Computational Fluctuating Hydrodynamics Modeling of Giant Fluctuations

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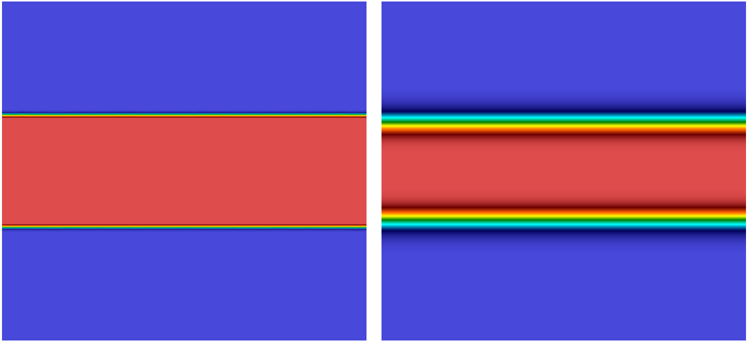
Outline

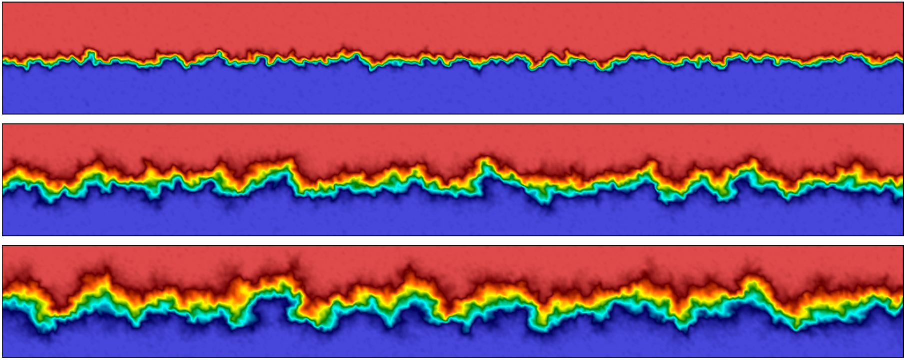
Micro- and nano-hydrodynamics

- 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 hope to demonstrate the general conclusion that **fluctuations should be taken into account at all levels**.

Deterministic Diffusive Mixing



Fractal Fronts in Diffusive Mixing



Snapshots of concentration in a miscible mixture showing the development of a *rough* diffusive interface between two miscible fluids in zero gravity [1, 2, 3]. A similar pattern is seen over a broad range of Schmidt numbers and is affected strongly by nonzero gravity.

Fluctuating Navier-Stokes Equations

- We will consider a binary fluid mixture with mass **concentration** $c = \rho_1/\rho$ for two fluids that are dynamically **identical**, where $\rho = \rho_1 + \rho_2$ (e.g., **fluorescently-labeled** molecules).
- Ignoring density and temperature fluctuations, equations of **incompressible isothermal fluctuating hydrodynamics** are

$$\partial_t \mathbf{v} + \mathbf{v} \cdot \boldsymbol{\nabla} \mathbf{v} = -\boldsymbol{\nabla}\pi + \nu \boldsymbol{\nabla}^2 \mathbf{v} + \boldsymbol{\nabla} \cdot \left(\sqrt{2\nu\rho^{-1}\, k_B T}\, \boldsymbol{\mathcal{W}} \right)$$

$$\partial_t c + \mathbf{v} \cdot \boldsymbol{\nabla} c = \chi \boldsymbol{\nabla}^2 c + \boldsymbol{\nabla} \cdot \left(\sqrt{2m\chi\rho^{-1}\, c(1-c)}\, \boldsymbol{\mathcal{W}}^{(c)} \right),$$

  where the **kinematic viscosity** $\nu = \eta/\rho$, and π is determined from incompressibility, $\boldsymbol{\nabla} \cdot \mathbf{v} = 0$.
- We assume that $\boldsymbol{\mathcal{W}}$ can be modeled as spatio-temporal **white noise** (a delta-correlated Gaussian random field), e.g.,

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

Fluctuating Hydrodynamics Equations

- Adding stochastic fluxes to the **non-linear** NS equations produces **ill-behaved stochastic PDEs** (solution is too irregular).
- No problem if we **linearize** the equations around a **steady mean state**, to obtain equations for the fluctuations around the mean.
- Finite-volume discretizations naturally impose a grid-scale **regularization** (smoothing) of the stochastic forcing.
- A **renormalization** of the transport coefficients is also necessary [1].
- We have algorithms and codes to solve the compressible equations (**collocated** and **staggered grid**), and recently also the incompressible and **low Mach number** ones (staggered grid) [4, 3].
- Solving these sort of equations numerically requires paying attention to **discrete fluctuation-dissipation balance**, in addition to the usual deterministic difficulties [4].

Finite-Volume Schemes

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

- 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 $\boldsymbol{\nabla} \cdot \mathbf{v} = 0$,

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

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:

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

- The velocity fluctuations drive and amplify the concentration fluctuations leading to so-called **giant fluctuations** [2].

Back of the Envelope

- The coupled *linearized velocity*-concentration system in **one dimension**:

$$\begin{aligned} v_t &= \nu v_{xx} + \sqrt{2\nu}\, W_x \\ c_t &= \chi c_{xx} - v\bar{c}_x, \end{aligned}$$

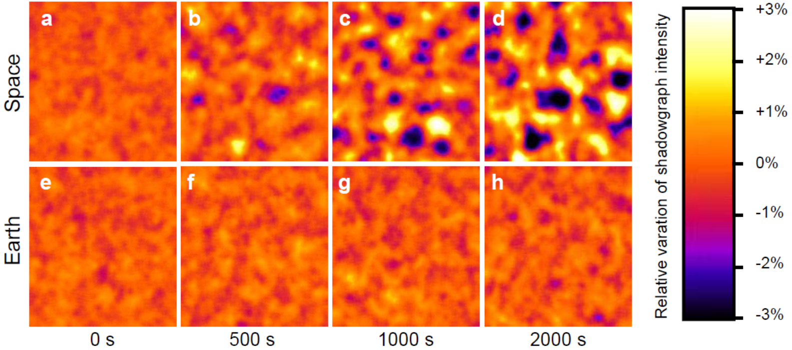
where $g = \bar{c}_x$ is the imposed background concentration gradient.

- The linearized system can be easily solved in Fourier space to give a **power-law divergence** for the spectrum of the concentration fluctuations as a function of wavenumber k,

$$\langle \hat{c}\hat{c}^\star \rangle \sim \frac{(\bar{c}_x)^2}{\chi(\chi+\nu)k^4}.$$

- Concentration fluctuations become **long-ranged** and are enhanced as the square of the gradient, to values much larger than equilibrium fluctuations.
- In real life the divergence is **suppressed** by surface tension, gravity, or boundaries (usually in that order).

Giant Fluctuations in Experiments



Experimental results by A. Vailati *et al.* from a microgravity environment [2] showing the enhancement of concentration fluctuations in space (box scale is **macroscopic**: 5mm on the side, 1mm thick).

Giant Fluctuations in Simulations

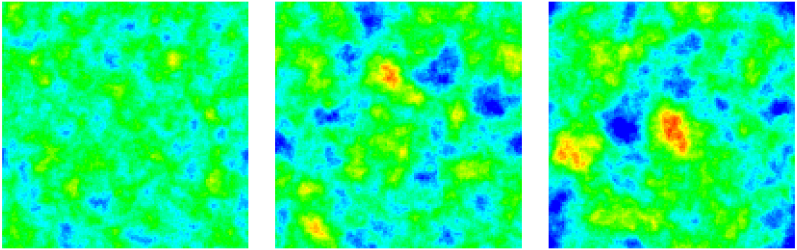


Figure: Computer simulations of microgravity experiments.

Spectrum of Concentration Fluctuations

- The **linearized equations** can be solved in the Fourier domain (ignoring boundaries for now) for any wavenumber **k**, denoting $k_\perp = k \sin\theta$ and $k_\parallel = k\cos\theta$.
- One finds **giant concentration fluctuations** proportional to the square of the applied gradient,

$$S_{c,c}^{\text{neq}} = \langle (\widehat{\delta c})(\widehat{\delta c}^\star) \rangle = \frac{k_B T}{\rho\chi(\nu+\chi)k^4} \left(\sin^2\theta\right) \left(\nabla\bar{c}\right)^2, \tag{1}$$

- The **finite height** of the container h imposes no-slip **boundary conditions**, which damps the power law at wavenumbers $k \sim 2\pi/h$.
- This is difficult to calculate analytically and one has to make drastic approximations, and **simulations** are ideal to compare to experiments.
- However, the **separation of time scales** between the slow diffusion and fast vorticity fluctuations poses a big challenge.

Simulation vs. Experiments

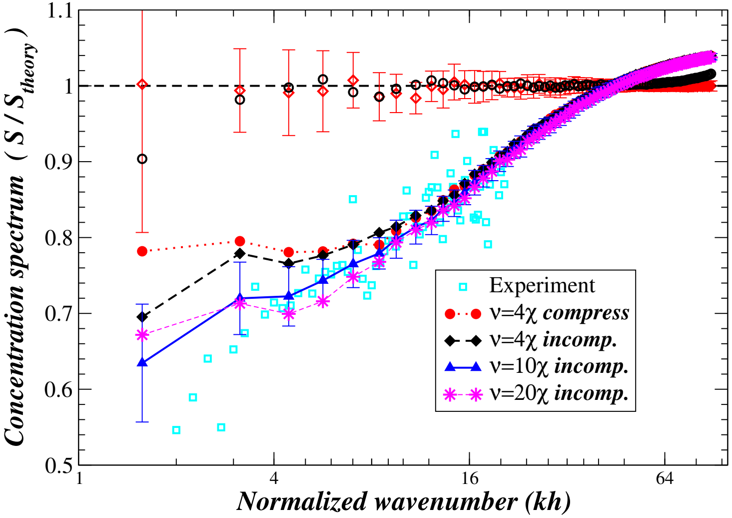


Figure: Giant fluctuations: simulation vs. experiment vs. *approximate* theory.

Low Mach Approximation

For isothermal mixtures of fluids with unequal densities, the incompressible approximation needs to be replaced with a **low Mach approximation**

$$\begin{aligned} D_t\rho &= -\rho\left(\boldsymbol{\nabla}\cdot\mathbf{v}\right) \\ \rho\left(D_t\mathbf{v}\right) &= -\boldsymbol{\nabla}\pi+\boldsymbol{\nabla}\cdot\left[\eta\left(\boldsymbol{\nabla}\mathbf{v}+\boldsymbol{\nabla}^T\mathbf{v}\right)+\boldsymbol{\Sigma}\right]+\rho\mathbf{g} \\ \rho\left(D_t c\right) &= \boldsymbol{\nabla}\cdot\left[\rho\chi\left(\boldsymbol{\nabla}c\right)+\boldsymbol{\Psi}\right], \end{aligned}$$

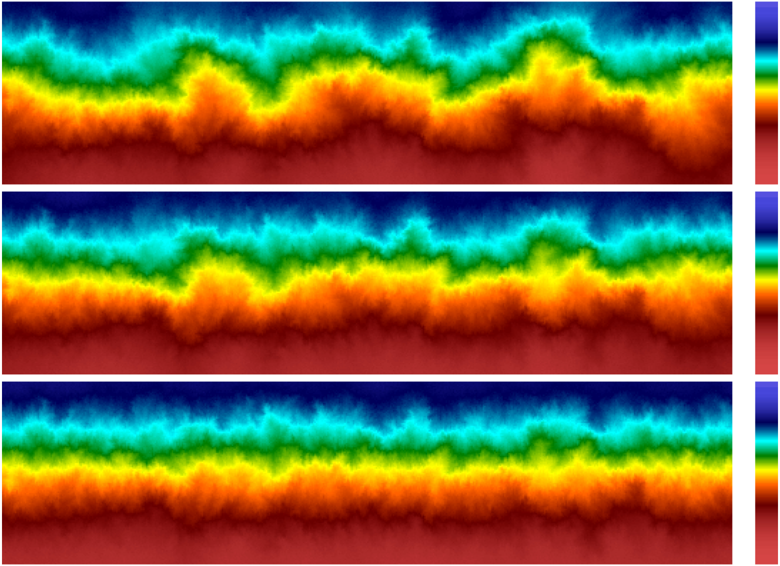
where $D_t\square=\partial_t\square+\mathbf{v}\cdot\boldsymbol{\nabla}\left(\square\right)$ and $\boldsymbol{\Sigma}$ and $\boldsymbol{\Psi}$ are stochastic fluxes determined from fluctuation-dissipation balance.

The incompressibility condition is replaced by the **equation of state (EOS) constraint**

$$\boldsymbol{\nabla}\cdot\mathbf{v}=\rho^{-1}\left(\frac{\partial\rho}{\partial c}\right)_{P,T}\left(D_t c\right)=\beta\left(D_t c\right),$$

where β is the **solutal expansion coefficient**.

Diffusive Mixing in Gravity



Boussinesq Approximation

- When $\beta \neq 0$ changes in composition (concentration) due to diffusion cause local expansion and contraction of the fluid and thus a nonzero $\boldsymbol{\nabla} \cdot \mathbf{v}$.
- The low Mach number equations are **substantially harder** to solve computationally because of the nontrivial constraint. They are also more problematic mathematically...
- Note that the usual incompressibility constraint $\boldsymbol{\nabla} \cdot \mathbf{v} = 0$ is obtained as $\beta \to 0$.
- A commonly-used simplification is the **Boussinesq approximation**, in which it is assumed that $\beta \ll 1$. More precisely, take the limit $\beta \to 0$ and $g \to \infty$ while keeping the product βg fixed.
- In theoretical calculations it is **assumed** that the **transport coefficients**, i.e., the viscosity and diffusion coefficients, **are constant**.
- This is definitely not so for viscosity in a water glycerol mixture as used by Croccolo et al. [5]!

Theoretical Approximations

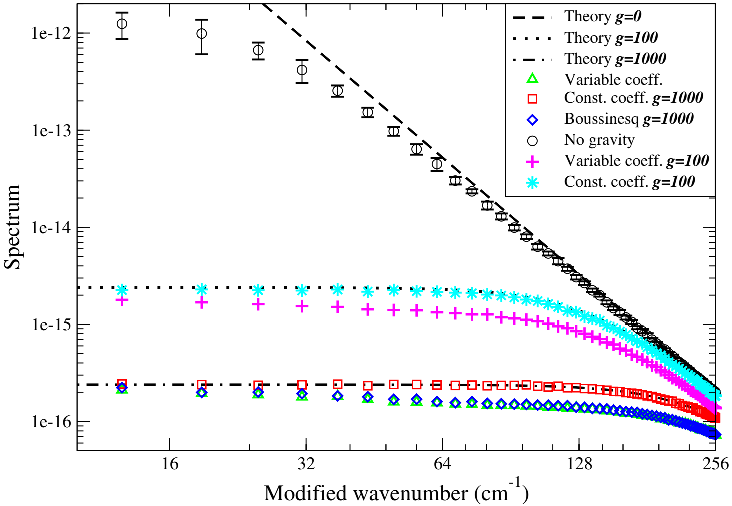
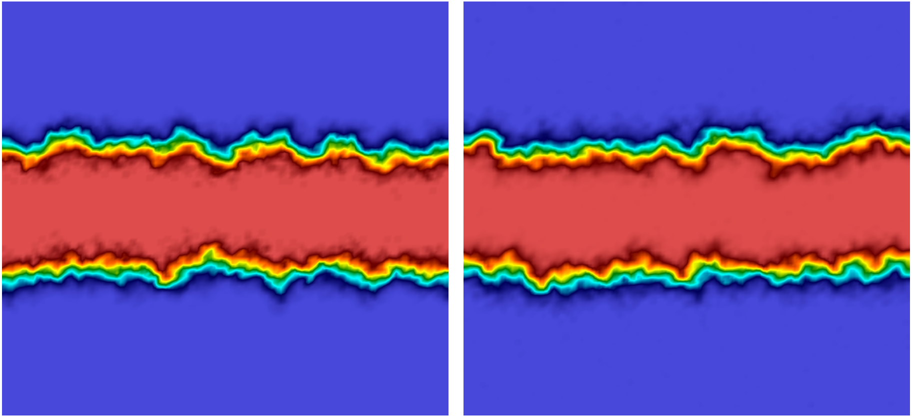


Figure: Comparison between the simple constant-coefficient Boussinesq theory and numerical results.

Molecular Dynamics Simulations

- We performed event-driven **hard disk simulations** of diffusive mixing with about 1.25 million disks.
- The two species had equal molecular diameter but potentially different molecular masses, with density ratio $R = m_2/m_1 = 1$, 2 or 4.
- In order to convert the particle data to hydrodynamic data, we employed finite-volume averaging over a grid of 128^2 hydrodynamic cells 10×10 molecular diameters (about 76 disks per hydrodynamic cell).
- We also performed fluctuating low Mach number **finite-volume simulations** using the same grid of hydrodynamic cells, at only a small fraction of the computational cost [6].
- Quantitative statistical comparison between the molecular dynamics and fluctuating hydrodynamics was excellent once the values of the **bare diffusion** and **viscosity** were adjusted based on the level of coarse-graining.

Hard-Disk Simulations



MD vs. Hydrodynamics

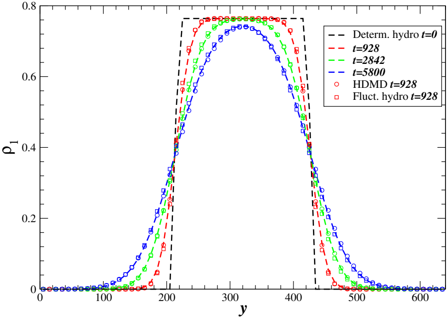


Figure: Diffusive evolution of the horizontally-averaged density for density ratio $R = 4$, as obtained from HDMD simulations (circles), deterministic hydrodynamics with effective diffusion coefficient $\chi_{\text{eff}} = 0.2$ (dashed lines), and fluctuating hydrodynamics with bare diffusion coefficient $\chi_0 = 0.09$ (squares).

MD vs. Hydrodynamics contd.

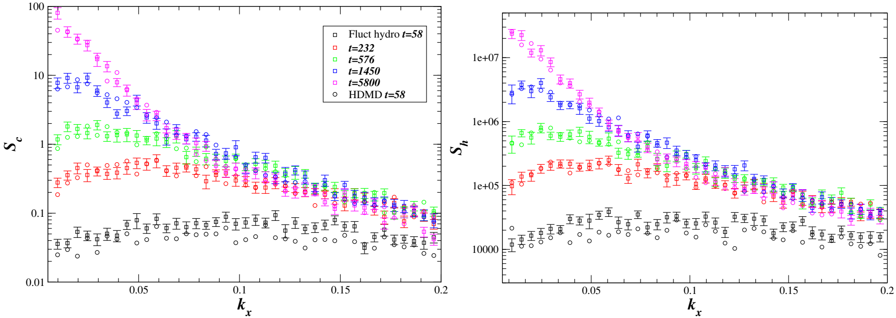
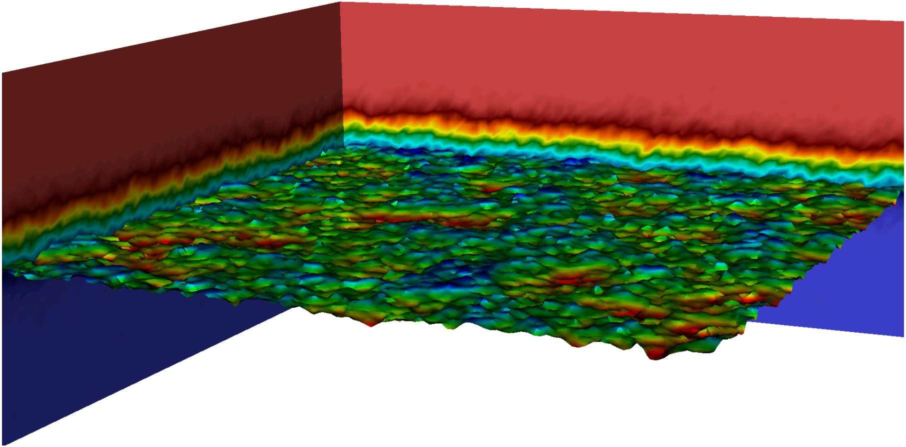
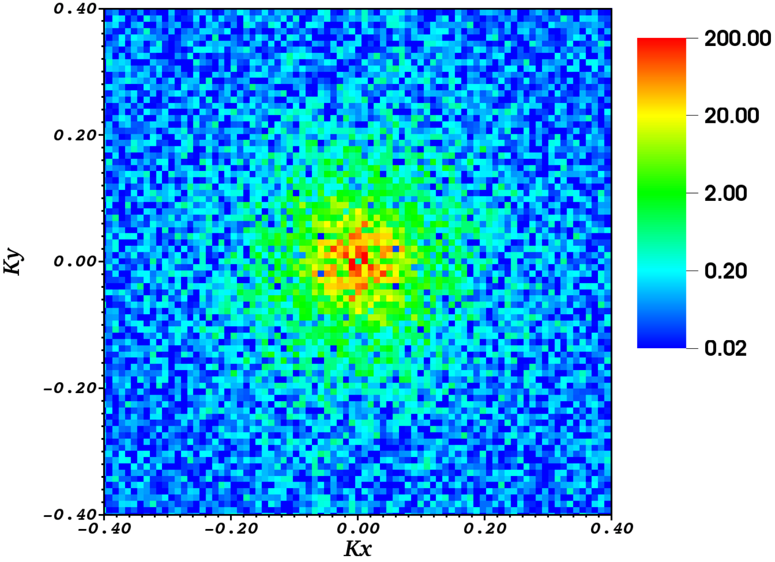


Figure: Discrete spatial spectrum of the interface fluctuations for mass ratio $R = 4$ at several points in time, for fluctuating hydrodynamics (squares with error bars) and HDMD (circles, error bars comparable to those for squares).

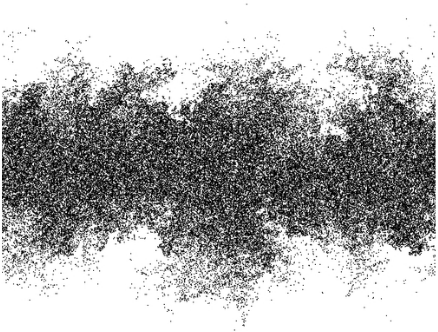
"Hard-Sphere" Simulations

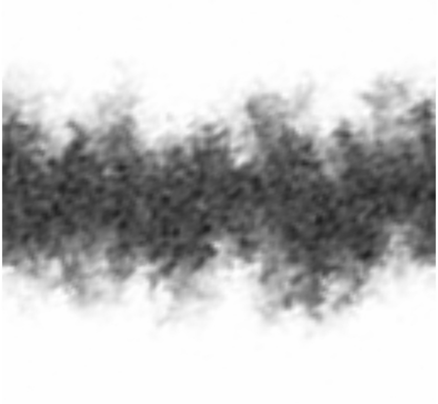


Interface Spectrum in 3D



Passively-Advected (Fluorescent) Tracers





Diffusion by Velocity Fluctuations

- Consider a large collection of **passively-advected particles** immersed in a fluctuating Stokes velocity field,

$$\begin{aligned} \partial_t \mathbf{v} &= \mathcal{P}\left[\nu \boldsymbol{\nabla}^2 \mathbf{v} + \boldsymbol{\nabla} \cdot \left(\sqrt{2\nu\rho^{-1}\, k_B T}\, \boldsymbol{\mathcal{W}}\right)\right] \\ \partial_t c &= -\mathbf{v} \cdot \boldsymbol{\nabla} c + \chi \boldsymbol{\nabla}^2 c + \boldsymbol{\nabla} \cdot \left(\sqrt{2\chi c}\, \boldsymbol{\mathcal{W}}^{(c)}\right), \end{aligned}$$

  where c is the number density for the particles, and $\mathcal{P}$ is the orthogonal projection onto the space of divergence-free velocity fields.
- In liquids diffusion of mass is much slower than diffusion of momentum, $\chi \ll \nu$, leading to a **Schmidt number**

$$S_c = \frac{\nu}{\chi} \sim 10^3.$$

- [With *Eric Vanden-Eijnden*]: There exists a limiting dynamics for c in the limit $S_c \to \infty$ in the scaling

$$\nu = \chi S_c, \quad \chi(\chi + \nu) \approx \chi\nu = \text{const}$$

Limiting Dynamics

- Formal adiabatic elimination of **v** as a fast variable gives *approximately* the following limiting **stochastic advection-diffusion equation** for concentration (common in turbulence models):

$$\partial_t c = -\mathbf{v} \cdot \boldsymbol{\nabla} c + (\chi + \Delta\chi)\, \boldsymbol{\nabla}^2 c,$$

  where $\Delta\chi$ is a **renormalization** of the diffusion coefficient [1], approximated here by a local diffusion.

- The advection velocity here is a **white-in-time** process that can be sampled by solving the steady Stokes equation

$$\begin{aligned} \boldsymbol{\nabla}\pi &= \nu \boldsymbol{\nabla}^2 \mathbf{v} + \boldsymbol{\nabla} \cdot \left(\sqrt{2\nu\rho^{-1}\, k_B T}\, \boldsymbol{\mathcal{W}} \right) \\ \boldsymbol{\nabla} \cdot \mathbf{v} &= 0. \end{aligned}$$

Simulating the Limiting Dynamics

The limiting dynamics can be efficiently simulated using the following **predictor-corrector algorithm**:

1. Generate a random advection velocity

$$\boldsymbol{\nabla}\pi^{n+\frac{1}{2}} = \nu\left(\boldsymbol{\nabla}^2\mathbf{v}^n\right) + \Delta t^{-\frac{1}{2}}\boldsymbol{\nabla}\cdot\left(\sqrt{2\nu\rho^{-1}\,k_B T}\,\boldsymbol{\mathcal{W}}^n\right)$$
$$\boldsymbol{\nabla}\cdot\mathbf{v}^n = 0.$$

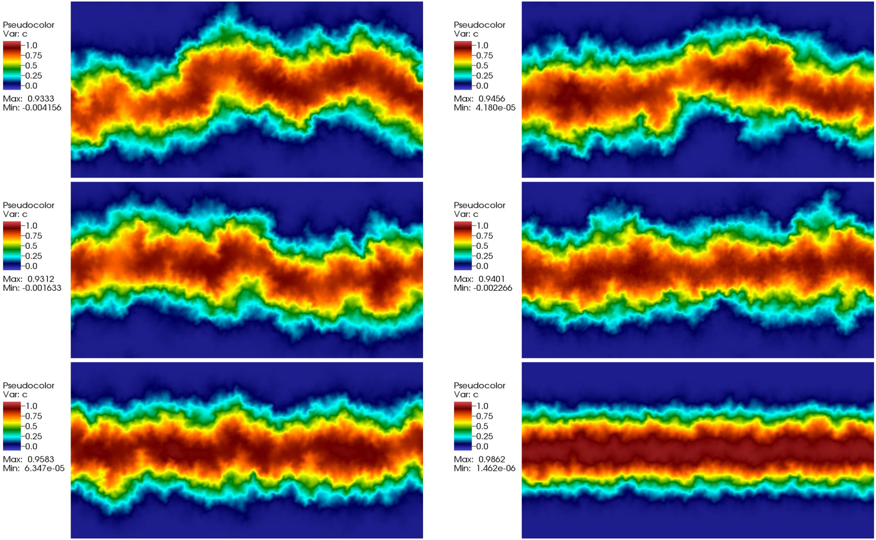
2. Take a predictor step for concentration, e.g., using Crank-Nicolson,

$$\frac{\tilde{c}^{n+1} - c^n}{\Delta t} = -\mathbf{v}^n\cdot\boldsymbol{\nabla}c^n + \chi\boldsymbol{\nabla}^2\left(\frac{c^n + \tilde{c}^{n+1}}{2}\right).$$

3. Take a corrector step for concentration

$$\frac{c^{n+1} - c^n}{\Delta t} = -\mathbf{v}^n\cdot\boldsymbol{\nabla}\left(\frac{c^n + \tilde{c}^{n+1}}{2}\right) + \chi\boldsymbol{\nabla}^2\left(\frac{c^n + c^{n+1}}{2}\right).$$

Changing S_c from 1 to ∞



Questionable Separation

- The above animation makes it clear S_c needs to be very large to be close to the limiting dynamics.
- The separation of time scales between the slowest velocity mode and the fastest concentration mode is

$$\frac{k_{\max}^2 \nu}{k_{\min}^2 \chi} = \frac{S_c}{N_c^2},$$

  where N_c is the number of modes (along a direction).
- Full separation of scales requires $S_c \gg N_c^2$, which is often not met in practice, e.g., $S_c \sim 500$ in a typical liquid like water.
- Similarly **questionable** is the **assumption** that particles immersed in a fluid follow a diffusion equation: what about large-scale slow velocity fluctuations?
- Under certain conditions the limiting dynamics should be a good approximation, but seems hard to justify in general.

Conclusions

- Fluctuations are **not just a microscopic phenomenon**: giant fluctuations can reach macroscopic dimensions or certainly dimensions much larger than molecular.
- **Fluctuating hydrodynamics** agrees with molecular dynamics of diffusive mixing in mixtures of hard disks and seems to be a very good coarse-grained model for fluids, despite unresolved issues.
- Diffusion is strongly affected and often dominated by **advection by velocity fluctuations**.
- In the presence of density variations one should use the **low Mach number equations** instead of the incompressible approximation.
- Even coarse-grained methods need to be accelerated due to **large separation of time scales** between advective and diffusive phenomena. One can both decrease or increase the separation of scales to allow for efficient simulation.

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