Multiscale Problems in Fluctuating Hydrodynamics

Aleksandar Donev

Courant Institute, *New York University*
&
Eric Vanden-Eijnden, *Courant*
John B. Bell, *LBNL*
Boyce Griffith, *Courant*
and others

Modelling the Dynamics of Complex Molecular Systems
Lorentz Center Workshop
August 2012

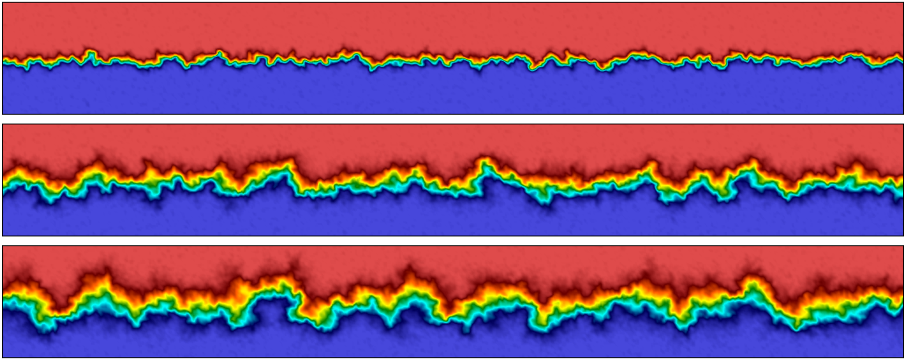
Outline

1. Giant Fluctuations

2. Fluctuating Hydrodynamics

3. Limiting Diffusive Dynamics

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.,

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

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** [2].

Back of the Envelope

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

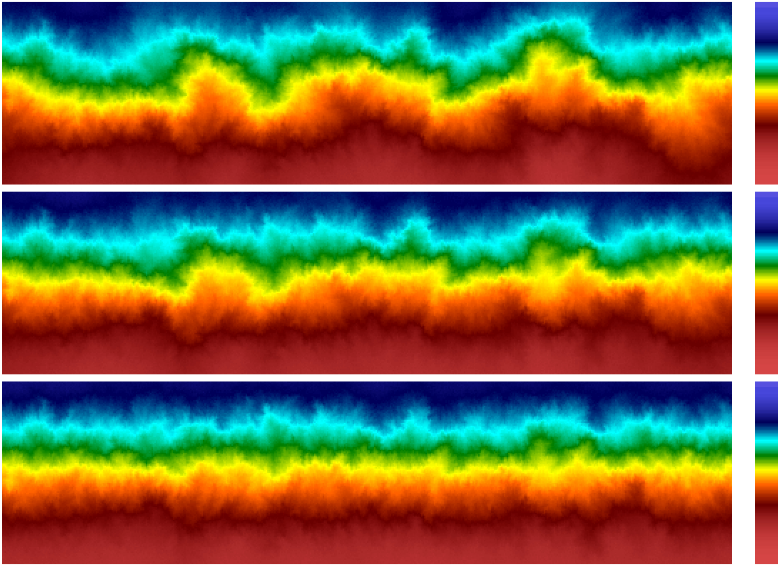
$$\begin{aligned} v_t &= \nu v_{xx} + \sqrt{2\nu}\, W_x \\ c_t &= \chi c_{xx} - v\bar{c}_x, \end{aligned}$$

  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,

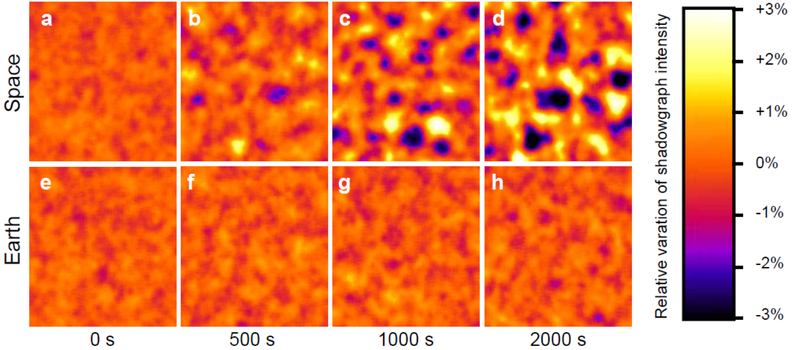
$$\langle \hat{c}\hat{c}^{\star} \rangle \sim \frac{(\bar{c}_x)^2}{\chi(\chi+\nu)k^4}.$$

- 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).

Diffusive Mixing in Gravity



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).

Low Mach Approximation

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

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

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**

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

.

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

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

- Generic **finite-volume spatial discretization**

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

  where $\mathbf{D}$: faces $\rightarrow$ cells is a **conservative** discrete divergence, $\mathbf{G}$: cells $\rightarrow$ faces is a discrete gradient.

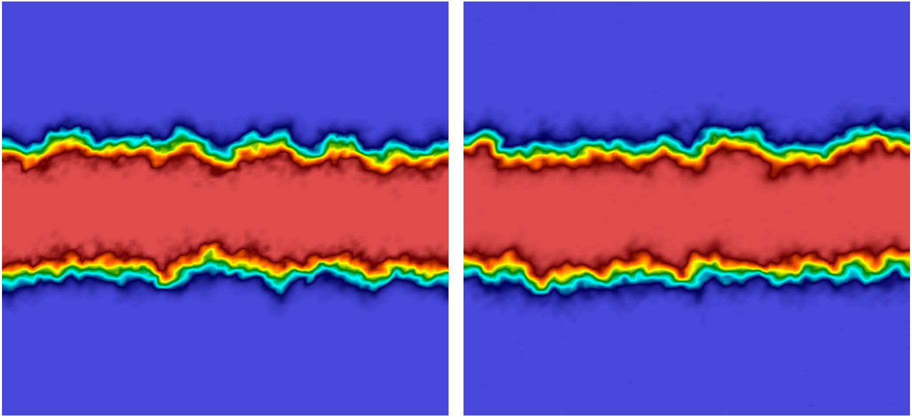
- Here $\mathbf{W}$ is a collection of random normal numbers representing the (face-centered) stochastic fluxes.
- The **divergence** and **gradient** should be **duals**, $\mathbf{D}^\star = -\mathbf{G}$.
- Advection should be **skew-adjoint** (non-dissipative) if $\boldsymbol{\nabla} \cdot \mathbf{v}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

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

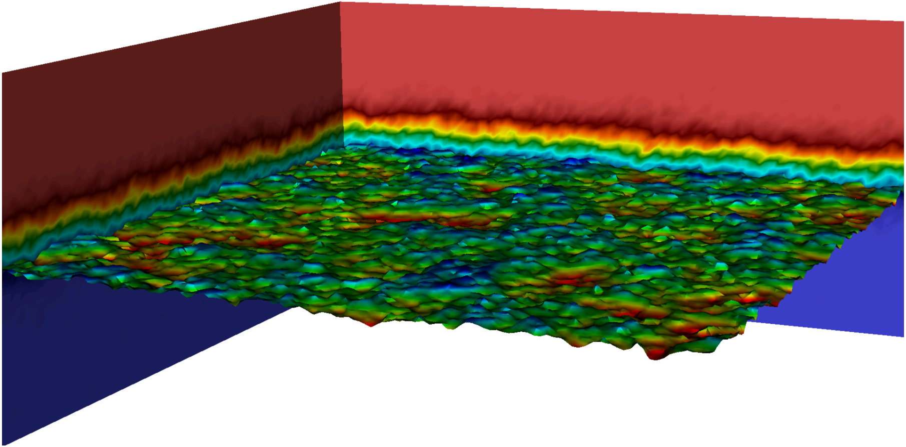
Molecular Dynamics Simulations

- 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 [5].
- 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.

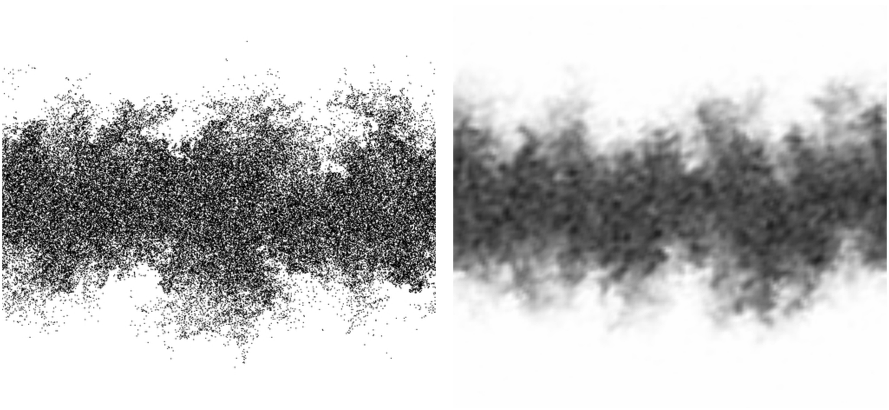
Hard-Disk Simulations



"Hard-Sphere" Simulations



Passively-Advected (Fluorescent) Tracers



Diffusion by Velocity Fluctuations

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

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

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

  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

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

Rescaling Dynamics

- Consider a family of equations with **rescaled coefficients**

$$\nu' = \epsilon^{-1}\nu, \quad \chi' = \epsilon\chi,$$

  which has $\nu'\chi' = \chi\nu$ but $S_c' = \epsilon^{-2} S_c$.
- For $\epsilon = 1$ we get the original dynamics, and as $\epsilon \to 0$ we get the limiting dynamics $S_c \to \infty$.
- **Rescale time** as $t' = \epsilon^{-1} t$, to get the rescaled equations

$$\begin{aligned} \partial_{t'}\mathbf{v} =& \mathcal{P}\left[\epsilon^{-2}\nu\boldsymbol{\nabla}^2\mathbf{v} + \boldsymbol{\nabla}\cdot\left(\sqrt{2\epsilon^{-2}\nu\rho^{-1}\,k_B T}\,\boldsymbol{\mathcal{W}}\right)\right] \\ \partial_{t'} c =& -\epsilon^{-1}\mathbf{v}\cdot\boldsymbol{\nabla} c + \chi\boldsymbol{\nabla}^2 c + \text{stoch.} \end{aligned}$$

- On the rescaled time scale the dynamics will be very similar to the limiting dynamics if ϵ is small, specifically, if there is a very large separation of scales between the velocity and concentration dynamics.

Adiabatic Elimination of **v**

- The existence of the limiting dynamics follows from well-established limit theorems, see review by Eric Vanden-Eijnden in Section IV of [6] or a series of three articles on "Adiabatic elimination in stochastic systems" in Phys. Rev. A [7].
- Briefly, if the Liouville operator has the form

$$L = L_0 + \epsilon^{-1} L_1 + \epsilon^{-2} L_2,$$

in the limiting dynamics $\epsilon \to 0$ we have

$$L' = \mathcal{P}_\epsilon L_0 \mathcal{P}_\epsilon - \mathcal{P}_\epsilon L_1 L_2^{-1} L_1 \mathcal{P}_\epsilon,$$

where $\mathcal{P}_\epsilon$ is a Zwanzig projection operator

- In our specific case

$$L_0 \longleftrightarrow \chi \boldsymbol{\nabla}^2 c, \quad L_1 \longleftrightarrow -\epsilon^{-1} \mathbf{v} \cdot \boldsymbol{\nabla} c, \quad L_2 \longleftrightarrow \boldsymbol{\mathcal{P}} \epsilon^{-2} \nu \boldsymbol{\nabla}^2 \mathbf{v}$$

Limiting Dynamics

- A Fourier-space calculation 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

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

$$\boldsymbol{\nabla} \cdot \mathbf{v} = 0.$$

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

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

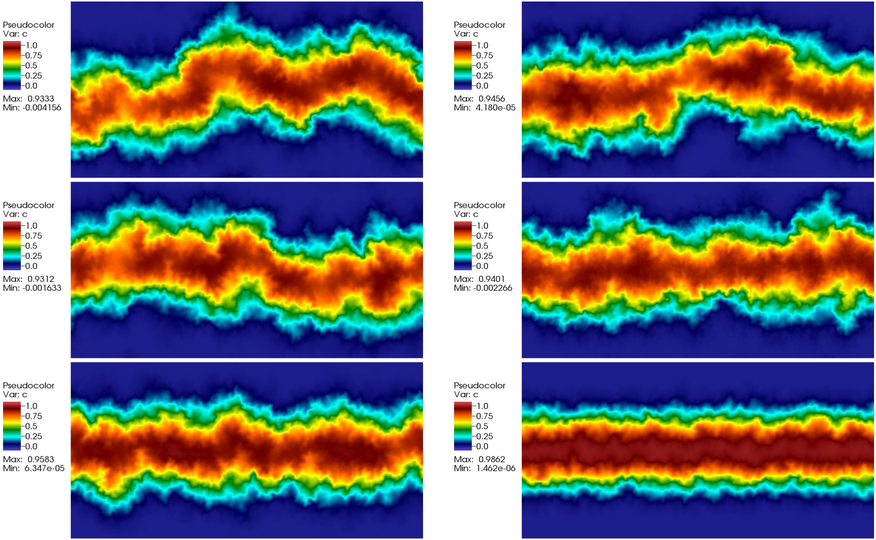
Multiscale Temporal Integrators

- There are two ways in which the rescaled dynamics can be used. Recall that

$$\nu' = \epsilon^{-1}\nu, \quad \chi' = \epsilon\chi \quad \Rightarrow \quad S_c' = \epsilon^{-2} S_c.$$

- The first one is to **increase** $\epsilon \gg 1$ and use this to *decrease* the separation of time scales to the point where the rescaled dynamics is computationally feasible, in the spirit of the **seamless HMM method** (Vanden-Eijnden *et al*).
- The second one is to **decrease** $\epsilon \to 0$ and directly simulate the liming dynamics, which assumes infinite separation of scales (overdamped limit).
- If there is strong separation of scales in the original problem either will do and in fact there may exist integrators that can handle the limit $\epsilon \to 0$ gracefully (**stiffly-accurate integrators**).

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

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

  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**.
- 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.
- In the case of SPDEs there are many ($\gg 1$) length and time scales. Can one construct **many-scale temporal integrators** that are accurate even when they under-resolve the fast fluctuations?

References

A. Donev, A. L. Garcia, Anton de la Fuente, and J. B. Bell.
Enhancement of Diffusive Transport by Nonequilibrium Thermal Fluctuations.
J. of Statistical Mechanics: Theory and Experiment, 2011:P06014, 2011.

A. Vailati, R. Cerbino, S. Mazzoni, C. J. Takacs, D. S. Cannell, and M. Giglio.
Fractal fronts of diffusion in microgravity.
Nature Communications, 2:290, 2011.

F. Balboa Usabiaga, J. B. Bell, R. Delgado-Buscalioni, A. Donev, T. G. Fai, B. E. Griffith, and C. S. Peskin.
Staggered Schemes for Incompressible Fluctuating Hydrodynamics.
To appear in SIAM J. Multiscale Modeling and Simulation, 2012.

A. Donev, E. Vanden-Eijnden, A. L. Garcia, and J. B. Bell.
On the Accuracy of Explicit Finite-Volume Schemes for Fluctuating Hydrodynamics.
CAMCOS, 5(2):149–197, 2010.

A. J. Nonaka, A. Donev, T. Fai, A. L. Garcia, and J. B. Bell.
Low Mach Number Fluctuating Hydrodynamics of Fluid Mixtures.
In preparation, 2012.

C. Hijón, P. Español, E. Vanden-Eijnden, and R. Delgado-Buscalioni.
Mori-zwanzig formalism as a practical computational tool.
Faraday Discuss., 144:301–322, 2009.

C. W. Gardiner and M. L. Steyn-Ross.
Adiabatic elimination in stochastic systems. i-iii.
Phys. Rev. A, 29:2814–2822, 2823–2833, 2834–2844, 1984.