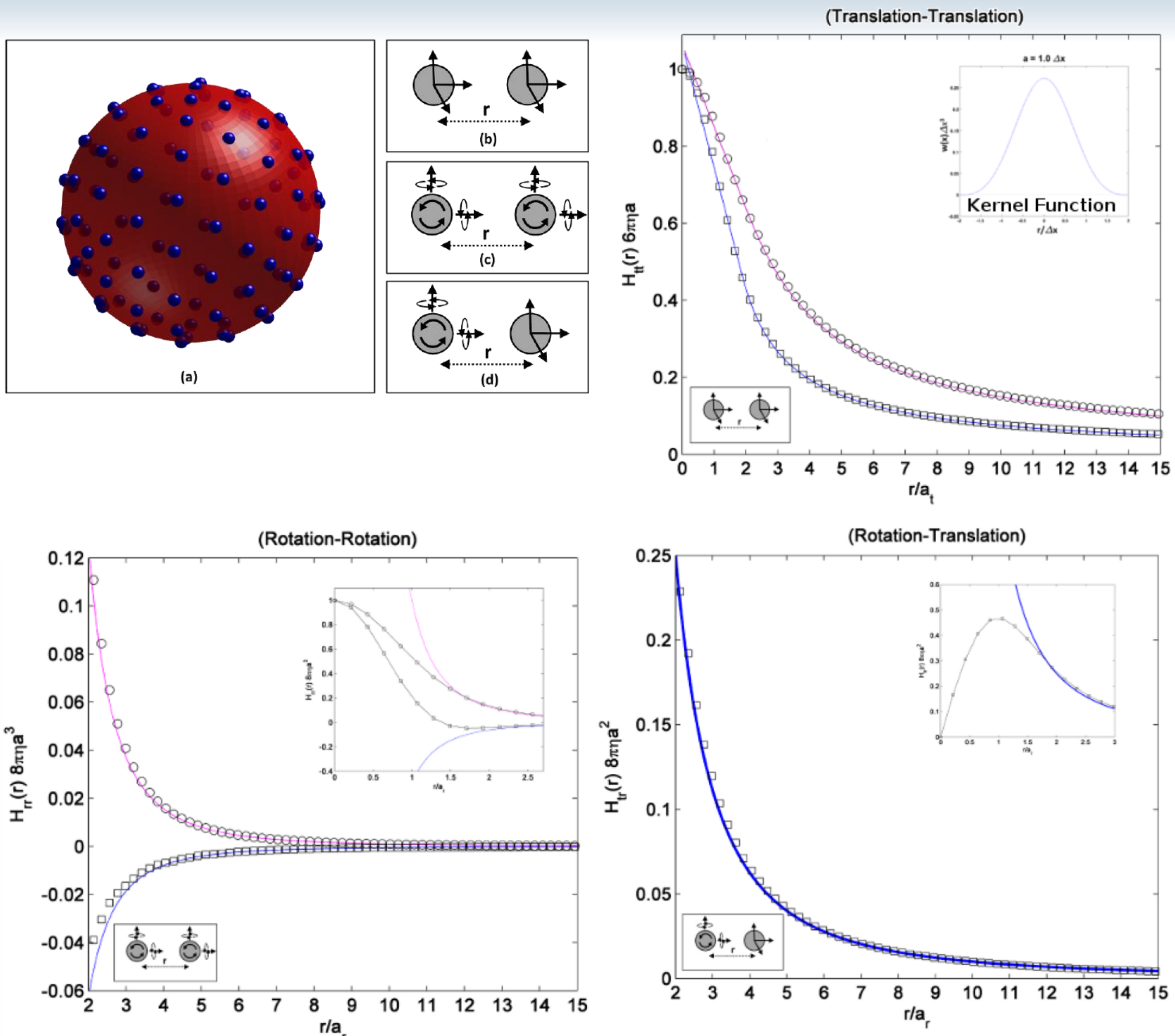
$$\Gamma_1 \mathbf{u} = \frac{3}{2R^2} \sum_{\mathbf{m}} \langle\, \eta_1(\mathbf{y_m} - (\mathbf{X}_{cm} + \mathbf{z}))\, (\mathbf{z} \times \mathbf{u_m}) \,\rangle_{\tilde{\mathcal{S}},|\mathbf{z}|=R} \Delta x_{\mathbf{m}}^3.$$

Adjoint Condition → Λ :

$$\Lambda_0(\mathbf{x_m}) = \left(\langle\, \eta_0(\mathbf{x_m} - (\mathbf{X}_{cm} + \mathbf{z})) \,\rangle_{\tilde{\mathcal{S}},|\mathbf{z}|=R} \right) \mathbf{F}$$

$$\Lambda_1(\mathbf{x_m}) = -\frac{3}{2R^2} \left(\langle\, \mathbf{z}\eta_1(\mathbf{x_m} - (\mathbf{X}_{cm} + \mathbf{z})) \,\rangle_{\tilde{\mathcal{S}},|\mathbf{z}|=R} \right) \times \mathbf{T}.$$

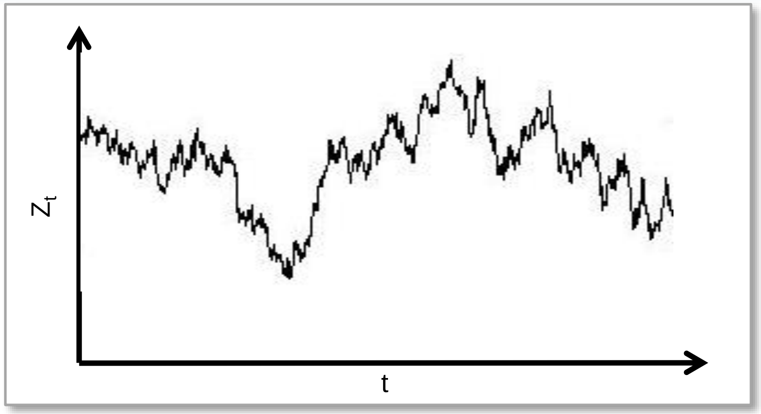
Coupling Operators based on Faxen Relations



Excellent agreement for r > 2a !

Numerical Stiffness

Time-scales	
Fluid Modes	**Particle Diffusion**
$\tau_\lambda = \frac{\rho}{4\pi^2\mu}\lambda^2$	$\tau_{\text{diff}}(a) \approx \frac{a^2}{D_a}$
λ = 10nm : $\tau = 10^{-3}$ns	$\tau_{\text{diff}}(1\text{nm}) \approx 10^0\text{ns}$
λ = 1000nm : τ = 10ns	$\tau_{\text{diff}}(10\text{nm}) \approx 10^3\text{ns}$



Sources of stiffness

- In SELM additional sources of stiffness from
  - microstructure inertia
  - fluid-structure slip $-\Upsilon\left(\mathbf{v} - \Gamma\mathbf{u}\right)$
- Thermal fluctuations also excite coupling modes and all fluid modes.
- Elasticity of microstructures.
- Equilibration time-scales of system vary over wide range.

Two approaches

- Develop stiff stochastic time-step integrators (as for SIBM).
- Perturbation analysis of SPDEs : reduced descriptions.

Stochastic Reduction

Stochastic differential equation:

$$d\mathbf{Z}(t) = \mathbf{a}(\mathbf{Z}(t))dt + \mathbf{b}(\mathbf{Z}(t))d\mathbf{W}_t \longrightarrow \mathcal{A}_\epsilon = \mathrm{a} \cdot \frac{\partial}{\partial \mathrm{z}} + \frac{1}{2}\mathrm{b}\mathrm{b}^T : \frac{\partial^2}{\partial \mathrm{z}^2}$$

Backward-Kolomogorov PDE:

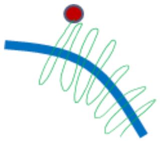
$$\frac{\partial u}{\partial t} = \mathcal{A}_\epsilon u$$

$$u(x,0) = f(x) \longrightarrow u(x,t) = E^{x,0}\left[f(X_t)\right]$$

Perturbation Analysis:

$$u(\mathbf{z},t) = u_0(\mathbf{z},t) + u_1(\mathbf{z},t)\epsilon + u_2(\mathbf{z},t)\epsilon^2 \cdots + u_n(\mathbf{z},t)\epsilon^n + \cdots$$

Split operator into "slow" and "fast" parts:



invariant distribution

$$\mathcal{A}_\epsilon = L_{slow} + L_{fast} \rightarrow L_{fast} = \frac{1}{\epsilon}\left(L_2 + \epsilon \tilde{L}_2\right), \quad L_2^*\Psi = 0, \int \Psi d\mathbf{z}_f = 1$$

average drift part — remainder

$$\rightarrow L_{slow} = \bar{L}_1 + L_1 \rightarrow \bar{L}_1 = \int \Psi(\mathbf{z}_f|\mathbf{z}_s) L_{slow} d\mathbf{z}_f, \quad L_1 = L_{slow} - \bar{L}_1$$

$$\mathcal{A}_\epsilon = \bar{L}_1 + \epsilon L_\epsilon, \quad L_\epsilon = \frac{1}{\epsilon}\left(L_1 + \tilde{L}_2\right) + \frac{1}{\epsilon^2} L_2$$

ε → 0 : compare orders

$$\mathcal{A} = \bar{L}_1 + \epsilon \bar{L}_0$$ leading order dynamics. $$\bar{L}_0 = -\int \Psi \left(L_1 + \tilde{L}_2\right) L_2^{-1} L_1 d\mathbf{z}_f$$

Reduced dynamics:

$$\mathcal{A} = \tilde{\mathrm{a}} \cdot \frac{\partial}{\partial \mathrm{z}} + \frac{1}{2}\tilde{\mathrm{b}}\tilde{\mathrm{b}}^T : \frac{\partial^2}{\partial \mathrm{z}^2} \longrightarrow d\tilde{\mathbf{Z}}_t = \tilde{\mathbf{a}}(\tilde{\mathbf{Z}}_t)dt + \tilde{\mathbf{b}}(\tilde{\mathbf{Z}}_t)d\tilde{\mathbf{W}}_t$$

Atzberger & Tabak 2015

Summary of regimes

Stochastic Eulerian Lagrangian Method (SELM)

Fluid dynamics:

$$\rho\frac{\partial \mathbf{u}}{\partial t} = \mu\Delta\mathbf{u} - \nabla p + \Lambda[\Upsilon(\mathbf{v} - \Gamma\mathbf{u})] + \mathbf{f}_{\text{thm}}$$
$$\nabla\cdot\mathbf{u} = 0$$

Structure dynamics:

$$\frac{d\mathbf{X}}{dt} = \mathbf{v}$$
$$m\frac{d\mathbf{v}}{dt} = -\Upsilon(\mathbf{v} - \Gamma\mathbf{u}) - \nabla_{\mathbf{X}}\Phi[\mathbf{X}] + \zeta + \mathbf{F}_{\text{thm}}$$

Thermal Fluctuations

$$\langle \mathbf{f}_{\text{thm}}(s)\mathbf{f}_{\text{thm}}^T(t)\rangle = -(2k_BT)(\mu\Delta - \Lambda\Upsilon\Gamma)\,\delta(t-s)$$
$$\langle \mathbf{F}_{\text{thm}}(s)\mathbf{F}_{\text{thm}}^T(t)\rangle = (2k_BT)\,\Upsilon\,\delta(t-s)$$
$$\langle \mathbf{f}_{\text{thm}}(s)\mathbf{F}_{\text{thm}}^T(t)\rangle = -(2k_BT)\,\Lambda\Upsilon\,\delta(t-s).$$

Microstructure density matched with fluid

$m \ll \rho\ell^3$

Fluid-structure dynamics:

$$\frac{d\mathbf{p}}{dt} = \rho^{-1}\mathcal{L}\mathbf{p} + \Lambda[-\nabla_{\mathbf{X}}\Phi(\mathbf{X})] + (\nabla_{\mathbf{X}}\cdot\Lambda)\,k_BT + \lambda + \mathbf{g}_{\text{thm}}$$
$$\frac{d\mathbf{X}}{dt} = \rho^{-1}\Gamma\mathbf{p} + \Upsilon^{-1}[-\nabla_{\mathbf{X}}\Phi(\mathbf{X})] + \zeta + \mathbf{G}_{\text{thm}}.$$
$$\nabla_{\mathbf{X}}\cdot\Lambda = \text{Tr}[\nabla_{\mathbf{X}}\Lambda]$$

Phase space compressibility (p,X).

Thermal Fluctuations:

$$\langle \mathbf{g}_{\text{thm}}(s)\mathbf{g}_{\text{thm}}^T(t)\rangle = -(2k_BT)\,\mathcal{L}\,\delta(t-s)$$
$$\langle \mathbf{G}_{\text{thm}}(s)\mathbf{G}_{\text{thm}}^T(t)\rangle = (2k_BT)\,\Upsilon^{-1}\,\delta(t-s)$$
$$\langle \mathbf{g}_{\text{thm}}(s)\mathbf{G}_{\text{thm}}^T(t)\rangle = 0.$$

- Structure momentum no longer tracked.
- Removes a source of stiffness.
- Non-conjugate Hamiltonian formulation yields metric-factor in phase-space.

Microstructure-fluid no-slip coupling (S-Immersed-Boundary)

$\Upsilon \to \infty$

Fluid-Structure Equations:

$$\frac{d\mathbf{p}}{dt} = \rho^{-1}\mathcal{L}\mathbf{p} + \Lambda[-\nabla_{\mathbf{X}}\Phi(\mathbf{X})] + (\nabla_{\mathbf{X}}\cdot\Lambda)\,k_BT + \lambda + \mathbf{g}_{\text{thm}}$$
$$\frac{d\mathbf{X}}{dt} = \rho^{-1}\Gamma\mathbf{p}$$

Thermal Fluctuations:

$$\langle \mathbf{g}_{\text{thm}}(s)\mathbf{g}_{\text{thm}}^T(t)\rangle = -(2k_BT)\,\mathcal{L}\,\delta(t-s).$$

- Structure dynamics no-longer inertial.
- Removes additional sources of stiffness.
- Regime of the Stochastic Immersed Boundary Method.
- Phase-space metric reflected in the drift term.

Microstructure-fluid stress balance

$\mu \to \infty$

Fluid-Structure Equations:

$$\frac{d\mathbf{X}}{dt} = H_{\text{SELM}}[-\nabla_{\mathbf{X}}\Phi(\mathbf{X})] + (\nabla_{\mathbf{X}}\cdot H_{\text{SELM}})k_BT + \mathbf{h}_{\text{thm}}$$
$$H_{\text{SELM}} = \Gamma(-\wp\mathcal{L})^{-1}\Lambda$$

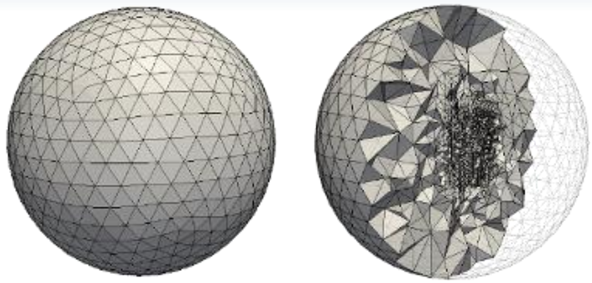
Thermal Fluctuations:

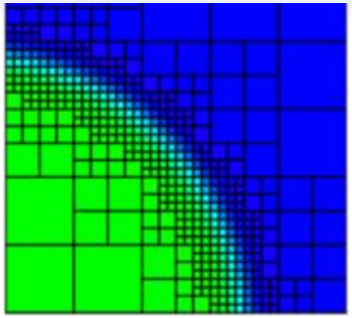
$$\langle \mathbf{h}_{\text{thm}}(s)\mathbf{h}_{\text{thm}}^T(t)\rangle = (2k_BT)\,H_{\text{SELM}}\,\delta(t-s).$$

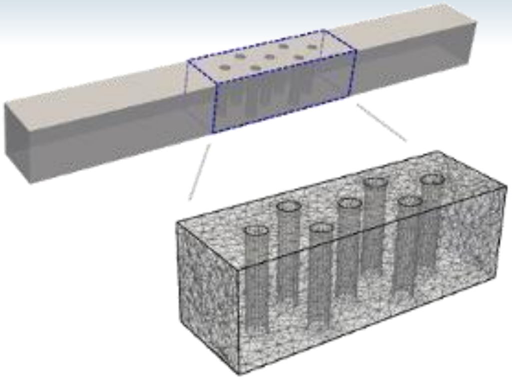
- Fluid momentum no longer tracked.
- Balance of hydrodynamic stresses with elastic stresses.
- Removes additional sources of stiffness.
- Regime of the Stokesian-Brownian Dynamics (Brady 1980, McCammond 1980's).
- Phase-space metric reflected in the drift term.

Atzberger & Tabak 2015

Adaptive Meshes







Fluid dynamics

$$
\begin{aligned}
\rho \frac{\partial \mathbf{u}}{\partial t} &= \mathcal{L}\mathbf{u} + \Lambda[\Upsilon(\mathbf{v} - \Gamma\mathbf{u})] + \lambda + \mathbf{f}_{\mathrm{thm}} \\
\nabla \cdot \mathbf{u} &= 0
\end{aligned}
$$

Structure dynamics

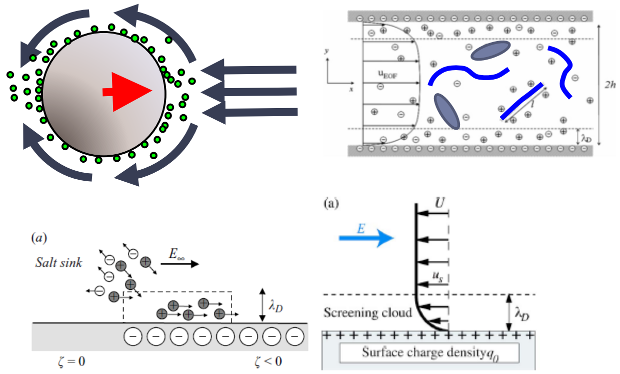
$$
\begin{aligned}
\frac{d\mathbf{X}}{dt} &= \mathbf{v} \\
m\frac{d\mathbf{v}}{dt} &= -\Upsilon\left(\mathbf{v} - \Gamma\mathbf{u}\right) - \nabla_{\mathbf{X}}\Phi[\mathbf{X}] + \zeta + \mathbf{F}_{\mathrm{thm}}
\end{aligned}
$$

- Thermal fluctuation propagation pose challenges for non-uniform discretizations.
- Dissipative numerical operators need to be compatible stochastic driving fields.
- Additional time-scales arise from the microstructure – fluid momentum coupling.
- We developed Finite Element Methods + Stochastic Iterative Methods for SELM.

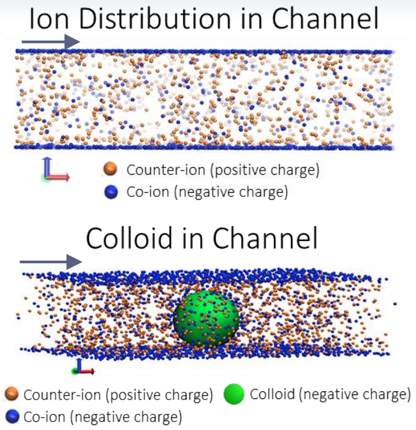
Atzberger, Plunkett, Hu, Siefert 2014

Fluidics Transport

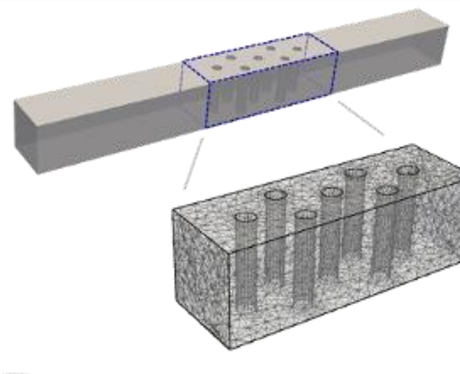
Fluidic Devices



Electrokinetics



Geometry / Confinement



Fluidic Devices

- Developed to miniaturize and automate many laboratory tests, diagnostics, characterization.
- Hydrodynamic transport at such scales must grapple with dissipation / friction.
- Electrokinetic effects utilized to drive flow.

Key Features

- Large surface area to volume.
- Ionic double-layers can be comparable to channel width.
- Brownian motion plays important role in ion distribution and analyte diffusion across channel.
- Hydrodynamic flow effected by close proximity to walls or other geometric features.
- Ionic concentrations often in regime with significant discrete correlations /density fluctuations.

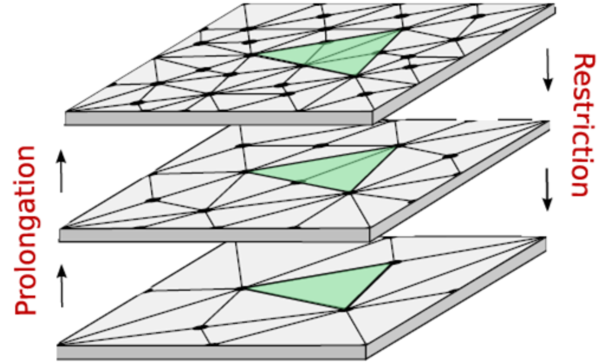
Challenges

- Develop theory and methods beyond mean-field Poisson-Boltzmann theory.
- Methods capable of handling hydrodynamics, fluctuations, geometry/confinement.

Stochastic Iterative Methods

Stochastic iteration

$$\xi^{n+1} = M\xi^n + N\mathrm{b} \boxed{+ \eta^n}$$

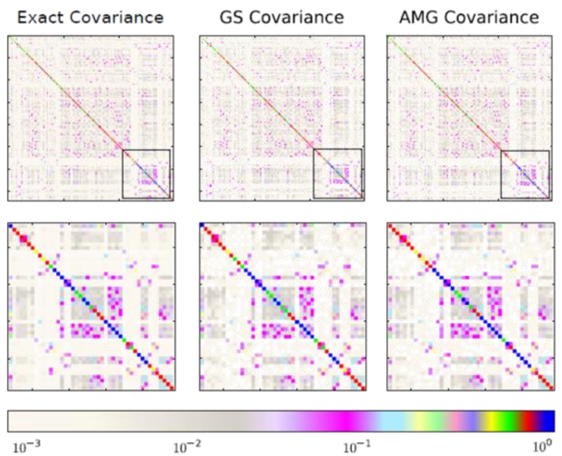


Choice related to target covariance C by

$$G = \langle \eta^n \eta^{n,T} \rangle \qquad G = C - MCM^T$$

Autocorrelation in the sampler satisfies

$$\Phi^m = \langle \xi^n (\xi^{n+m})^T \rangle$$
$$\Phi^{m+1} = M\Phi^m$$



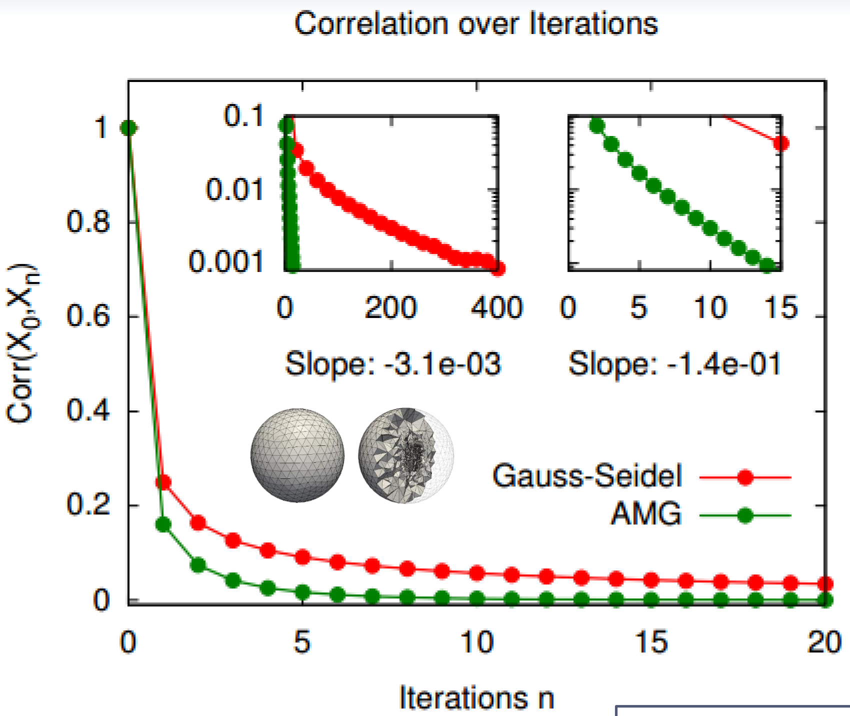
(Atzberger & Plunkett 2014)

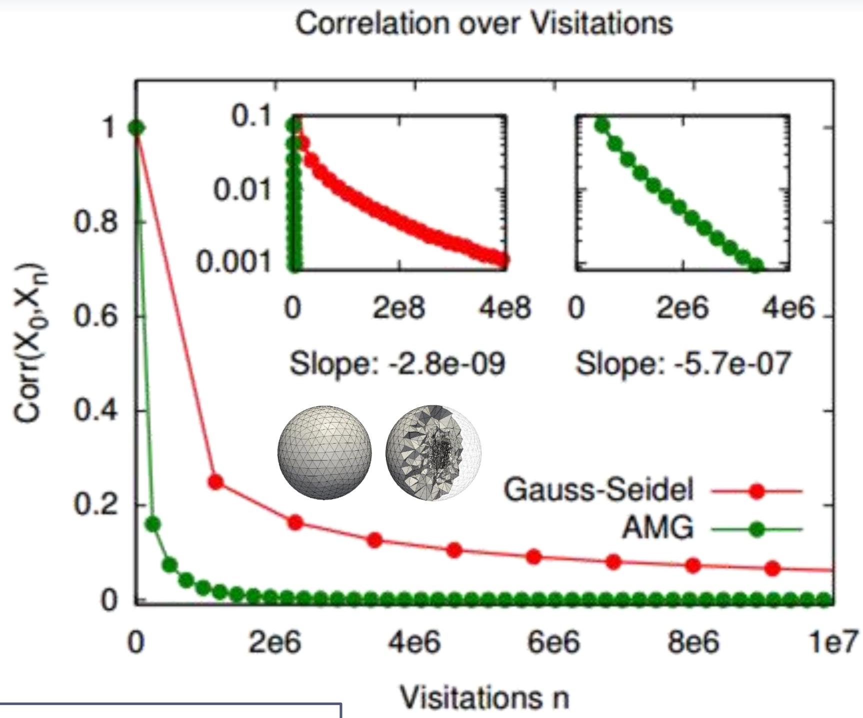
Gauss-Siedel iterations (Goodman & Sokal 1986)

$$G = (D-L-U)^{-1} - (D-L)^{-1}U(D-L-U)^{-1}L(D-L)^{-T}$$

Preconditioners improve decorrelations (multigrid , cg).

Stochastic Multigrid



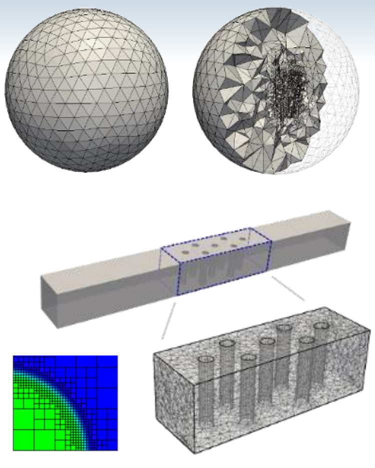


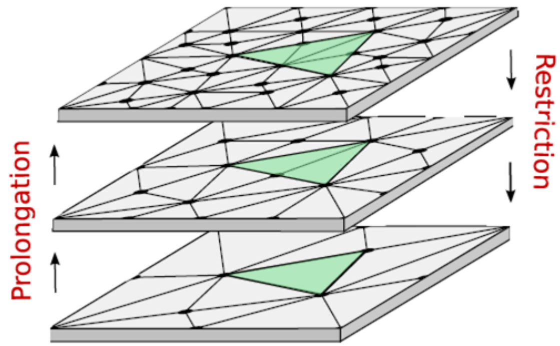
Error < 1%

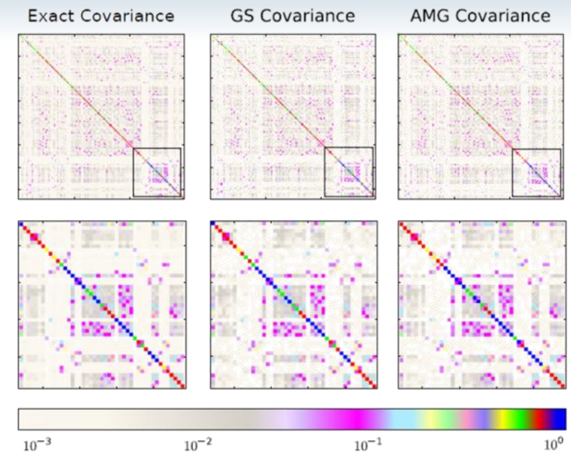
S-Gauss-Siedel	S-Multigrid
~100 iterations	~ 10 iterations
$\sim 10^8$ visitations	$\sim 10^6$ visitations
-3×10^{-3} rate exp	-1×10^{-1} rate exp

2014 Atzberger & Plunkett

Summary of Solvers







(Atzberger & Plunkett 2014)

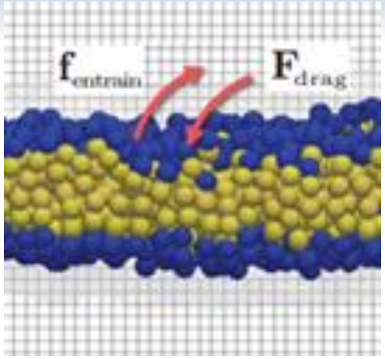
Solvers Developed

- Compatible stochastic discretizations developed for SELM (F-D, G-B).
  - Finite Volume / Spectral / Finite Element
- Stochastic field generation methods
  - Factorization methods.
  - Fast Fourier Transforms (FFTs).
  - Stochastic Iterative Methods (multigrid, cg).
- Complex geometries and spatial adaptivity.
- Stiff numerical time-step integrators.
- Stochastic reduction analysis.
- Open source software package.

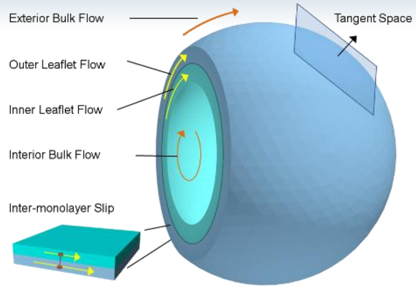
Mango-Selm package for Fluctuating Hydrodynamics
http://atzberger.org

(more on this later)

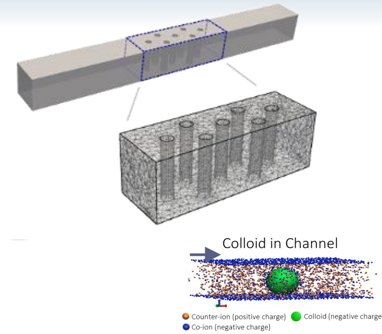
Conclusions



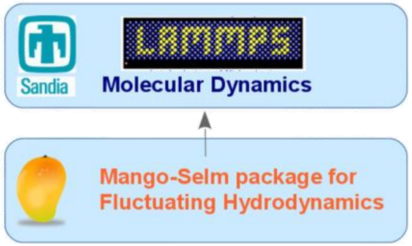
Coarse-Grained Lipid Models
Fluctuating Hydrodynamics Approaches



Continuum Mechanics of Bilayer Membranes
Fluctuating Hydrodynamics Approaches



Hybrid Descriptions for Fluidics
Fluctuating Hydrodynamics Approaches



http://mango-selm.org/

SELM Fluctuating Hydrodynamics
Software Packages

Summary

- Stochastic Eulerian Lagrangian Method (SELM) for fluctuating hydrodynamic descriptions of mesoscale systems.
- SELM incorporates into traditional hydrodynamic and CFD approaches the role of thermal fluctuations.
- Developed both coarse-grained and continuum approaches for soft-materials and fluidics.
- Many applications: polymeric fluids, colloidal systems, lipid bilayer membranes, electrokinetics, fluidics.
- Open source package in LAMMPS MD for SELM simulations: http://mango-selm.org/

Recent Students / Post-docs

- B. Gross
- J. K. Sigurdsson
- Y. Wang
- P. Plunkett
- G. Tabak
- M. Gong
- I. Sidhu

CM4 Collaborators

- C. Siefert, J. Hu, M. Parks (Sandia)
- A. Frischknecht (Sandia)
- H. Lei, G. Schenter, N. Baker (PNNL)
- N. Trask (Brown / Sandia)

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- DOE CM4
- Keck Foundation

More information: http://atzberger.org/

Publications

Hydrodynamic Coupling of Particle Inclusions Embedded in Curved Lipid Bilayer Membranes, J.K. Sigurdsson and P.J. Atzberger, (submitted), (2016) http://arxiv.org/abs/1601.06461

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More information: http://atzberger.org/