A Hybrid Particle-Continuum Method Coupling a Fluctuating Fluid with Suspended Structures

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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.

An important feature of small-scale flows, not discussed here, is surface/boundary effects (e.g., slip in the contact line problem).

Essential distinguishing feature from “ordinary” CFD: thermal fluctuations!

I focus here not on the technical details of hybrid methods, but rather, on using our method to demonstrate the general conclusion that fluctuations should be taken into account at the continuum level.
Example: DNA Filtering

*Fu et al., Nature Nanotechnology* 2 (2007)  

How to coarse grain the fluid (solvent) and couple it to the suspended microstructure (e.g., polymer chain)?
Levels of Coarse-Graining

Figure: From Pep Español, “Statistical Mechanics of Coarse-Graining”
Figure: Hybrid method for a polymer chain.
Particle Methods for Complex Fluids

- 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)**

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

- The stiff repulsion among beads demands small time steps, and chain-chain crossings are a problem.
- 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.
- We need to coarse grain the fluid model further: **Replace deterministic interactions with stochastic collisions**.
Stochastic conservative collisions of randomly chosen nearby solvent particles, as in DSMC (also related to MPCD/SRD and DPD).

Solute particles still interact with both solvent and other solute particles as hard or soft spheres.

No fluid structure: Viscous ideal gas.

One can introduce biased collision models to give the fluids consistent structure and a non-ideal equation of state. [1].

(MNG)
Tethered polymer chain in shear flow.
Formally, we consider the continuum field of \textit{conserved quantities}

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

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

Formal coarse-graining of the microscopic dynamics has been performed to derive an \textit{approximate closure} for the macroscopic dynamics [2].

This leads to \textit{SPDEs of Langevin type} formed by postulating a \textit{white-noise random flux} term in the usual Navier-Stokes-Fourier equations with magnitude determined from the \textit{fluctuation-dissipation balance} condition, following Landau and Lifshitz.
Compressible Fluctuating Hydrodynamics

\[ D_t \rho = - \rho \nabla \cdot \mathbf{v} \]
\[ \rho \left( D_t \mathbf{v} \right) = - \nabla P + \nabla \cdot \left( \eta \nabla \mathbf{v} + \Sigma \right) \]
\[ \rho c_p \left( D_t T \right) = D_t P + \nabla \cdot \left( \mu \nabla T + \Xi \right) + \left( \eta \nabla \mathbf{v} + \Sigma \right) : \nabla \mathbf{v}, \]

where the variables are the density \( \rho \), velocity \( \mathbf{v} \), and temperature \( T \) fields,

\[ D_t \Box = \partial_t \Box + \mathbf{v} \cdot \nabla (\Box) \]
\[ \nabla \mathbf{v} = (\nabla \mathbf{v} + \nabla \mathbf{v}^T) - 2 (\nabla \cdot \mathbf{v}) \mathbf{I} / 3 \]

and capital Greek letters denote stochastic fluxes:

\[ \Sigma = \sqrt{2 \eta k_B T} \mathbf{W}. \]
\[ \langle \mathcal{W}_{ij}(\mathbf{r}, t) \mathcal{W}_{kl}^*(\mathbf{r}', t') \rangle = (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} - 2 \delta_{ij} \delta_{kl} / 3) \delta(t - t') \delta(\mathbf{r} - \mathbf{r}'). \]
The non-linear LLNS equations are ill-behaved stochastic PDEs, and we do not really know how to interpret the nonlinearities precisely.

Finite-volume discretizations naturally impose a grid-scale regularization (smoothing) of the stochastic forcing.

A renormalization of the transport coefficients is also necessary [3].

We have algorithms and codes to solve the compressible equations (collocated and staggered grid), and recently also the incompressible ones (staggered grid) [4, 5].

Solving the LLNS equations numerically requires paying attention to discrete fluctuation-dissipation balance, in addition to the usual deterministic difficulties [4].
Fluctuating Hydrodynamics

Finite-Volume Schemes

\[ c_t = -\mathbf{v} \cdot \nabla c + \chi \nabla^2 c + \nabla \cdot \left( \sqrt{2\chi} \mathcal{W} \right) = \nabla \cdot \left[ -c\mathbf{v} + \chi \nabla c + \sqrt{2\chi} \mathcal{W} \right] \]

- **Generic finite-volume spatial discretization**

\[ c_t = D \left[ (-\mathbf{V}c + Gc) + \sqrt{2\chi/ (\Delta t \Delta V)} \mathcal{W} \right], \]

where \( D : \text{faces} \rightarrow \text{cells} \) is a **conservative** discrete divergence, \( G : \text{cells} \rightarrow \text{faces} \) is a discrete gradient.

- **Here** \( \mathcal{W} \) is a collection of random normal numbers representing the (face-centered) stochastic fluxes.

- **The divergence and gradient** should be **duals**, \( D^* = -G \).

- **Advection** should be **skew-adjoint** (non-dissipative) if \( \nabla \cdot \mathbf{v} = 0 \),

\[ (\mathbf{D}\mathbf{V})^* = - (\mathbf{D}\mathbf{V}) \text{ if } (\mathbf{D}\mathbf{V}) \mathbf{1} = 0. \]
Figure: Equilibrium discrete spectra (static structure factors) $S_{\rho,\rho}(k) \sim \langle \hat{\rho}\hat{\rho}^* \rangle$ (should be unity for all discrete wavenumbers) and $S_{\rho,v}(k) \sim \langle \hat{\rho}\hat{v}_x^* \rangle$ (should be zero) for our RK3 collocated scheme.
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.
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 ideal particle fluid has no internal structure.

Our Hybrid Algorithm

1. The hydro solution $u_H$ 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, 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, $u_H \leftarrow u_H - \Phi_H + \Phi_p$. 
For molecular dynamics (non-ideal particle fluids) the insertion of reservoir particles is greatly complicated by the need to account for the internal structure of the fluid and requires an overlap region.

A hybrid method based on a flux-flux coupling between molecular dynamics and isothermal compressible fluctuating hydrodynamics has been developed by Coveney, De Fabritiis, Delgado-Buscalioni and co-workers [6].

Some comparisons between different forms of coupling (state-state, state-flux, flux-state, flux-flux) has been performed by Ren [7].

Reaching relevant time scales ultimately requires a stochastic immersed structure approach coupling immersed structures directly to a fluctuating solver (work in progress).
Thermal fluctuations push a sphere of size $a$ and density $\rho'$ suspended in a stationary fluid with density $\rho$ and viscosity $\eta$ (Brownian walker) with initial velocity $V_{th} \approx \sqrt{kT/M}$, $M \approx \rho'a^3$.

The classical picture of Brownian motion indicates three widely-separated timescales:

- **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 $t_{sonic} \approx a/c$, where $c$ is the (adiabatic) sound speed.
- **Viscous dissipation** then takes over and slows the particle non-exponentially over a viscous time $t_{visc} \approx \rho a^2/\eta$, where $\eta$ is the shear viscosity.
- **Thermal fluctuations** get similarly dissipated, but their constant presence pushes the particle diffusively over a diffusion time $t_{diff} \approx a^2/D$, where $D \sim kT/(\alpha\eta)$. 

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

\[ C(t) = 2d^{-1} \langle v(t_0) \cdot v(t_0 + t) \rangle \]

From equipartition theorem \( C(0) = \frac{k_B T}{M} \).

For a **neutrally-boyant** particle, \( \rho' = \rho \), incompressible hydrodynamic theory gives \( C(0) = \frac{2k_B T}{3M} \) because one third of the kinetic energy decays at the sound time scale.

Hydrodynamic persistence (conservation) gives a **long-time power-law tail** \( C(t) \sim \left( \frac{k_B T}{M} \right) \left( \frac{t}{t_{visc}} \right)^{-3/2} \) that can be quantified using fluctuating hydrodynamics.

The diffusion coefficient is the **integral of the VACF** and is strongly-affected by the tail.
The adiabatic piston problem

MNG
The Importance of Thermal Fluctuations

Relaxation Toward Equilibrium

Figure: Massive rigid piston ($M/m = 4000$) not in mechanical equilibrium: The deterministic hybrid gives the wrong answer!
The Importance of Thermal Fluctuations

Adiabatic Piston

VACF for Piston

\[ C(t) \]

Figure: The VACF for a rigid piston of mass \( M/m = 1000 \) at thermal equilibrium: Increasing the width of the particle region does not help: One must include the thermal fluctuations in the continuum solver!
When macroscopic gradients are present, steady-state thermal fluctuations become long-range correlated.

Consider a binary mixture of fluids and consider concentration fluctuations around a steady state $c_0(r)$:

$$c(r, t) = c_0(r) + \delta c(r, t)$$

The concentration fluctuations are advected by the random velocities $v(r, t) = \delta v(r, t)$, approximately:

$$\partial_t (\delta c) + (\delta v) \cdot \nabla c_0 = \chi \nabla^2 (\delta c) + \sqrt{2\chi k_B T} (\nabla \cdot \mathcal{W}_c)$$

The velocity fluctuations drive and amplify the concentration fluctuations leading to so-called giant fluctuations [8].
Figure: Snapshots of concentration in a miscible mixture showing the development of a rough diffusive interface between two miscible fluids in zero gravity [3, 8, 5].
Figure: Experimental results by A. Vailati et al. from a microgravity environment [8] showing the enhancement of concentration fluctuations in space (box scale is macroscopic: 5mm on the side, 1mm thick).
The **nonlinear** concentration equation includes a contribution to the mass flux due to **advection by the fluctuating velocities**, 

\[
\partial_t (\delta c) + (\delta \mathbf{v}) \cdot \nabla c_0 = \nabla \cdot \left[ - (\delta c)(\delta \mathbf{v}) + \chi \nabla (\delta c) \right] + \ldots
\]

Simple (quasi-linear) perturbative theory suggests that concentration and velocity fluctuations become correlated and

\[-\langle (\delta c)(\delta \mathbf{v}) \rangle \approx (\Delta \chi) \nabla c_0.\]

The **fluctuation-renormalized diffusion coefficient** is \(\chi + \Delta \chi\) (think of **eddy diffusivity** in turbulent transport).

Because fluctuations are affected by boundaries, \(\Delta \chi\) is **system-size dependent**.
Consider the effective diffusion coefficient in a system of dimensions $L_x \times L_y \times L_z$ with a concentration gradient imposed along the $y$ axis.

In **two dimensions**, $L_z \ll L_x \ll L_y$, linearized fluctuating hydrodynamics predicts a **logarithmic divergence**

$$\chi_{\text{eff}}^{(2D)} \approx \chi + \frac{k_B T}{4\pi \rho (\chi + \nu) L_z \ln \frac{L_x}{L_0}}$$

In **three dimensions**, $L_x = L_z = L \ll L_y$, $\chi_{\text{eff}}$ converges as $L \to \infty$ to the **macroscopic diffusion coefficient**,

$$\chi_{\text{eff}}^{(3D)} \approx \chi + \frac{\alpha k_B T}{\rho (\chi + \nu)} \left( \frac{1}{L_0} - \frac{1}{L} \right)$$

We have verified these predictions using particle (DSMC) simulations at hydrodynamic scales [3].
Particle Simulations

Figure: Divergence of diffusion coefficient in two dimensions.

Kinetic theory
\( \chi_{\text{eff}} \) (System A)
\( \chi_0 \) (System A)
\( \chi_{\text{eff}} \) (System B)
\( \chi_0 \) (System B)
\( \chi_{\text{eff}} \) (SPDE, A)
Theory \( \chi_0 \) (A)
Theory \( \chi_0 \) (B)
Theory \( \chi_{\text{eff}} \)
Instead of an ill-defined “molecular” or “bare” diffusivity, one should define a **locally renormalized diffusion coefficient** $\chi_0$ that depends on the length-scale of observation.

This coefficient accounts for the arbitrary division between continuum and particle levels inherent to fluctuating hydrodynamics.

A deterministic continuum limit does not exist in two dimensions, and is not applicable to small-scale finite systems in three dimensions.

**Fluctuating hydrodynamics** is applicable at a broad range of scales if the transport coefficient are renormalized based on the cutoff scale for the random forcing terms.
Coarse-grained particle methods can be used to accelerate hydrodynamic calculations at small scales.

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

It is necessary to include fluctuations in the continuum solver in hybrid methods.

Thermal fluctuations affect the macroscopic transport in fluids.
Future Directions

- Improve and implement stochastic particle methods (parallelize, add chemistry, analyze theoretically).
- Direct fluid-structure coupling between fluctuating hydrodynamics and microstructure.
- Develop numerical schemes for Low-Mach Number fluctuating hydrodynamics.
- Ultimately we require an Adaptive Mesh and Algorithm Refinement (AMAR) framework that couples a particle model (micro), with compressible fluctuating Navier-Stokes (meso), and incompressible or low Mach solver (macro).
Stochastic Hard-Sphere Dynamics for Hydrodynamics of Non-Ideal Fluids.

P. Español.
Stochastic differential equations for non-linear hydrodynamics.

Diffusive Transport Enhanced by Thermal Velocity Fluctuations.

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

Staggered Schemes for Incompressible Fluctuating Hydrodynamics.
Submitted, 2011.

G. De Fabritiis, M. Serrano, R. Delgado-Buscalioni, and P. V. Coveney.
Fluctuating hydrodynamic modeling of fluids at the nanoscale.

W. Ren.
Analytical and numerical study of coupled atomistic-continuum methods for fluids.

Fractal fronts of diffusion in microgravity.