A Hybrid Particle-Continuum Approach to Hydrodynamics at Small Scales

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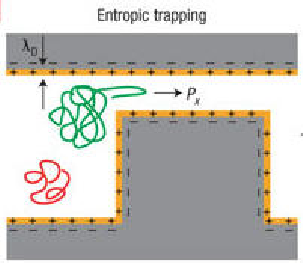
Outline

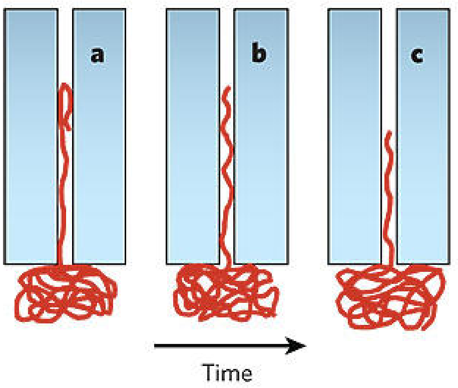
Micro- and nano-hydrodynamics

- Flows of fluids (gases and liquids) through micro- (μm) 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**!

Example: DNA Filtering

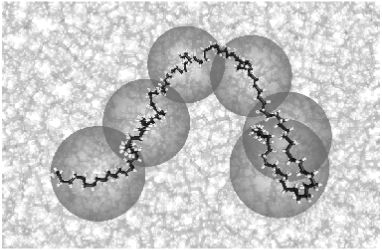


Fu et al., Nature Nanotechnology 2 (2007)



H. Craighead, Nature 442 (2006)

Polymer chains



Johan Padding, Cambridge

- 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 **coarse-grained** to a model of spherical **beads**.
- E.g., Kuhn segments of the chain are represented as **spherical beads** connected by non-linear elastic springs (FENE, worm-like, etc.)

The issue: **How to coarse grain the fluid (solvent) and couple it to the suspended structures?**

Our approach: Particle/Continuum Hybrid

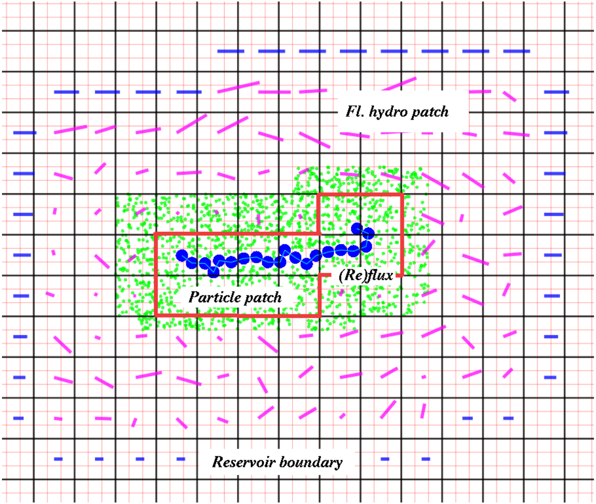


Figure: Hybrid method for a polymer chain.

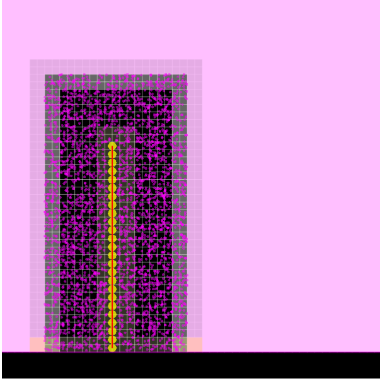
Particle Methods for Complex Fluids

- 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)**

$$m\ddot{\mathbf{r}}_i = \sum_j \mathbf{f}_{ij}(\mathbf{r}_{ij})$$

- 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 ones.*

Direct Simulation Monte Carlo (DSMC)



(MNG)

Tethered polymer chain in shear flow [1].

- **Stochastic conservative collisions** of randomly chosen nearby solvent particles, as in DSMC (also related to MPCD/SRD).
- Solute particles still interact with **both** solvent and other solute particles as hard or soft spheres [2].
- **No fluid structure**: Viscous ideal gas.
- One can introduce biased collision models to give the fluids consisten structure and a **non-ideal equation of state**. [3, 4].

The Need for Coarse-Graining

- In order to examine the time-scales involved, we focus on a fundamental problem:
  A single bead of size a and density ρ' *suspended in a stationary fluid with density* ρ and viscosity η (**Brownian walker**).
- By increasing the size of the bead obviously the **number of solvent particles** increases as $N \sim a^3$. But this is not the biggest problem (we have large supercomputers).
- The real issue is that a wide **separation of timescales occurs**: The *gap between the timescales of microscopic and macroscopic* processes widens as the bead becomes much bigger than the solvent particles (water molecules).
- Typical bead sizes are *nm* (nano-colloids, short polymers) or μm (colloids, DNA), while typical atomistic sizes are $1\text{Å} = 0.1 nm$.

Brownian Bead

- 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** $t_{sonic} \approx a/c$, 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 η is the shear viscosity. Note that the classical **Langevin time** scale $t_{Lang} \approx m/\eta a$ applies only to unrealistically dense beads!
  - **Thermal fluctuations** get similarly dissipated, but their constant presence pushes the particle diffusively over a **diffusion time** $t_{diff} \approx a^2/D$, where $D \sim kT/(a\eta)$.

Timescale Estimates

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

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

- The **sound time**

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

Estimates contd...

- **Viscous time** estimates

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

- Finally, the **diffusion time** can be estimated to be

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

which can now reach **macroscopic timescales**!

Levels of Coarse-Graining

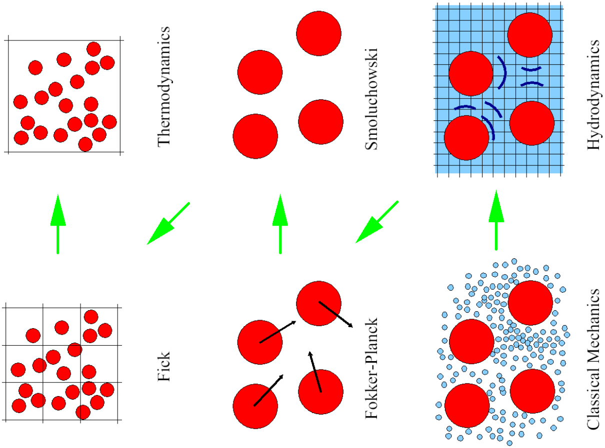


Figure: From Pep Español, "Statistical Mechanics of Coarse-Graining"

Continuum Models of Fluid Dynamics

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

$$\mathbf{U}(\mathbf{r},t)=\begin{bmatrix}\rho\\ \mathbf{j}\\ e\end{bmatrix}\cong\widetilde{\mathbf{U}}(\mathbf{r},t)=\sum_i\begin{bmatrix}m_i\\ m_i\boldsymbol{\upsilon}_i\\ m_i\upsilon_i^2/2\end{bmatrix}\delta\left[\mathbf{r}-\mathbf{r}_i(t)\right],$$

  where the symbol $\cong$ means that $\mathbf{U}(\mathbf{r},t)$ approximates the true atomistic configuration $\widetilde{\mathbf{U}}(\mathbf{r},t)$ over **long length and time scales**.
- Formal coarse-graining of the microscopic dynamics has been performed to derive an **approximate closure** for the macroscopic dynamics [5].
- This leads to **SPDEs of Langevin type** formed by postulating a random flux term in the usual Navier-Stokes-Fourier equations with magnitude determined from the **fluctuation-dissipation balance** condition, following Landau and Lifshitz.

The SPDEs of Fluctuating Hydrodynamics

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

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

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

- Here $\mathcal{W}$ is spatio-temporal **white noise**, i.e., a Gaussian random field with covariance

$$\langle \mathcal{W}_i(\mathbf{r}, t) \mathcal{W}_j^\star(\mathbf{r}, t') \rangle = (\delta_{ij})\, \delta(t - t') \delta(\mathbf{r} - \mathbf{r}').$$

- Adding stochastic fluxes to the **non-linear** NS equations produces **ill-behaved stochastic PDEs** (solution is too irregular), but we will ignore that for now...

Compressible Fluctuating Hydrodynamics

$$
\begin{aligned}
D_t\rho &= -\rho\boldsymbol{\nabla}\cdot\mathbf{v} \\
\rho\left(D_t\mathbf{v}\right) &= -\boldsymbol{\nabla}P + \boldsymbol{\nabla}\cdot\left(\eta\overline{\boldsymbol{\nabla}}\mathbf{v} + \boldsymbol{\Sigma}\right) \\
\rho c_p\left(D_t T\right) &= D_t P + \boldsymbol{\nabla}\cdot\left(\mu\boldsymbol{\nabla}T + \boldsymbol{\Xi}\right) + \left(\eta\overline{\boldsymbol{\nabla}}\mathbf{v} + \boldsymbol{\Sigma}\right):\boldsymbol{\nabla}\mathbf{v},
\end{aligned}
$$

where the variables are the **density** ρ, **velocity** $\mathbf{v}$, and **temperature** T fields,

$$
\begin{aligned}
D_t\square &= \partial_t\square + \mathbf{v}\cdot\boldsymbol{\nabla}\left(\square\right) \\
\overline{\boldsymbol{\nabla}}\mathbf{v} &= \left(\boldsymbol{\nabla}\mathbf{v} + \boldsymbol{\nabla}\mathbf{v}^T\right) - 2\left(\boldsymbol{\nabla}\cdot\mathbf{v}\right)\mathbf{I}/3
\end{aligned}
$$

and capital Greek letters denote stochastic fluxes:

$$
\boldsymbol{\Sigma} = \sqrt{2\eta k_B T}\,\mathcal{W}.
$$

$$
\langle\mathcal{W}_{ij}(\mathbf{r},t)\mathcal{W}^{\star}_{kl}(\mathbf{r}',t')\rangle = \left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - 2\delta_{ij}\delta_{kl}/3\right)\delta(t-t')\delta(\mathbf{r}-\mathbf{r}')
$$

Incompressible Fluctuating Navier-Stokes

- Ignoring density and temperature fluctuations, we obtain the **incompressible approximation**:

$$\rho D_t \mathbf{v} = \eta \boldsymbol{\nabla}^2 \mathbf{v} - \boldsymbol{\nabla} \pi + \sqrt{2\eta k_B T} \left(\boldsymbol{\nabla} \cdot \boldsymbol{\mathcal{W}} \right),$$
$$\boldsymbol{\nabla} \cdot \mathbf{v} = 0$$

  where the stochastic stress tensor $\boldsymbol{\mathcal{W}}$ is a white-noise random Gaussian tensor field with covariance

$$\langle \mathcal{W}_{ij}(\mathbf{r}, t) \mathcal{W}^{\star}_{kl}(\mathbf{r}', t') \rangle = \left(\delta_{ik} \delta_{jl} \right) \delta(t - t') \delta(\mathbf{r} - \mathbf{r}').$$

- We have algorithms and codes to solve the compressible equations, and we are now working on the incompressible ones.
- Solving them numerically requires paying attention to **discrete fluctuation-dissipation balance**, in addition to the usual deterministic difficulties [6].

Solute-Solvent Coupling using Particles

- 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.

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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*", A. Donev and J. B. Bell and A. L. Garcia and B. J. Alder, **SIAM J. Multiscale Modeling and Simulation 8(3):871-911, 2010**

Hybrid Algorithm

Steps of the coupling algorithm [7]:

1. The hydro solution is computed everywhere, including the **particle patch**, giving an estimated total flux $\boldsymbol{\Phi}_H$.
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 Δt and *collisions* are processed (including virtual particles!), giving the total particle flux $\boldsymbol{\Phi}_p$.
4. The hydro solution is overwritten in the particle patch based on the particle state $\mathbf{u}_p$.
5. The hydro solution is corrected based on the more accurate flux, $\mathbf{u}_H \leftarrow \mathbf{u}_H - \boldsymbol{\Phi}_H + \boldsymbol{\Phi}_p$.

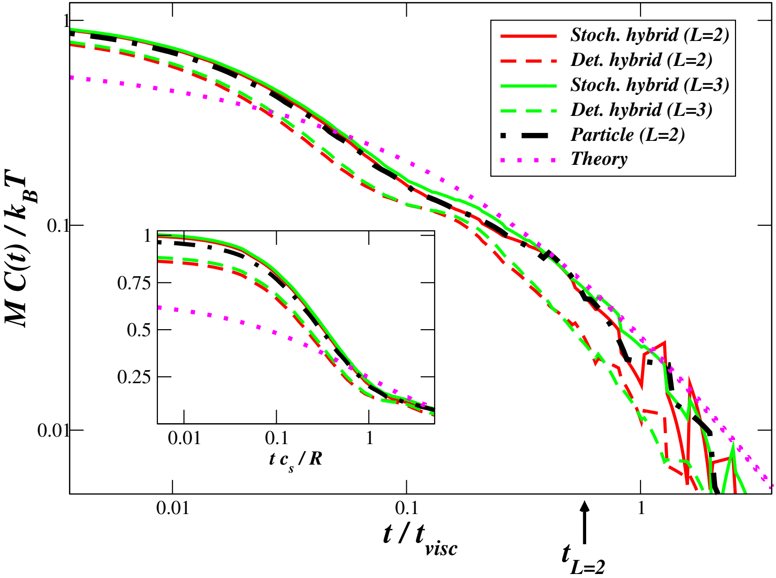
Velocity Autocorrelation Function

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

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

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

VACF


Stoch. hybrid (L=2)
Det. hybrid (L=2)
Stoch. hybrid (L=3)
Det. hybrid (L=3)
Particle (L=2)
Theory
$M\,C(t) / k_B T$
1
0.1
0.01
0.01
0.1
1
t / t_{visc}
$t_{L=2}$
0.75
0.5
0.25
$t\,c_s / R$

The adiabatic piston problem

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Relaxation Toward Equilibrium

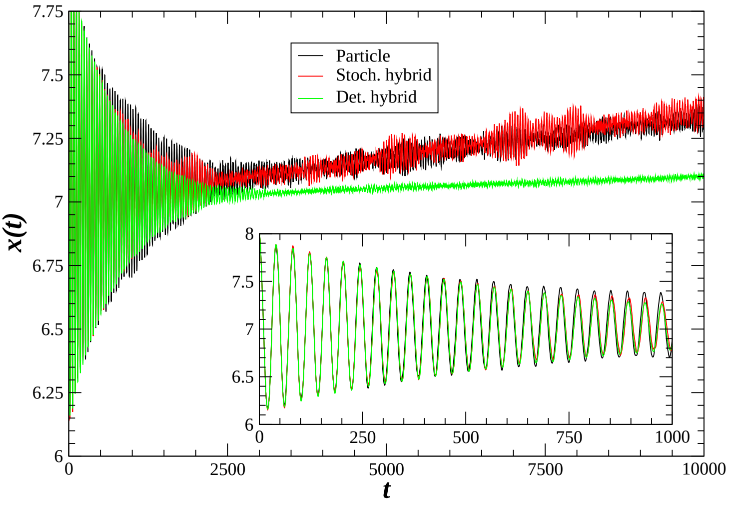


Figure: Massive rigid piston ($M/m = 4000$) not in mechanical equilibrium: **The deterministic hybrid gives the wrong answer!**

VACF for Piston

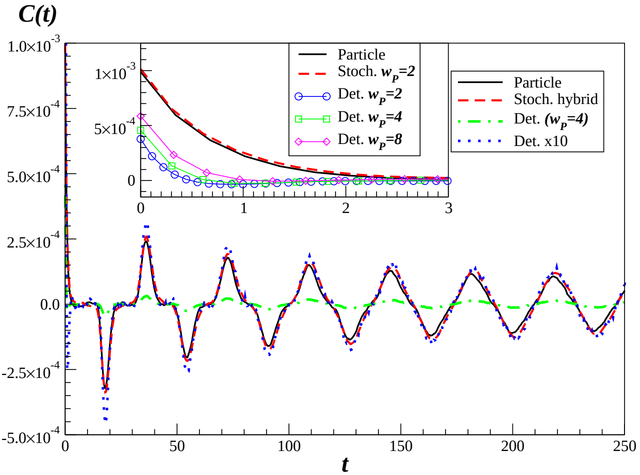


Figure: The VACF for a rigid piston of mas $M/m = 1000$ at thermal equilibrium: **Increasing the width of the particle region does not help: One must include the thermal fluctuations in the continuum solver**!

Fluctuations in the presence of gradients

- At **equilibrium**, hydrodynamic fluctuations have non-trivial temporal correlations, but there are no spatial correlations between any variables.
- When macroscopic gradients are present, however, **long-ranged correlated fluctuations** appear.
- Consider a **binary mixture** of fluids and consider **concentration fluctuations** around a steady state $c_0(\mathbf{r})$:

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

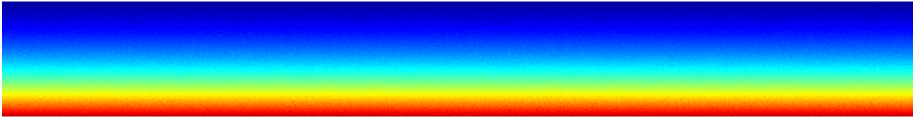
- The concentration fluctuations are **advected by the random velocities** $\mathbf{v}(\mathbf{r}, t)$, approximately:

$$(\delta c)_t + \mathbf{v} \cdot \boldsymbol{\nabla} c_0 = D \boldsymbol{\nabla}^2 (\delta c) + \sqrt{2Dk_B T} \left(\boldsymbol{\nabla} \cdot \boldsymbol{\mathcal{W}}_c \right)$$

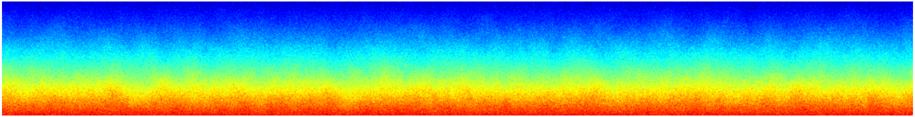
- The velocity fluctuations drive and amplify the concentration fluctuations leading to so-called **giant fluctuations**.

Equilibrium versus Non-Equilibrium

Results obtained using our fluctuating continuum compressible solver.



Concentration for a mixture of two (heavier red and lighter blue) fluids at **equilibrium**, in the presence of gravity.



No gravity but a similar **non-equilibrium** concentration gradient is imposed via the boundary conditions.

Giant Fluctuations during diffusive mixing

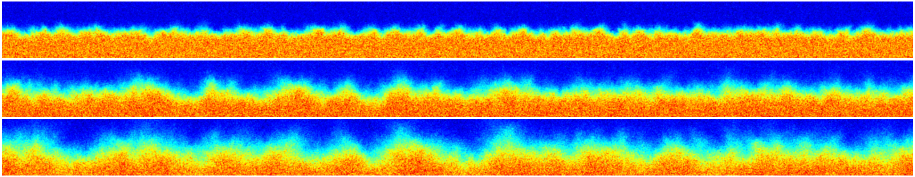


Figure: Snapshots of the concentration during the diffusive mixing of two fluids (red and blue) at $t = 1$ (top), $t = 4$ (middle), and $t = 10$ (bottom), starting from a flat interface (phase-separated system) at $t = 0$.

Giant Fluctuations in Experiments

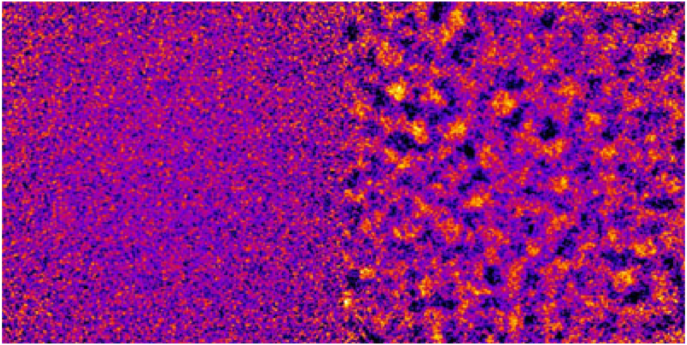


Figure: Experimental snapshots of the steady-state concentration fluctuations in a solution of polystyrene in water with a strong concentration gradient imposed via a stabilizing temperature gradient, in Earth gravity (left), and in microgravity (right) [private correspondence with Roberto Cerbino]. The strong enhancement of the fluctuations in microgravity is evident.

Fluctuation-Enhanced Diffusion Coefficient

- We study the following simple **model steady-state system**: *A quasi-two dimensional mixture of identical but* labeled *(as components 1 and 2) fluids is enclosed in a box of lengths* $L_x \times L_y \times L_z$*, where* $L_z \ll L_{x/y}$*. Periodic boundary conditions are applied in the x (horizontal) and z (depth) directions, and impermeable constant-temperature walls are placed at the top and bottom boundaries. A weak constant concentration gradient* $\boldsymbol{\nabla} c_0 = g_c \hat{\mathbf{y}}$ *is imposed along the y axes by enforcing constant concentration boundary conditions at the top and bottom walls.*
- Incompressible (isothermal) linearized fluctuating hydrodynamics is given by:

$$(\delta c)_t + \mathbf{v} \cdot \boldsymbol{\nabla} c_0 = -D \boldsymbol{\nabla}^2 (\delta c) + \sqrt{2Dk_B T} \left(\boldsymbol{\nabla} \cdot \boldsymbol{\mathcal{W}}_c \right)$$

$$\rho \mathbf{v}_t = \eta \boldsymbol{\nabla}^2 \mathbf{v} - \boldsymbol{\nabla} \pi + \sqrt{2\eta k_B T} \left(\boldsymbol{\nabla} \cdot \boldsymbol{\mathcal{W}} \right) \text{ and } \boldsymbol{\nabla} \cdot \mathbf{v} = 0$$

Fluctuation-Enhanced Diffusion Coefficient

- Solve in Fourier space to obtain the correlations (**static structure factors**) between velocity and concentration fluctuations:

$$\widehat{S}_{c,v_y}(\mathbf{k}) = \langle (\widehat{\delta c})(\widehat{v}_y^\star) \rangle \sim -\left(k_\perp^2 k^{-4}\right) g_c,$$

which are seen to **diverge at small wavenumbers** k.

- The nonlinear concentration equation includes a contribution to the mass flux due to **advection by the fluctuating velocities**,

$$\partial_t (\delta c) + \rho_0 \mathbf{v} \cdot \boldsymbol{\nabla} c_0 = \boldsymbol{\nabla} \cdot (\mathbf{j} + \boldsymbol{\Psi}) = \boldsymbol{\nabla} \cdot [D_0 \boldsymbol{\nabla} (\delta c) - \rho_0 (\delta c) \mathbf{v}] + \boldsymbol{\nabla} \cdot \boldsymbol{\Psi},$$

where we have denoted the so-called **bare diffusion coefficient** with D_0.

- To leading order, the **renormalized diffusion coefficient** includes a **fluctuation enhancement** ΔD due to thermal velocity fluctuations,

$$\langle \mathbf{j} \rangle \approx (D_0 + \Delta D) \boldsymbol{\nabla} c_0 = \left[D_0 - (2\pi)^{-3} \int_{\mathbf{k}} \widehat{S}_{c,v_y}(\mathbf{k})\, d\mathbf{k} \right] \boldsymbol{\nabla} c_0.$$

Fluctuation-Enhanced Diffusion Coefficient

- The **effective transport** coefficient $D_{eff} = D_0 + \Delta D$ **depends on the** small wavenumber cutoff $k_{min} \sim 2\pi/L$, where L is the **system size**.
- For our quasi two-dimensional model, assuming $L_x \ll L_y$, one obtains [8] a logarithmic growth of the fluctuation-renormalized diffusion coefficient

$$\Delta D \approx k_B T \left[4\pi\rho(\chi_0 + \nu)L_z\right]^{-1} \ln L_x.$$

- This can be **tested in particle simulations** by calculating the mass current of the first fluid component:

$$\langle j_y \rangle = \langle \rho_1 v_{1,y} \rangle = \langle \rho_1 \rangle \langle v_{1,y} \rangle + \langle (\delta\rho_1)(\delta v_{1,y}) \rangle,$$

  defining a splitting of the total mass transfer into a **diffusive or bare** and an **advective or fluctuation** piece:

$$\langle \rho_1 v_{1,y} \rangle = D_{eff} \left(\boldsymbol{\nabla}_y c_0 \right)$$
$$\langle \rho_1 \rangle \langle v_{1,y} \rangle = D_0 \left(\boldsymbol{\nabla} c \right)$$

Particle Results (2D)

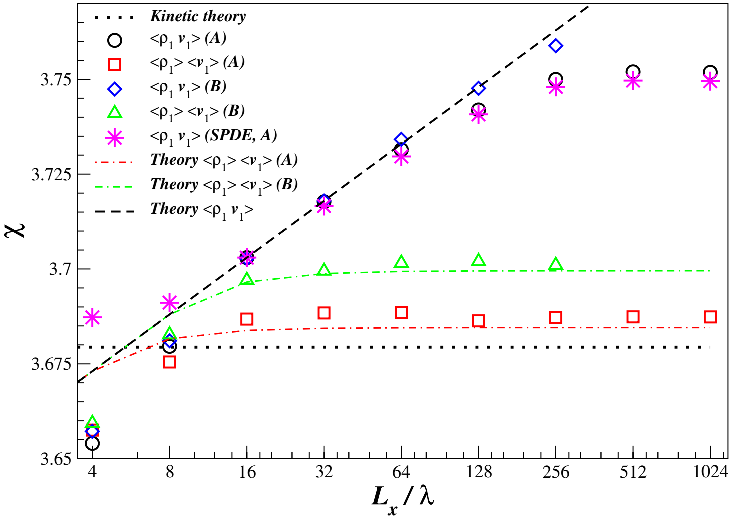


Figure: **Fluctuating hydro correctly predicts the dependence on system size!**

Particle Results (3D)

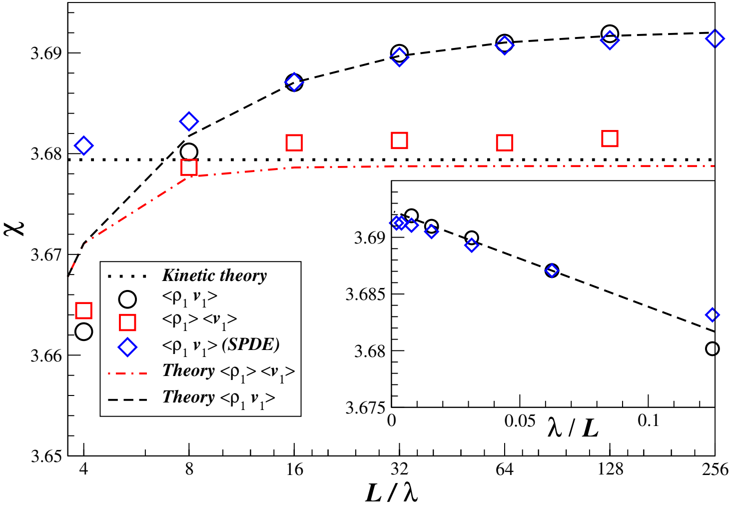


Figure: **Fluctuating hydro correctly predicts the dependence on system size!**

Conclusions

- **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 subdomain in hybrid methods.
- **Advection by the fluctuating velocities** fields leads to some very interesting physics and mathematics, such as giant fluctuations and renormalized transport coefficients.

Future Directions

- Improve and implement in a public-domain code the stochastic **particle methods** (parallelize, add chemistry, analyze theoretically).
- Develop numerical schemes for **incompressible** and **Low-Mach Number** fluctuating hydrodynamics.
- Theoretical work on the **equations of fluctuating hydrodynamics**: regularization, renormalization, systematic coarse-graining.
- **Direct fluid-structure coupling** between fluctuating hydrodynamics and microstructure (solute beads).
- 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 CFD (**macro**).

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