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

Aleksandar Donev[1]
Courant Institute, *New York University*
&
Alejandro L. Garcia, *San Jose State University*
John B. Bell, *Lawrence Berkeley National Laboratory*

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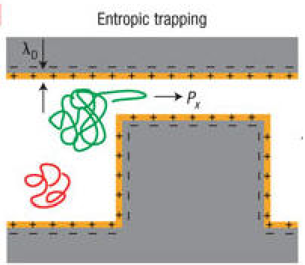
Outline

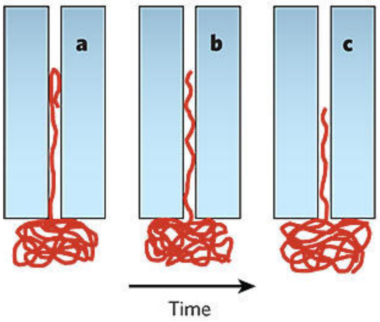
Micro- and nano-hydrodynamics

- 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.
- 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)



H. Craighead, Nature 442 (2006)

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

Levels of Coarse-Graining

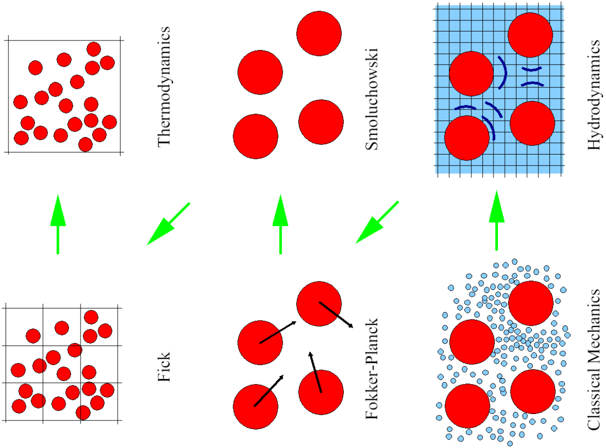


Figure: From Pep Español, "Statistical Mechanics of Coarse-Graining"

This talk: Particle/Continuum Hybrid

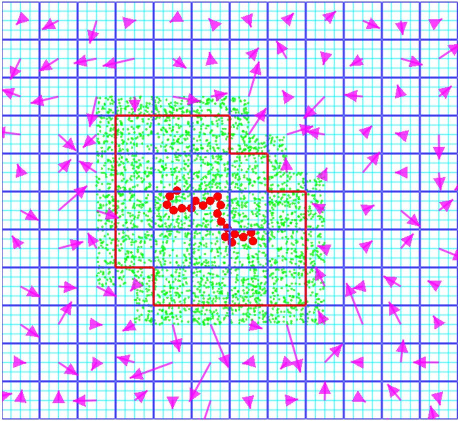


Figure: Hybrid method for a polymer chain.

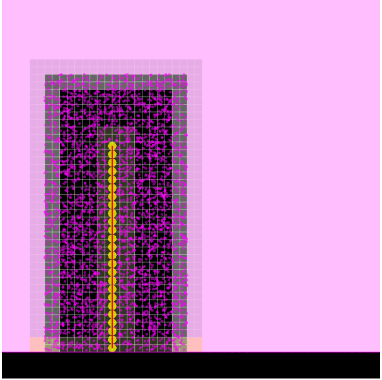
Particle Methods for Complex Fluids

- 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 collisions**.

Direct Simulation Monte Carlo (DSMC)



(MNG)

Tethered polymer chain in shear flow.

- **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 consisten structure and a **non-ideal equation of state**. [1].

Continuum Models of Fluid Dynamics

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

$$\mathbf{U}(\mathbf{r},t)=\left[\begin{array}{c}\rho\\ \mathbf{j}\\ e\end{array}\right]\cong\widetilde{\mathbf{U}}(\mathbf{r},t)=\sum_{i}\left[\begin{array}{c}m_{i}\\ m_{i}\boldsymbol{\upsilon}_{i}\\ m_{i}\upsilon_{i}^{2}/2\end{array}\right]\delta\left[\mathbf{r}-\mathbf{r}_{i}(t)\right],$$

  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 [2].
- This leads to **SPDEs of Langevin type** formed by postulating a **white-noise random flux** term in the usual Navier-Stokes-Fourier equations with magnitude determined from the **fluctuation-dissipation balance** condition, following Landau and Lifshitz.

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:

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

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

Landau-Lifshitz Navier-Stokes (LLNS) Equations

- 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].

Finite-Volume Schemes

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

- Generic **finite-volume spatial discretization**

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

  where $\mathbf{D}$: faces $\rightarrow$ cells is a **conservative** discrete divergence, $\mathbf{G}$: cells $\rightarrow$ faces is a discrete gradient.
- Here $\mathbf{W}$ is a collection of random normal numbers representing the (face-centered) stochastic fluxes.
- The **divergence** and **gradient** should be **duals**, $\mathbf{D}^\star = -\mathbf{G}$.
- Advection should be **skew-adjoint** (non-dissipative) if $\boldsymbol{\nabla} \cdot \mathbf{v} = 0$,

$$(\mathbf{D}\mathbf{V})^\star = -(\mathbf{D}\mathbf{V}) \;\text{ if }\; (\mathbf{D}\mathbf{V})\,\mathbf{1} = \mathbf{0}.$$

Weak Accuracy

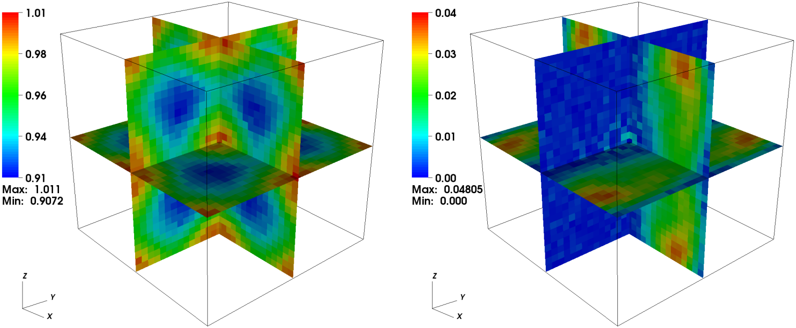


Figure: Equilibrium discrete spectra (static structure factors) $S_{\rho,\rho}(\mathbf{k}) \sim \langle \hat{\rho}\hat{\rho}^{\star} \rangle$ (should be unity for all discrete wavenumbers) and $S_{\rho,\mathbf{v}}(\mathbf{k}) \sim \langle \hat{\rho}\hat{v}_x^{\star} \rangle$ (should be zero) for our RK3 collocated scheme.

Fluid-Structure Coupling using Particles

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- 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.

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 ideal **particle fluid has no internal structure**.

"*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**

Our Hybrid Algorithm

1. The hydro solution $\mathbf{u}_H$ is computed everywhere, including the **particle patch**, giving an estimated total flux $\mathbf{\Phi}_H$.
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, giving the total particle flux $\mathbf{\Phi}_p$.
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, $\mathbf{u}_H \leftarrow \mathbf{u}_H - \mathbf{\Phi}_H + \mathbf{\Phi}_p$.

Other Hybrid Algorithms

- 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 progresss).

Brownian Bead

- Themal fluctuations *push a sphere of size* a *and density* ρ' *suspended in a stationary fluid with density* ρ *and viscosity* η (**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 η 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 $D \sim kT/(a\eta)$.

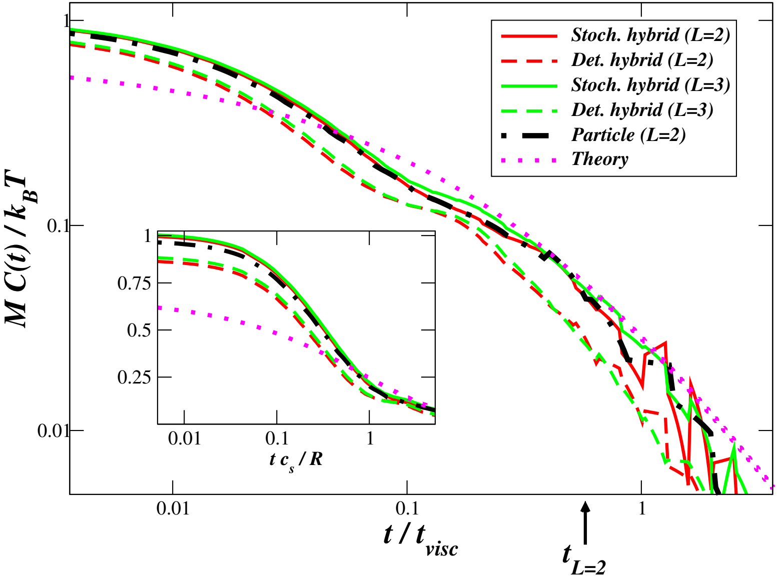
Velocity Autocorrelation Function

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

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

- 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 one third of the kinetic energy decays at the sound time scale.
- Hydrodynamic persistence (conservation) gives a **long-time power-law tail** $C(t) \sim (k_B T / M)(t / t_{visc})^{-3/2}$ that can be quantified using fluctuating hydrodynamics.
- The diffusion coefficient is the **integral of the VACF** and is strongly-affected by the tail.

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
$t\,/\,t_{visc}$
$t_{L=2}$
1
0.75
0.5
0.25
0.01
0.1
1
$t\,c_s\,/\,R$

The adiabatic piston problem

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Relaxation Toward Equilibrium

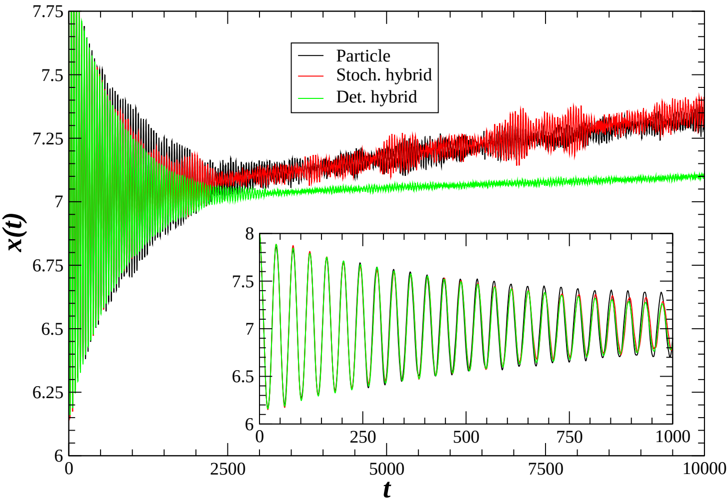


Figure: Massive rigid piston ($M/m = 4000$) not in mechanical equilibrium: **The deterministic hybrid gives the wrong answer!**

VACF for Piston

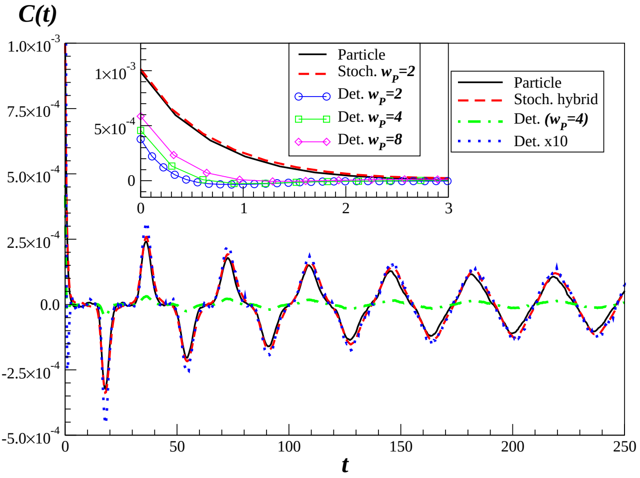


Figure: The VACF for a rigid piston of mas $M/m = 1000$ at thermal equilibrium: **Increasing the width of the particle region does not help: One must include the thermal fluctuations in the continuum solver**!

Nonequilibrium Fluctuations

- 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 $c_0(\mathbf{r})$:

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

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

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

- The velocity fluctuations drive and amplify the concentration fluctuations leading to so-called **giant fluctuations** [8].

Fractal Fronts in Diffusive Mixing

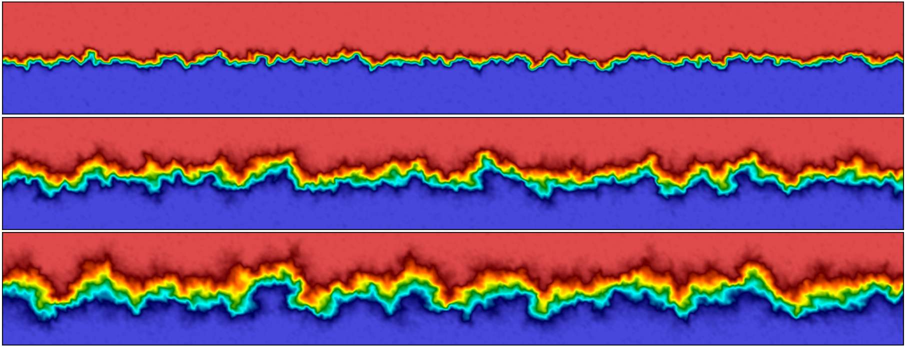


Figure: Snapshots of concentration in a miscible mixture showing the development of a *rough* diffusive interface between two miscible fluids in zero gravity [3, 8, 5].

Giant Fluctuations in Experiments

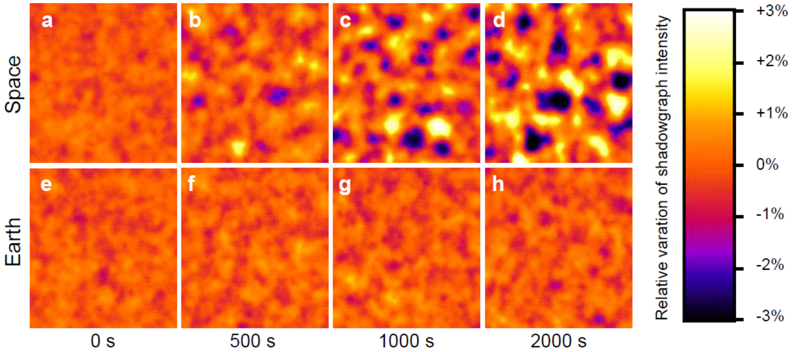


Figure: Experimental results by A. Vailati *et al.* from a microgravity environment [8] showing the enhancement of concentration fluctuations in space (box scale is **macroscopic**: 5mm on the side, 1mm thick).

Fluctuation-Enhanced Diffusion Coefficient

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

$$\partial_t (\delta c) + (\delta \mathbf{v}) \cdot \boldsymbol{\nabla} c_0 = \boldsymbol{\nabla} \cdot [-(\delta c)(\delta \mathbf{v}) + \chi \boldsymbol{\nabla} (\delta c)] + \dots$$

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

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

- 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**.

Fluctuation-Enhanced Diffusion Coefficient

- 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**

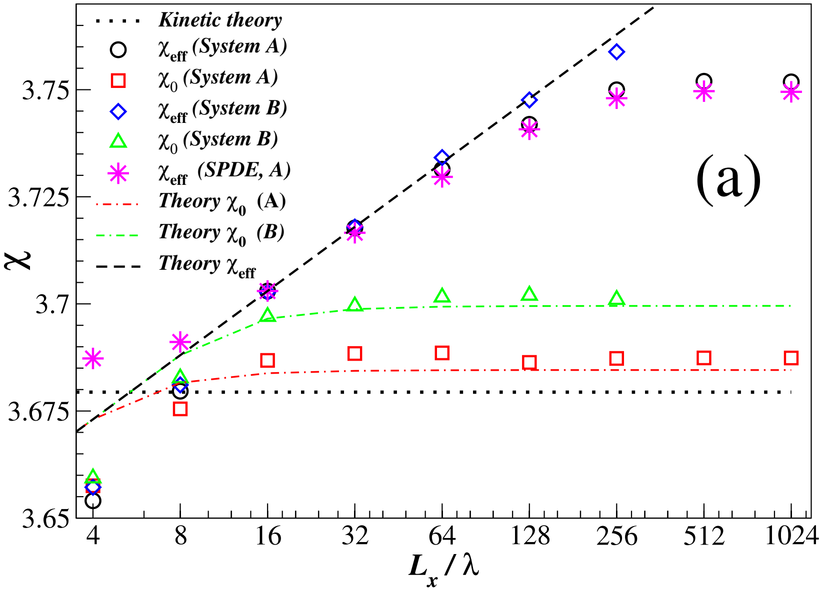
$$\chi_{\text{eff}}^{(2D)} \approx \chi + \frac{k_B T}{4\pi\rho(\chi+\nu)L_z} \ln \frac{L_x}{L_0}$$

- In **three dimensions**, $L_x = L_z = L \ll L_y$, χ_{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



Microscopic, Mesoscopic and Macroscopic Fluid Dynamics

- Instead of an ill-defined "molecular" or "bare" diffusivity, one should define a **locally renormalized diffusion coefficient** χ_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.

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 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**).

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