A hybrid particle-continuum method for hydrodynamics of complex fluids

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Micro- and nano-hydrodynamics

Flows of fluids (gases and liquids) through micro- (\(\mu m\)) and nano-scale (\(nm\)) structures has become technologically important, e.g., micro-fluidics, microelectromechanical systems (MEMS).

Biologically-relevant flows also occur at micro- and nano- scales.

The flows of interest often include suspended particles: colloids, polymers (e.g., DNA), blood cells, bacteria: complex fluids.

Essential distinguishing feature from “ordinary” CFD: thermal fluctuations!
Figure: From the work of David Trebotich (LLNL)
Example: Droplet Formation

Figure: From Jens Eggers, Reviews of Modern Physics, 69, 1997
I consider modeling of a polymer chain in a flowing solution, for example, DNA in a micro-array.

The detailed structure of the polymer chain is usually \textbf{coarse-grained} to a model of spherical \textbf{beads}:

- **Bead-Link**: The beads are free joints between inextensible links.
- **Bead-Spring**: Kuhn segments of the chain are point particles (beads) connected by non-linear elastic springs (FENE, worm-like, etc.)

The issue: \textbf{How to couple the polymer model with the surrounding fluid model?}
Particle/Continuum Hybrid Approach
The most direct and accurate way to simulate the interaction between the **solvent** (fluid) and **solute** (beads, chain) is to use a particle scheme for both: **Molecular Dynamics (MD)**

\[ m\ddot{r}_i = \sum_j f_{ij}(r_{ij}) \]

- Standard (time-driven) molecular dynamics:
  All of the particles are displaced *synchronously* in small *time steps* \( \Delta t \), calculating positions and forces on each particle at every time step.
- The stiff repulsion among beads demands small time steps, and chain-chain crossings are a problem.
- For hard spheres, one can use **asynchronous event-driven MD**. 
Tethered (square-well) hard-sphere chain polymers are the simplest but useful model.

Most of the computation is “wasted” on the unimportant solvent particles!

Over longer times it is hydrodynamics (local momentum and energy conservation) and fluctuations (Brownian motion) that matter.

Particle Methods

Direct Simulation Monte Carlo (DSMC)

- **Stochastic conservative collisions** of randomly chosen nearby solvent particles, as in Direct Simulation Monte Carlo (DSMC).
- Solute particles still interact with both solvent and other solute particles as hard spheres.
- Binary DSMC collisions can be replaced with **multiparticle collisions** (MPCD/SRD).

(2)

**No fluid structure**: Viscous ideal gas! [2]

In order to examine the time-scales involved, we focus on a fundamental problem: 

A single bead of size $a$ and density $\rho'$ suspended in a stationary fluid with density $\rho$ and viscosity $\eta$ (Brownian walker).

The issue: Wide separation of timescales occurs between the timescales of microscopic and macroscopic processes as the bead becomes much bigger than the mean free path $\lambda$ of the solvent particles.

Typical bead sizes are $nm$ (nano-colloids, short polymers) or $\mu m$ (colloids, DNA), while typical atomistic sizes are $0.1nm$. 
Classical picture for the following dissipation process: *Push a sphere suspended in a liquid with initial velocity* $V_{th} \approx \sqrt{kT/M}$, $M \approx \rho' a^3$, *and watch how the velocity decays:*

- **Sound waves** are generated from the sudden compression of the fluid and they take away a fraction of the kinetic energy during a **sonic time** $t_{\text{sonic}} \approx a/c$, where $c$ is the (adiabatic) sound speed.
- **Viscous dissipation** then takes over and slows the particle *non-exponentially* over a **viscous time** $t_{\text{visc}} \approx \rho a^2/\eta$, where $\eta$ is the shear viscosity.
- **Thermal fluctuations** get similarly dissipated, but their constant presence pushes the particle diffusively over a **diffusion time** $t_{\text{diff}} \approx a^2/D$, where $D \sim kT/(a\eta)$. 

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The mean collision time is $t_{coll} \approx \lambda / v_{th} \sim \eta / (\rho c^2)$, where the thermal velocity is $v_{th} \approx \sqrt{kT/m}$, for water

$$t_{coll} \sim 10^{-15} \text{s} = 1 \text{fs}$$

The sound time

$$t_{sonic} \sim \begin{cases} 1 \text{ns} & \text{for } a \sim \mu \text{m} \\ 1 \text{ps} & \text{for } a \sim \text{nm} \end{cases}, \text{ with gap } \frac{t_{sonic}}{t_{coll}} \sim \frac{a}{\lambda} \sim 10^2 - 10^5$$
Viscous time estimates

\[ t_{\text{visc}} \sim \begin{cases} 
1 \mu s & \text{for } a \sim \mu m \\
1 \text{ps} & \text{for } a \sim \text{nm} 
\end{cases}, \text{ with gap } \frac{t_{\text{visc}}}{t_{\text{sonic}}} \sim \sqrt{\frac{C}{\lambda}} \sim 1 - 10^3 \]

Finally, the diffusion time can be estimated to be

\[ t_{\text{diff}} \sim \begin{cases} 
1 \text{s} & \text{for } a \sim \mu m \\
1 \text{ns} & \text{for } a \sim \text{nm} 
\end{cases}, \text{ with gap } \frac{t_{\text{diff}}}{t_{\text{visc}}} \sim \frac{a}{\phi R} \sim 10^3 - 10^6 \]

which can now reach macroscopic timescales!
Levels of Coarse-Graining

Figure: From Pep Español, “Statistical Mechanics of Coarse-Graining”
The equations of hydrodynamics

Formally, we consider the continuum field of **conserved quantities**

\[ \mathbf{U}(r, t) = \begin{bmatrix} \rho \\ \mathbf{j} \\ e \end{bmatrix} \cong \tilde{\mathbf{U}}(r, t) = \sum_i \begin{bmatrix} m_i \\ m_i \mathbf{v}_i \\ m_i \mathbf{v}_i^2 / 2 \end{bmatrix} \delta [r - r_i(t)], \]

where the symbol \( \cong \) means that \( \mathbf{U}(r, t) \) approximates the true atomistic configuration \( \tilde{\mathbf{U}}(r, t) \) over long length and time scales.

- Due to the **microscopic conservation** of mass, momentum and energy,

\[ \partial_t \mathbf{U} = -\nabla \cdot [\mathbf{F}(\mathbf{U}) - \mathcal{Z}] = -\nabla \cdot [\mathbf{F}_H(\mathbf{U}) - \mathbf{F}_D(\nabla \mathbf{U}) - B \mathbf{W}], \]

where the flux is broken into a **hyperbolic, diffusive, and a stochastic flux**.

- Here \( \mathbf{W} \) is spatio-temporal white noise, i.e., a Gaussian random field with covariance

\[ \langle \mathbf{W}(r, t) \mathbf{W}^*(r', t') \rangle = \delta(t - t') \delta(r - r'). \]
Complete single-species fluctuating hydrodynamic equations:

\[ \mathbf{U}(r, t) = [\rho, \mathbf{j}, e]^T = [\rho, \rho\mathbf{v}, c_v\rho T + \frac{\rho v^2}{2}]^T \]

\[ \mathbf{F}_H = \begin{bmatrix} \rho\mathbf{v} \\ \rho\mathbf{v}\mathbf{v}^T + P(\rho, T) \mathbf{I} \\ (e + P)\mathbf{v} \end{bmatrix}, \quad \mathbf{F}_D = \begin{bmatrix} 0 \\ \sigma \\ \sigma \cdot \mathbf{v} + \xi \end{bmatrix}, \quad \mathbf{Z} = \begin{bmatrix} 0 \\ \Sigma \\ \Sigma \cdot \mathbf{v} + \Xi \end{bmatrix} \]

\[ \sigma = \begin{bmatrix} \eta(\nabla\mathbf{v} + \nabla\mathbf{v}^T) - \frac{\eta}{3} (\nabla \cdot \mathbf{v}) \mathbf{I} \end{bmatrix} \text{ and } \xi = \mu \nabla T \]

\[ \Sigma = \sqrt{2k_B\bar{T}} \left[ \mathbf{W}_T + \sqrt{\frac{1}{3}} \mathbf{W}_V \mathbf{I} \right] \text{ and } \Xi = \sqrt{2\bar{\mu}k_B \bar{T}^2} \mathbf{W}_S \]
Solving them numerically requires paying attention to **discrete fluctuation-dissipation balance**, in addition to the usual deterministic difficulties [3]!

It is not clear whether the Navier-Stokes equations apply at **nano-scales**.

Adding stochastic fluxes to the **non-linear** NS equations produces **ill-behaved stochastic PDEs**: At small scales one gets **negative densities** and **temperatures**.

Mathematically-rigorous limit theorems only give the **linearized fluctuations** around the nonlinear mean, which lacks important physics.

Fluctuations at scales smaller than the atomistic correlation length and time should be renormalized to account for discreteness of matter (recall *ultra-violet catastrophe*).
Consider the general linear SPDE

\[ U_t = LU + KW, \]

where the **generator** \( L \) and the **filter** \( K \) are linear operators.

Quite generally, numerical schemes use a **linear recursion** of the form

\[ U_{n+1}^j = (I + L_j \Delta t) U^n + \Delta t K_j W^n = (I + L_j \Delta t) U^n + \sqrt{\frac{\Delta t}{\Delta x}} K_j W^n \]

For analysis, convert the iteration to Fourier space.

When analyzing stochastic methods, it is natural to focus on the **second moments** (covariance).
Focus on the **discrete static spectrum**

\[ S_k = V \langle \hat{U}_k (\hat{U}_k)^* \rangle = S(k) + O(\Delta t^{p_1} k^{p_2}), \]

for a **weakly consistent** scheme.

- Recall that \( S(k) = I \) for fluctuating conservation laws.
- The remainder term quantifies the **stochastic accuracy** for large wavelengths \( (\Delta k = k\Delta x \ll 1) \) and small frequencies \( (\Delta \omega = \omega\Delta t \ll 1) \).
- A straightforward calculation \([3]\) gives

\[
\left( I + \Delta t \hat{L}_k \right) S_k \left( I + \Delta t \hat{L}_k^* \right) - S_k = -\Delta t \hat{K}_k \hat{K}_k^*. \]
For small $\Delta t$

\[ \hat{L}_k S_k^{(0)} + S_k^{(0)} \hat{L}^* = -\hat{K}_k \hat{K}^*_k, \]

and thus $S_k^{(0)} = \lim_{\Delta t \to 0} S_k = 1$ iff discrete fluctuation-dissipation balance [4] holds

\[ \hat{L}_k + \hat{L}^*_k = -\hat{K}_k \hat{K}^*_k. \]

Use the method of lines: first choose a spatial discretization consistent with the discrete fluctuation-dissipation balance condition, and then choose a temporal discretization.

In 3D, for **compressible flows**, the fluctuating velocities follow

\[
v_t = \eta \left[ \nabla^2 v + \frac{1}{3} \nabla (\nabla \cdot v) \right] + \sqrt{2} \eta \left[ (\nabla \cdot \mathbf{W}_T) + \sqrt{\frac{1}{3}} \nabla \mathbf{W}_V \right]
\]

\[
= \eta \left( \mathbf{D}_T \mathbf{G}_T + \frac{1}{3} \mathbf{G}_V \mathbf{D}_V \right) v + \sqrt{2} \eta \left( \mathbf{D}_T \mathbf{W}_T + \sqrt{\frac{1}{3}} \mathbf{G}_V \mathbf{W}_V \right).
\]

To obtain discrete fluctuation-dissipation balance, we require discrete **tensorial** divergence and gradient operators \( \mathbf{G}_T = \mathbf{D}_T^* \), and **vectorial** divergence and gradient \( \mathbf{G}_V = \mathbf{D}_V^* \).

We use the **MAC** (marker-and-cell) discretizations for the tensorial operators and the **Fortin** (corner) discretization for vectorial operators, both previously used in incompressible projection schemes.

For incompressible flows *only* the MAC discretization is required.
The **solvent** (fluid, liquid) can be modeled **implicitly** via analytical solutions (Brownian dynamics). But we want **reverse coupling** of the polymer motion on the flow (e.g., *drag reduction*)! We also need to resolve **shorter time scales** at nano systems.

Macroscopically, the coupling between flow and moving bodies/structures/beads relies on:

- **No-stick** boundary condition $v_{rel} = 0$ at the surface of the bead.
- Force on the bead is the integral of the stress tensor over the bead surface.

The above two conditions are **questionable at nanoscales**, but even worse, they are very hard to implement numerically in an efficient and stable manner, even in the (phenomenological) Lattice-Boltzmann method.
The coupling between the solute and solvent is phenomenological and approximate for most methods in use:

$$md\dot{v} = [F(R) - \gamma v] dt + \sqrt{2\gamma kT} dW$$

- Point beads with artificial friction coefficients $\gamma \approx 6\pi a\eta$ based on asymptotic Stokes law
- Point beads exerting (smeared) $\delta$-function forces on the fluid
- Uncorrelated fluctuating forces on the beads

Such a Langevin equation is physically inconsistent, except at (unrealistic?) asymptotic time-scales!

One can improve on this by giving the beads a physical size (not a point!) and consistently including thermal fluctuations in the fluid equations (see LB and SIB methods).
Split the domain into a particle and a continuum (hydro) subdomains, with timesteps $\Delta t_H = K \Delta t_P$.

Hydro solver is a simple explicit (fluctuating) compressible LLNS code and is not aware of particle patch.

The method is based on Adaptive Mesh and Algorithm Refinement (AMAR) methodology for conservation laws and ensures strict conservation of mass, momentum, and energy [5, 6].

**Instantaneous** (fluctuating) flow

**Mean** (plug) flow
Continuum-Particle Coupling

- Each macro (hydro) cell is either particle or continuum. There is also a reservoir region surrounding the particle subdomain.

- The coupling is roughly of the state-flux form:
  - The continuum solver provides state boundary conditions for the particle subdomain via reservoir particles.
  - The particle subdomain provides flux boundary conditions for the continuum subdomain.

- The fluctuating hydro solver is oblivious to the particle region: Any conservative explicit finite-volume scheme can trivially be substituted.

- The coupling is greatly simplified because the particle fluid is ideal (no internal structure): No overlap region.

"A hybrid particle-continuum method for hydrodynamics of complex fluids”
Steps of the coupling algorithm [7]:

1. The hydro solution is computed everywhere, including the particle patch, giving an estimated total flux $\Phi_H$.

2. **Reservoir particles** are inserted at the boundary of the particle patch based on Chapman-Enskog distribution from kinetic theory, accounting for both collisional and kinetic viscosities.

3. Reservoir particles are propagated by $\Delta t$ and collisions are processed (including virtual particles!), giving the total particle flux $\Phi_p$.

4. The hydro solution is overwritten in the particle patch based on the particle state $u_p$.

5. The hydro solution is corrected based on the more accurate flux, $u_H \leftarrow u_H - \Phi_H + \Phi_p$. 

Back to the Brownian Bead

MNG
We investigate the **velocity autocorrelation function** (VACF) for a Brownian bead

\[ C(t) = \langle v(t_0) \cdot v(t_0 + t) \rangle \]

- From equipartition theorem, \( C(0) = kT / M \).
- For a **neutrally-boyant** particle, \( \rho' = \rho \), incompressible hydrodynamic theory gives \( C(0) = 2kT / 3M \) because the momentum correlations decay instantly due to sound waves.
- Hydrodynamic persistence (conservation) gives a **long-time power-law tail** \( C(t) \sim (kT / M)(t / t_{\text{visc}})^{-3/2} \) not reproduced in Brownian dynamics.
Small Bead (~10 particles)

\[ \frac{M C(t)}{k_B T} \]

- Stoch. hybrid (L=1)
- Det. hybrid (L=1)
- Stoch. hybrid (L=2)
- Det. hybrid (L=2)
- Particle (L=1)
- Theory

\[ t / t_{\text{visc}} \]

\[ t_{L=1} \]
Large Bead (~1000 particles)

Results

Brownian bead VACF

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The adiabatic piston problem

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Figure: Massive rigid piston ($M/m = 4000$) not in mechanical equilibrium.
Figure: The VACF for a rigid piston of mass $M/m = 1000$ at thermal equilibrium.
Conclusions

- **Coarse-grained particle methods** can be used to accelerate hydrodynamic calculations at small scales.

- Designing numerical methods for fluctuating hydrodynamics requires attention to **fluctuation-dissipation balance**, in addition to the usual (deterministic) stability and accuracy considerations.

- Hybrid particle continuum methods closely reproduce purely particle simulations at a **fraction of the cost**.

- It is **necessary to include fluctuations** in the continuum subdomain in hybrid methods.
Future Directions

- Numerical schemes for **Low-Mach Number** fluctuating hydrodynamics.
- Theoretical work on the **equations of fluctuating hydrodynamics**: systematic coarse-graining and approximations.
- **Direct coupling** between fluctuating hydrodynamics and microstructure (solute beads).
- **Test, validate, and apply** the methodology for polymer problems.
- Couple our **non-ideal stochastic hard-sphere gas** to continuum hydrodynamics with **microscopic fidelity**.
- Ultimately we require an **Adaptive Mesh and Algorithm Refinement** (AMAR) framework that couples deterministic MD for the polymer chains (**micro**), a stochastic solvent (**micro-meso**), with compressible fluctuating Navier-Stokes (**meso**), and incompressible CFD (**macro**).
Stochastic Event-Driven Molecular Dynamics.

A Thermodynamically-Consistent Non-Ideal Stochastic Hard-Sphere Fluid.

On the Accuracy of Explicit Finite-Volume Schemes for Fluctuating Hydrodynamics.

P. J. Atzberger.
Preprint.

Adaptive Mesh and Algorithm Refinement using Direct Simulation Monte Carlo.

S. A. Williams, J. B. Bell, and A. L. Garcia.
Algorithm Refinement for Fluctuating Hydrodynamics.

A hybrid particle-continuum method for hydrodynamics of complex fluids.