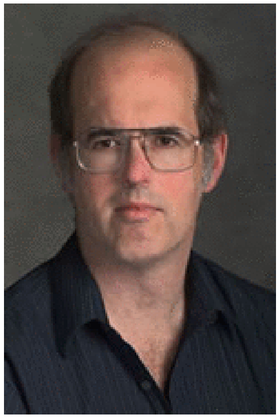
Enhancement of Diffusive Transport by Non-equilibrium Thermal Fluctuations



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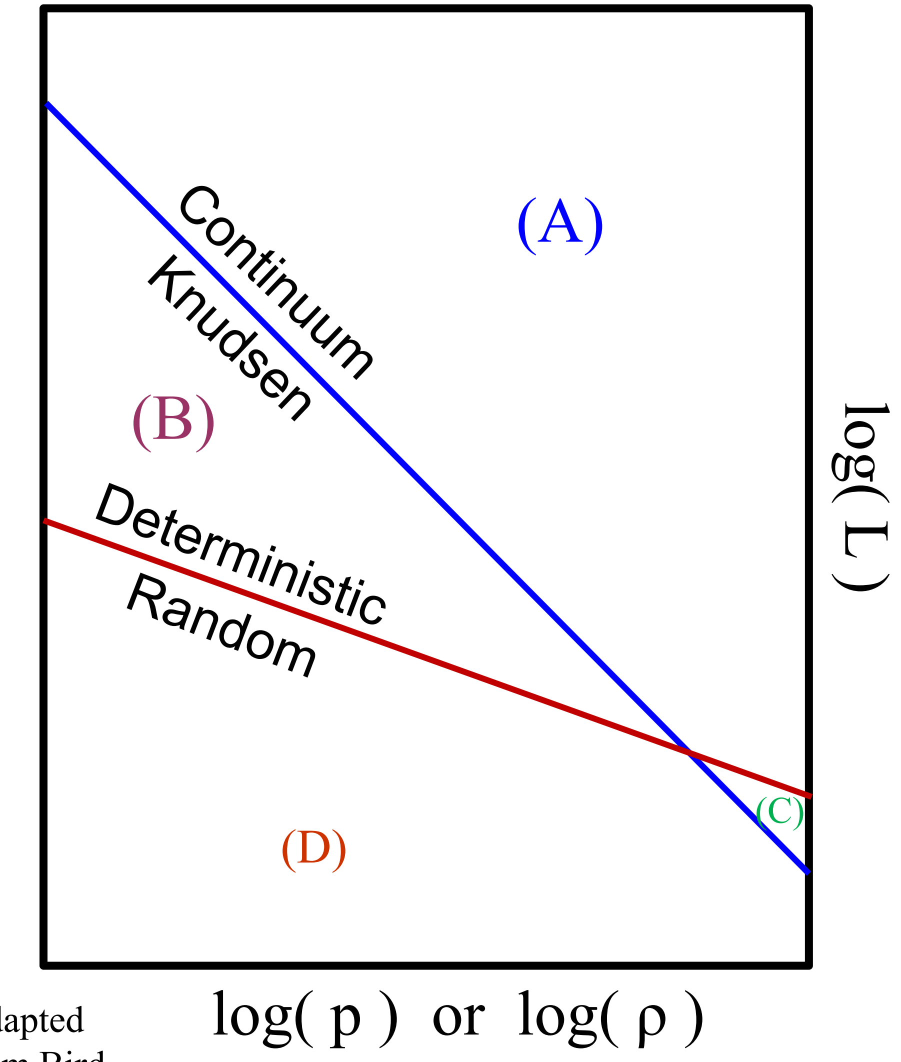
Regimes for Dilute Gases

Continuum vs. Knudsen
Deterministic vs. Random

(A) Continuum, Deterministic:
Fluid mechanics & CFD

(B) Knudsen, Deterministic:
Kinetic theory & RGD

(C) Continuum, Random:
Fluctuating hydrodynamics

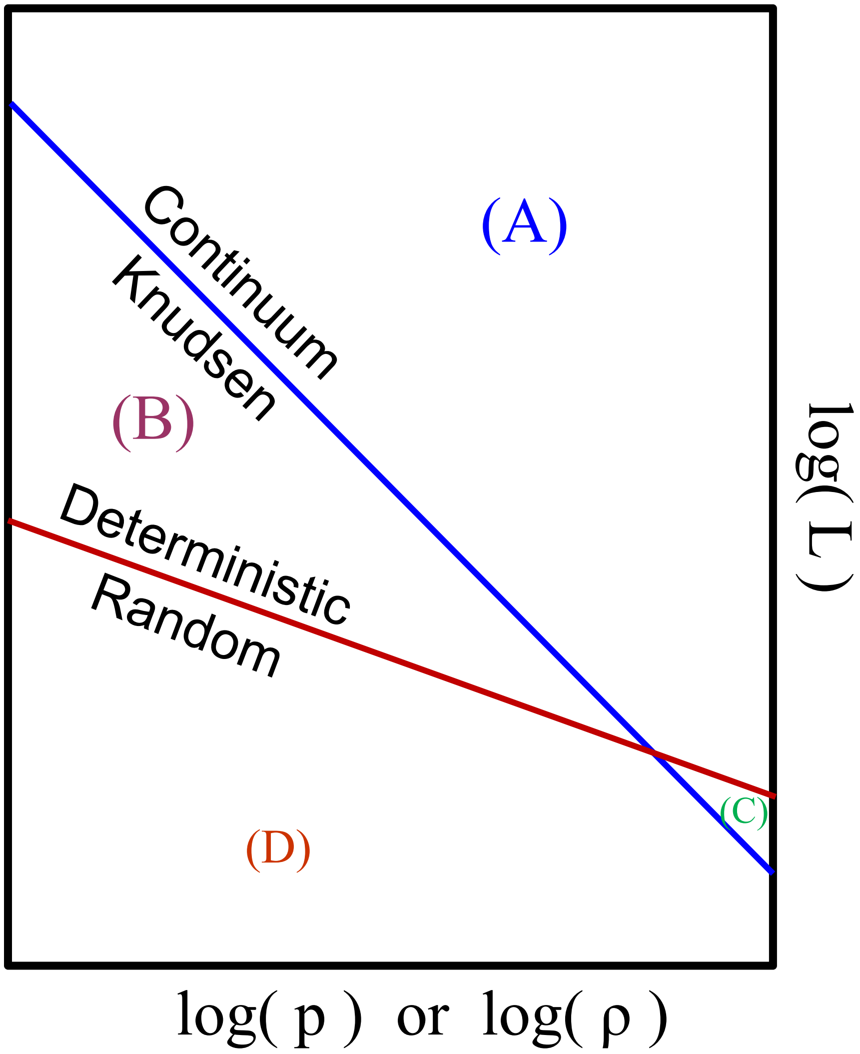


Adapted
from Bird

DSMC Simulations

DSMC can be used for *all* regimes but is primarily used in the Knudsen regime due to computational efficiency.

DSMC also useful for the study of fluctuations.



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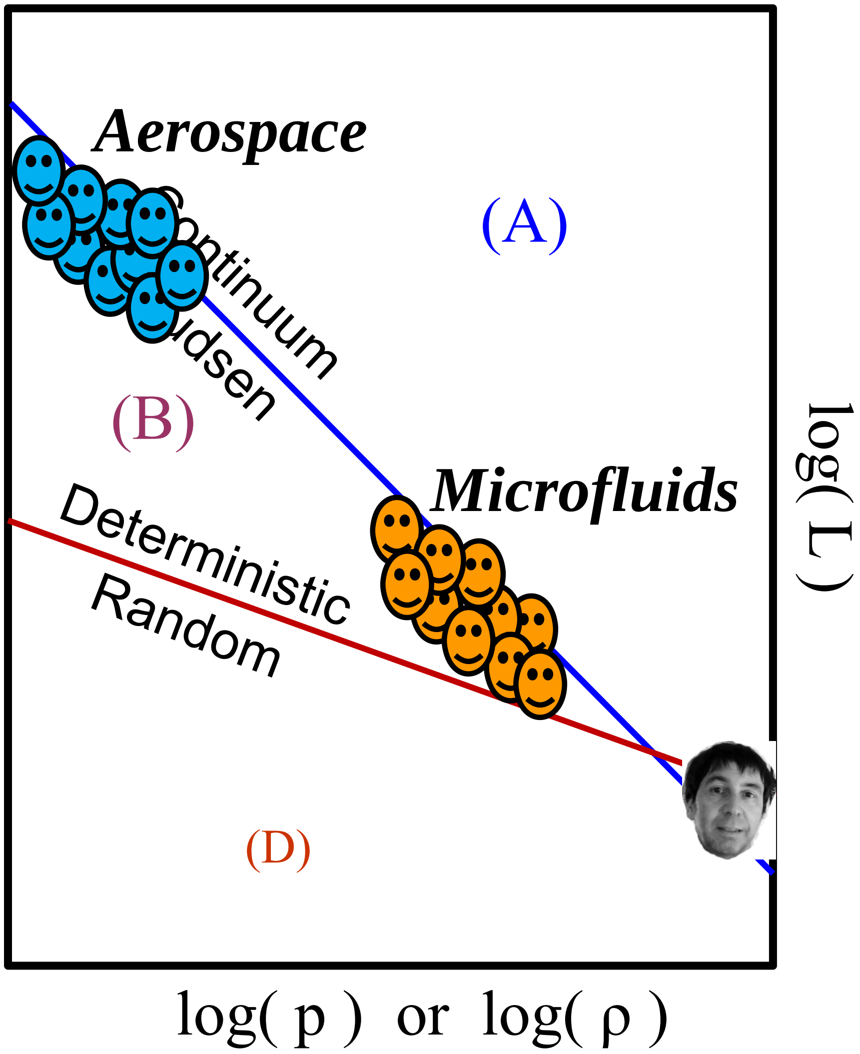
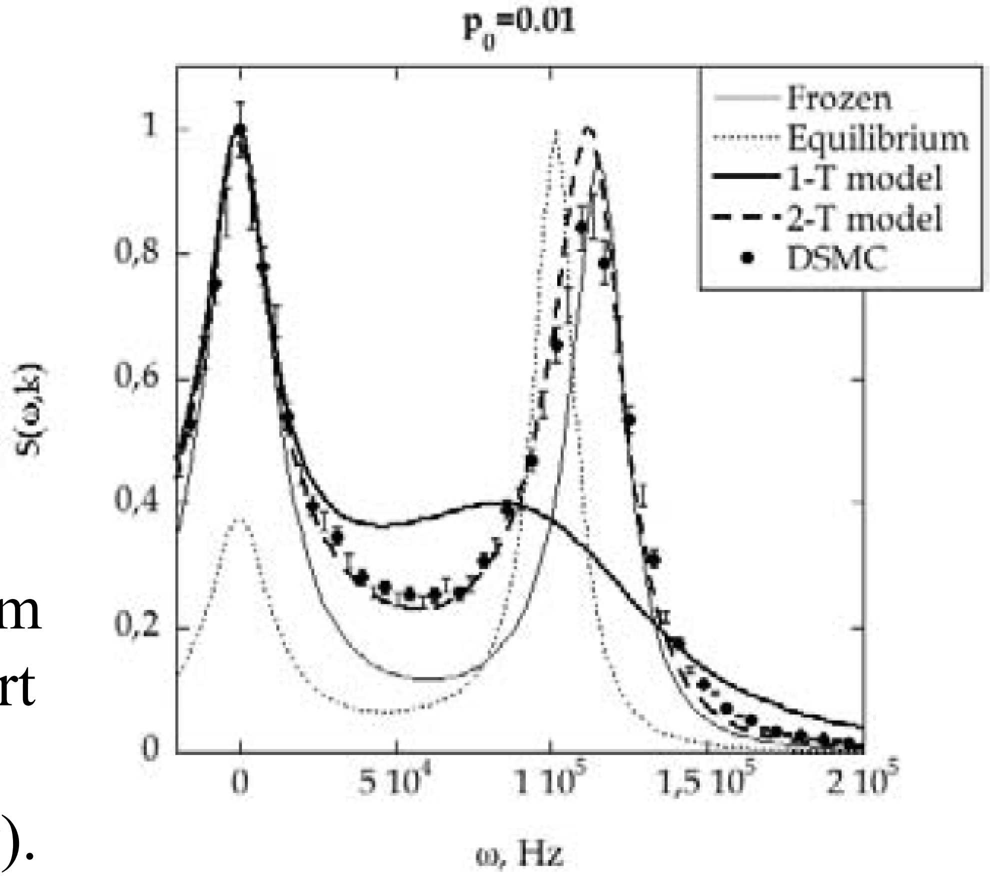


Fluctuations in DSMC

Fluctuations in DSMC are <u>not</u> due to Monte Carlo; they are physically correct at hydrodynamic and kinetic scales.

Fluctuation spectra at equilibrium may be used to measure transport properties (e.g., contribution of internal energy to bulk viscosity).

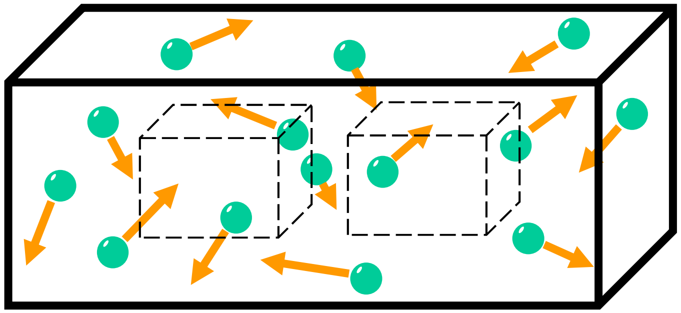


D. Bruno, 27th RGD Proceedings (2011)

Correlations of Fluctuations

At equilibrium, fluctuations of conjugate variables, such as density and fluid velocity, quantities are uncorrelated.

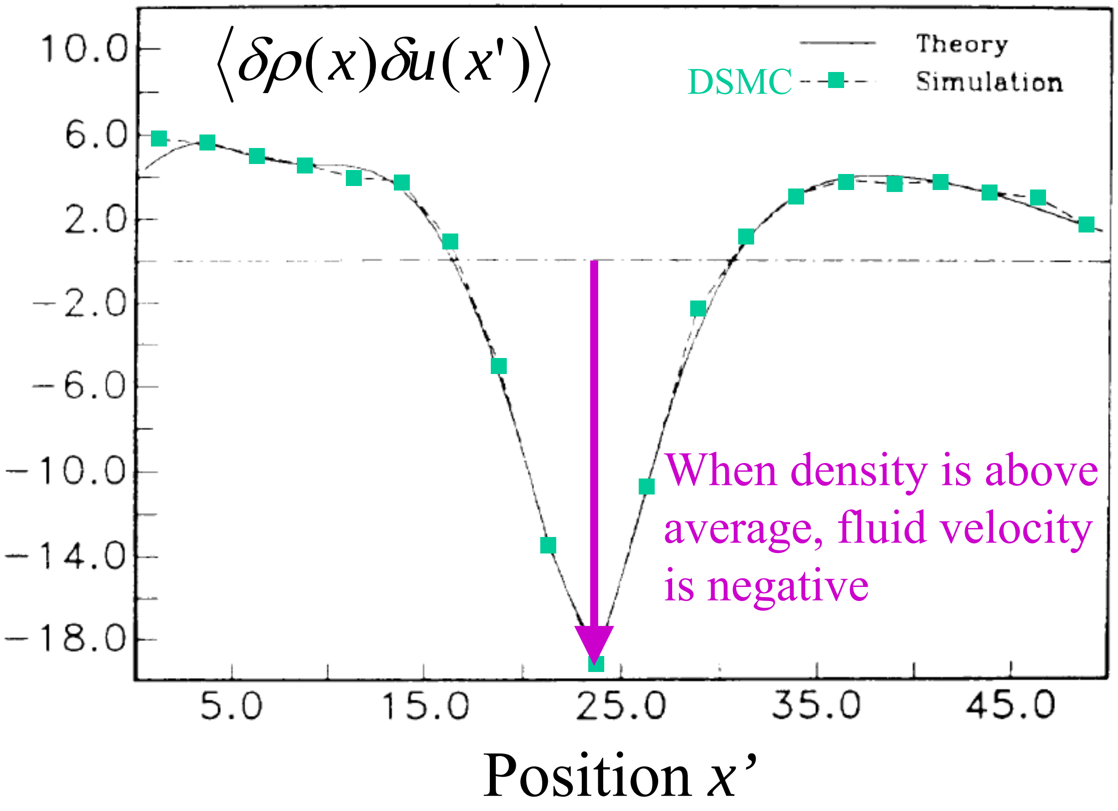
$$\langle \delta\rho(x,t)\delta u(x',t) \rangle = 0$$



Out of equilibrium, (e.g., gradient of temperature) these fluctuations become correlated at a distance.

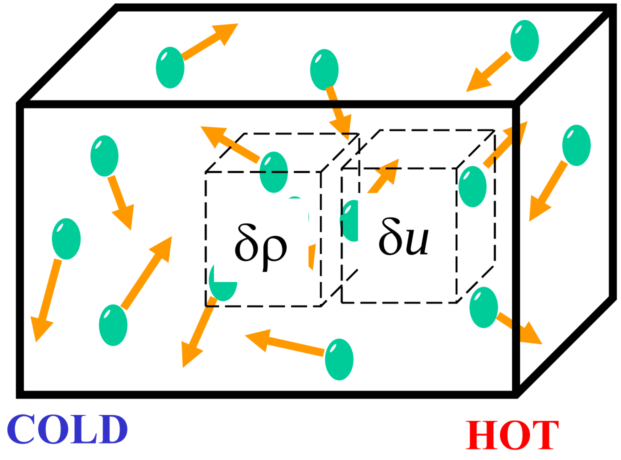
Density-Velocity Correlation

Correlation of density-velocity fluctuations under ∇T



When density is above average, fluid velocity is negative

Theory is Landau fluctuating hydrodynamics

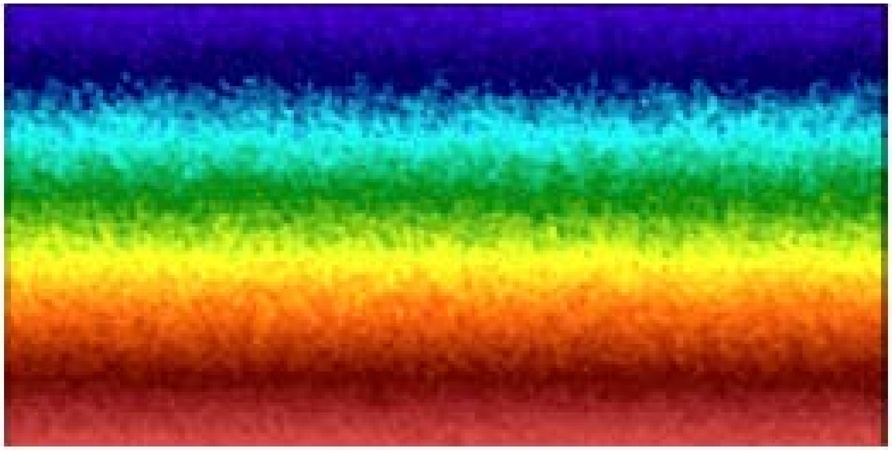


ALG, *Phys. Rev. A* **34** 1454 (1986).

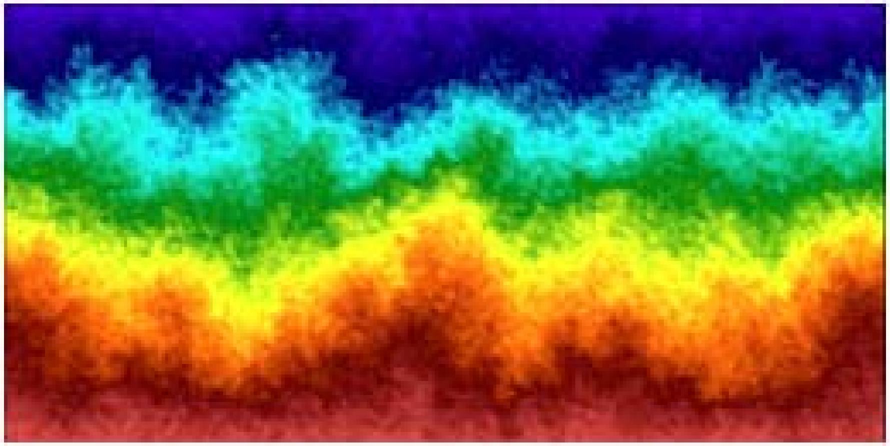
(My first DSMC paper)

Diffusion & Fluctuations

Concentration fluctuations are enhanced when a system is out of equilibrium.



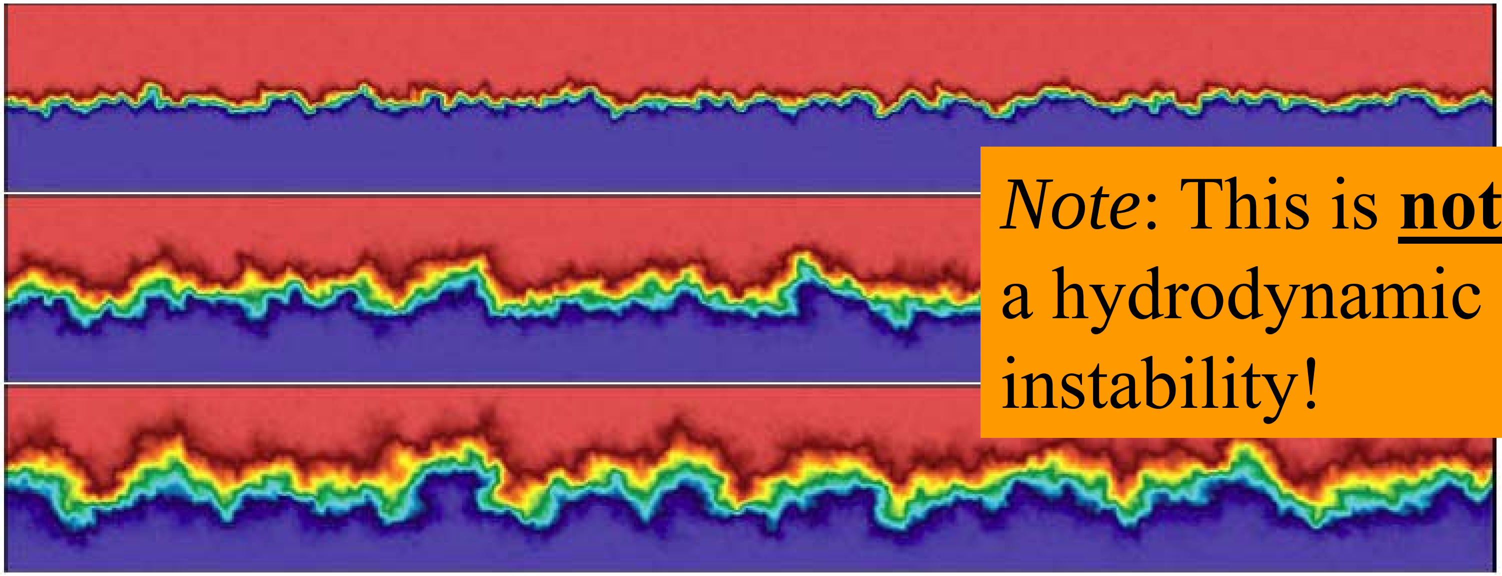
Equilibrium
concentration gradient
(induced by gravity)



Steady-state
concentration gradient
(induced by boundaries)

Giant Fluctuations in Mixing

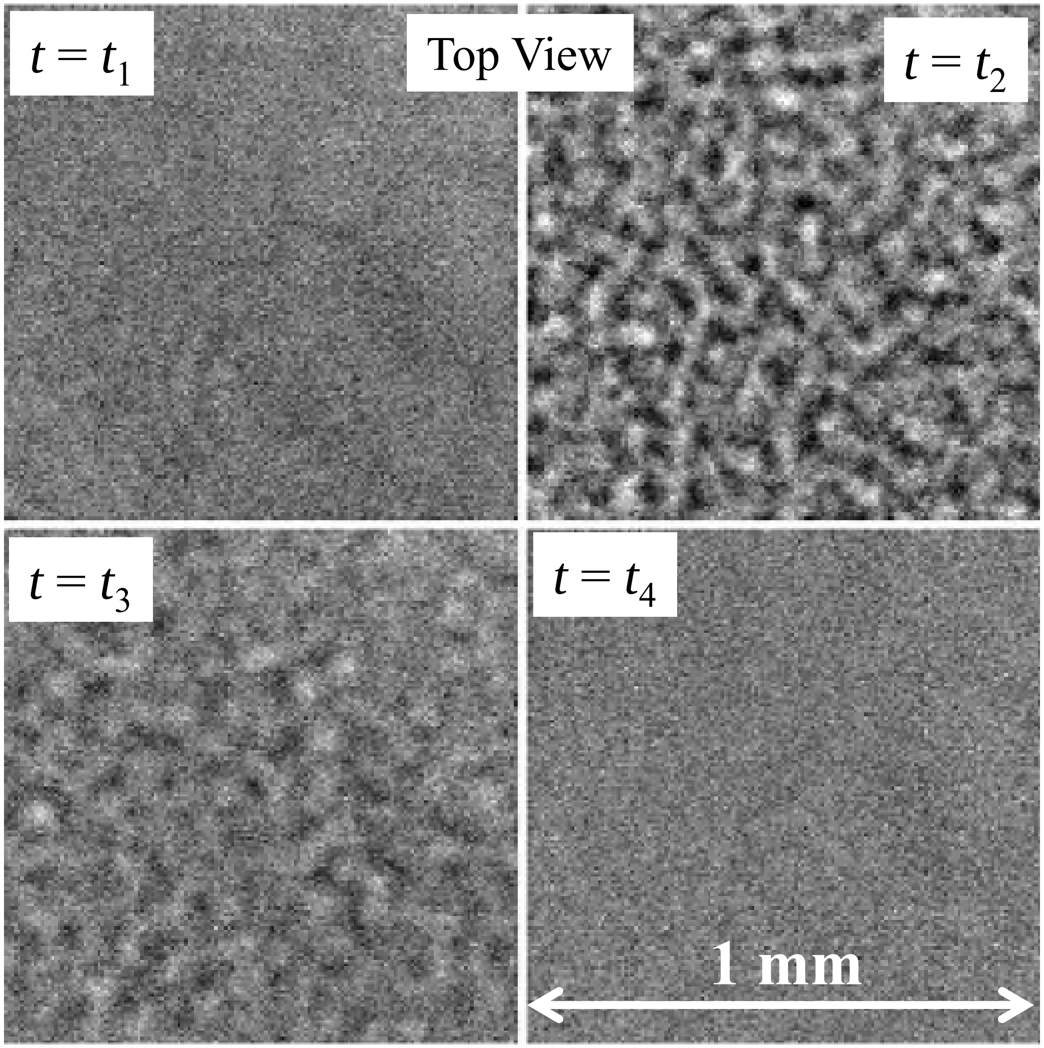
Fluctuations grow large during mixing even when the two species are identical (red & blue).



Note: This is **not** a hydrodynamic instability!

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.

Experimental Observations

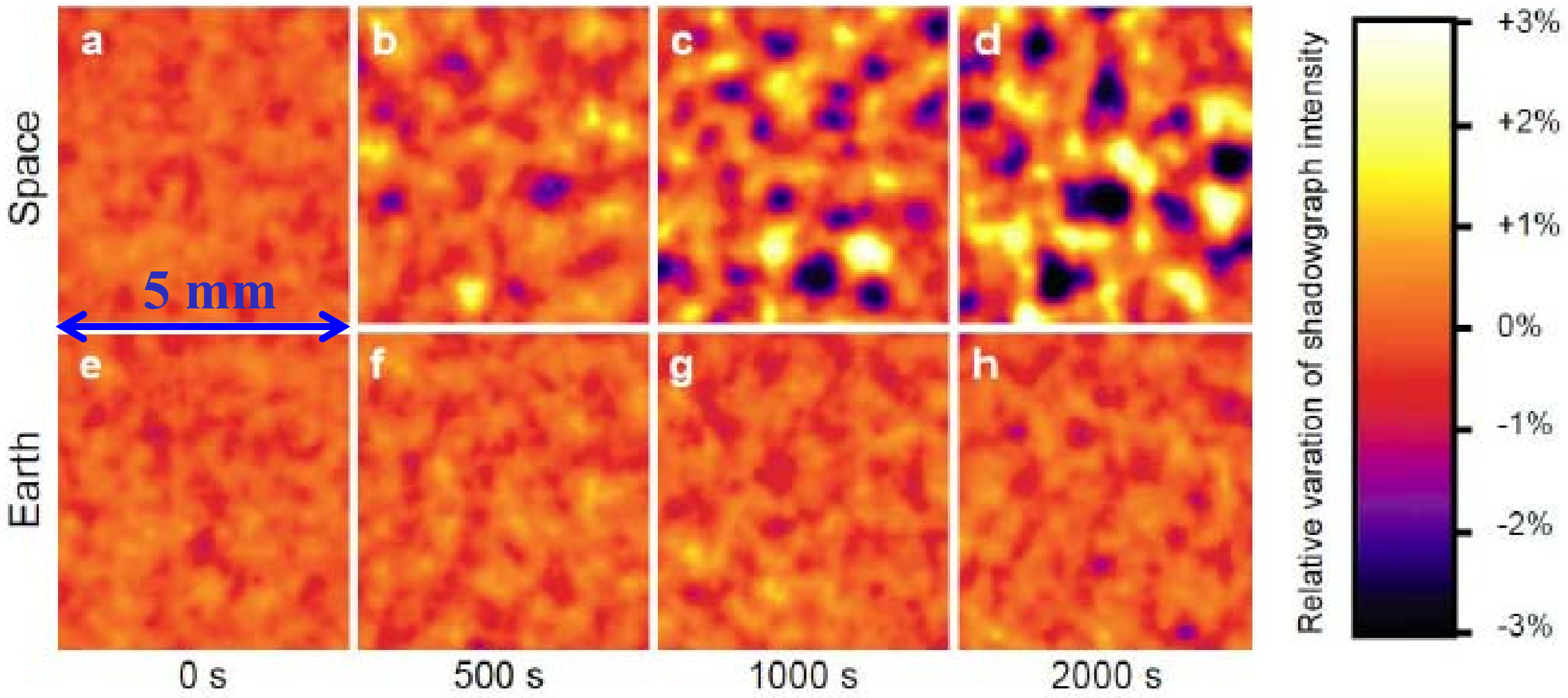


Giant fluctuations in diffusive mixing seen in lab experiments.

Experimental images of light scattering from the interface between two miscible fluids.

Vailati and Giglio,
Nature 390, 262 (1997).

Experimental Observations (cont.)

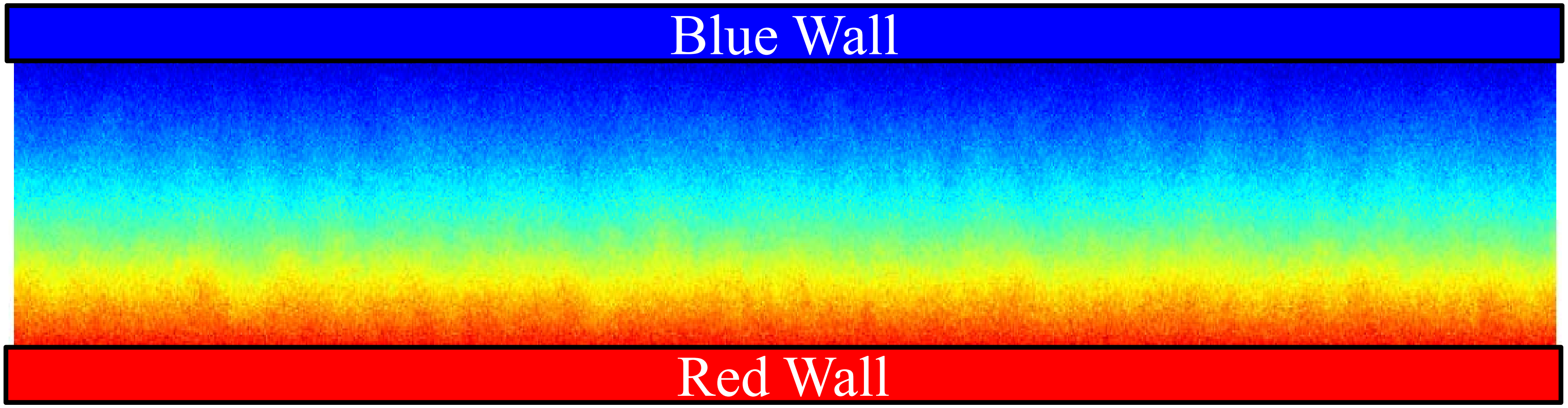


Experiments confirm that concentration fluctuations are reduced by gravity with a cut-off wavelength that is proportional to *g*.

Vailati, et al., Nature Comm., 2:290 (2011).

Diffusion & Fluctuations

Consider a monatomic gas of “red” and “blue” particles at a steady state gradient imposed by wall boundaries.



- Are concentration and velocity fluctuations correlated?
- Do these fluctuations change the transport rate?

Yes and Yes!

Fluctuating Hydrodynamic Theory

Using Landau-Lifshitz fluctuating hydrodynamics in the isothermal, incompressible approximation we may write,

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

$$\rho\mathbf{v}_t = \eta\boldsymbol{\nabla}^2\mathbf{v} - \boldsymbol{\nabla}\pi + \sqrt{2\eta k_BT}\,(\boldsymbol{\nabla}\cdot\mathcal{W}) \quad \text{and} \quad \boldsymbol{\nabla}\cdot\mathbf{v} = 0$$

for the fluctuations of concentration and velocity.

Solving in Fourier space gives the correlation function,

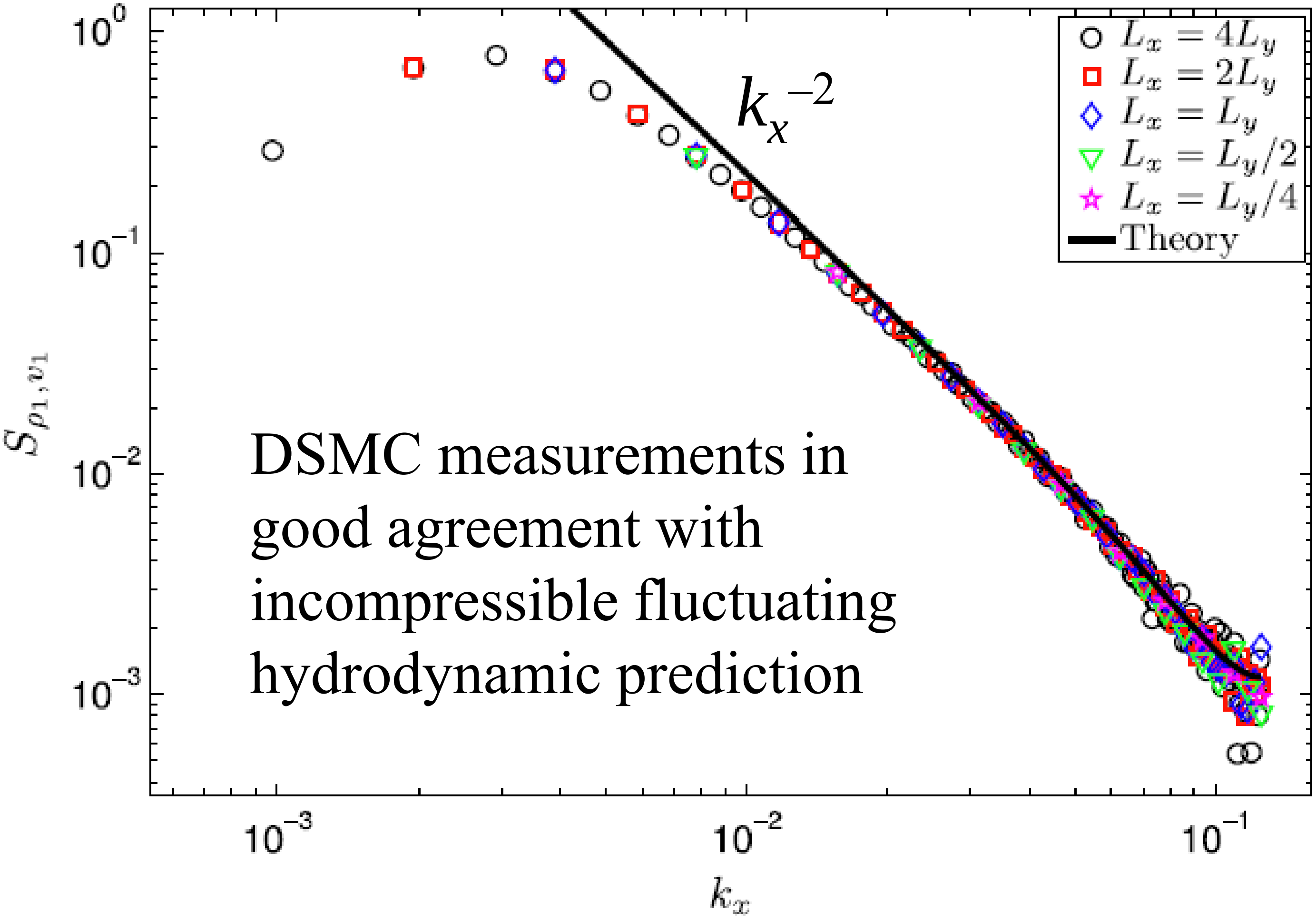
$$\widehat{S}_{c,v_y}(\boldsymbol{k}) = \langle(\widehat{\delta c})(\widehat{v_y^\star})\rangle \sim -\,[k_\perp^2 \boxed{(\nabla_y c)}]\, k^{-4}$$

Note: Linear in gradient

Donev, ALG, de la Fuente, and Bell, J. Stat. Mech. 2011:P06014 (2011)

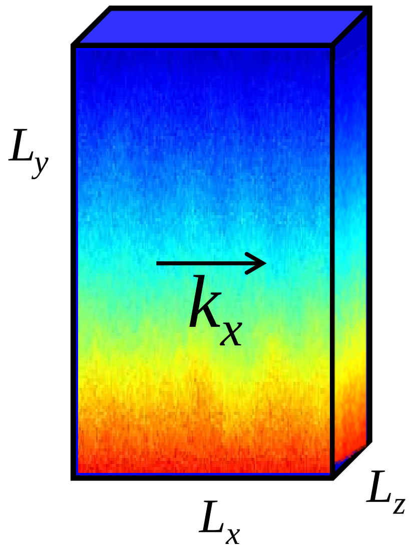
Donev, ALG, de la Fuente, and Bell, Phys. Rev. Lett., 106(20): 204501 (2011)

Concentration-Velocity Correlation



DSMC measurements in good agreement with incompressible fluctuating hydrodynamic prediction

Symbols are DSMC; $k_y = 0$



$L_y = 512\ \lambda$; $L_z = 2\ \lambda$

Diffusion Enhancement

The total mass flux for concentration species is,

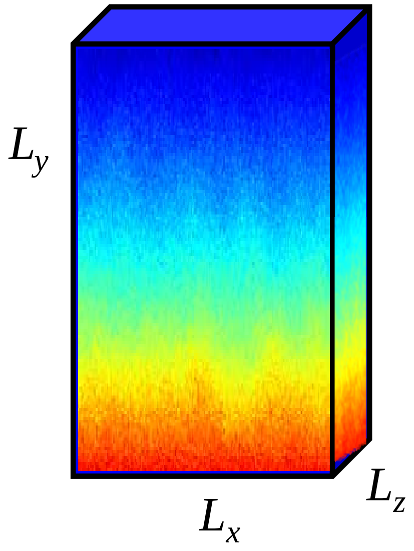
$$\langle \mathbf{j} \rangle \approx (D_0 + \Delta D)\, \nabla c_0 = \left[D_0 - (2\pi)^{-3} \int_{\mathbf{k}} \widehat{S}_{c,v_y}(\mathbf{k})\, d\mathbf{k} \right] \nabla c_0$$

where there are two contributions, the "bare" diffusion coefficient and the contribution due to correlation of fluctuations.

For a slab geometry ($L_z \ll L_x \ll L_y$) we have,

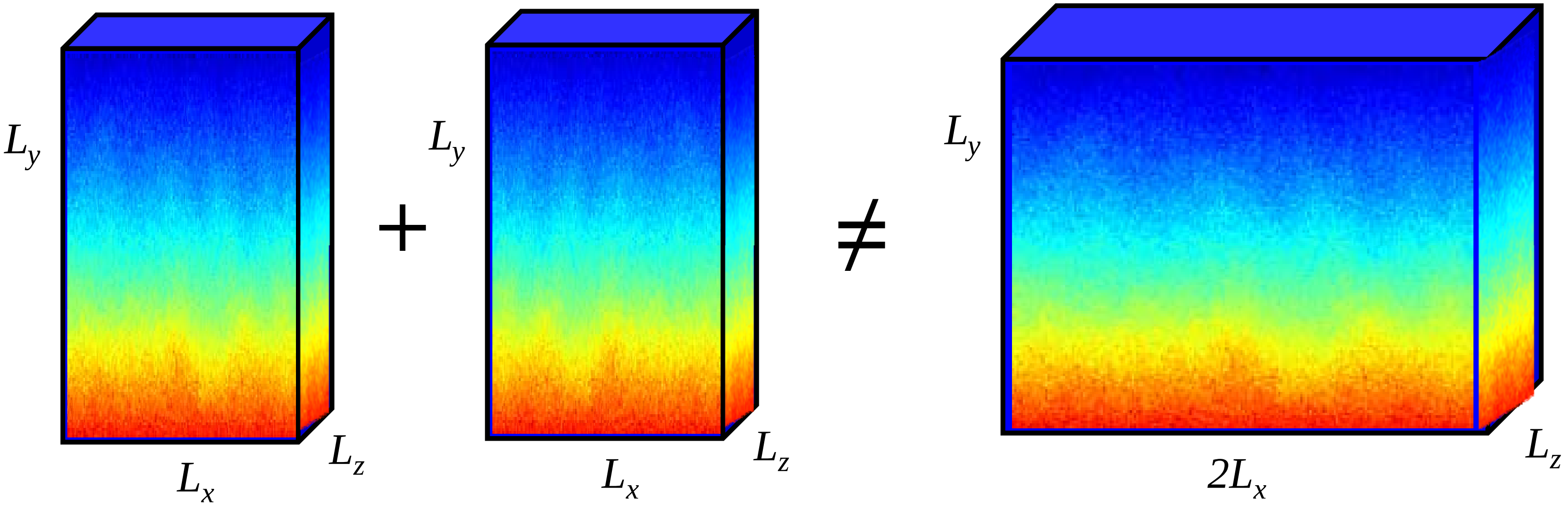
$$\Delta D \approx k_B T \left[4\pi\rho(D_0 + \nu) L_z \right]^{-1} \ln \frac{L_x}{L_{mol}}$$

Notice that diffusion enhancement goes as ln L_x



Enhancement of Diffusion

Spectrum of hydrodynamic fluctuations is truncated at wavenumbers given by the size of the physical system.



The wider system can accommodate long wavelength fluctuations, thus it has enhanced diffusion.

DSMC Measurements

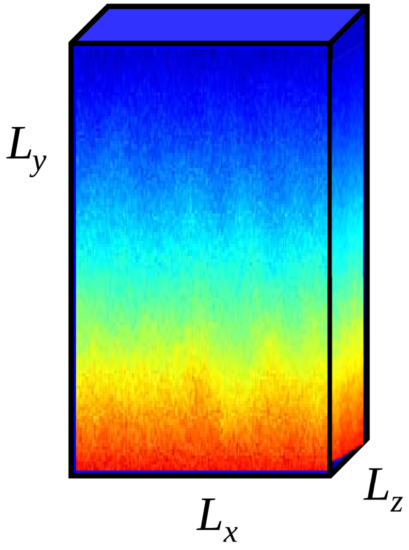
Can separate the contributions to the concentration flux as,

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

$$= D_{\mathrm{eff}} \nabla c \quad = \quad D_0 \nabla c \quad + \quad \Delta D \nabla c$$

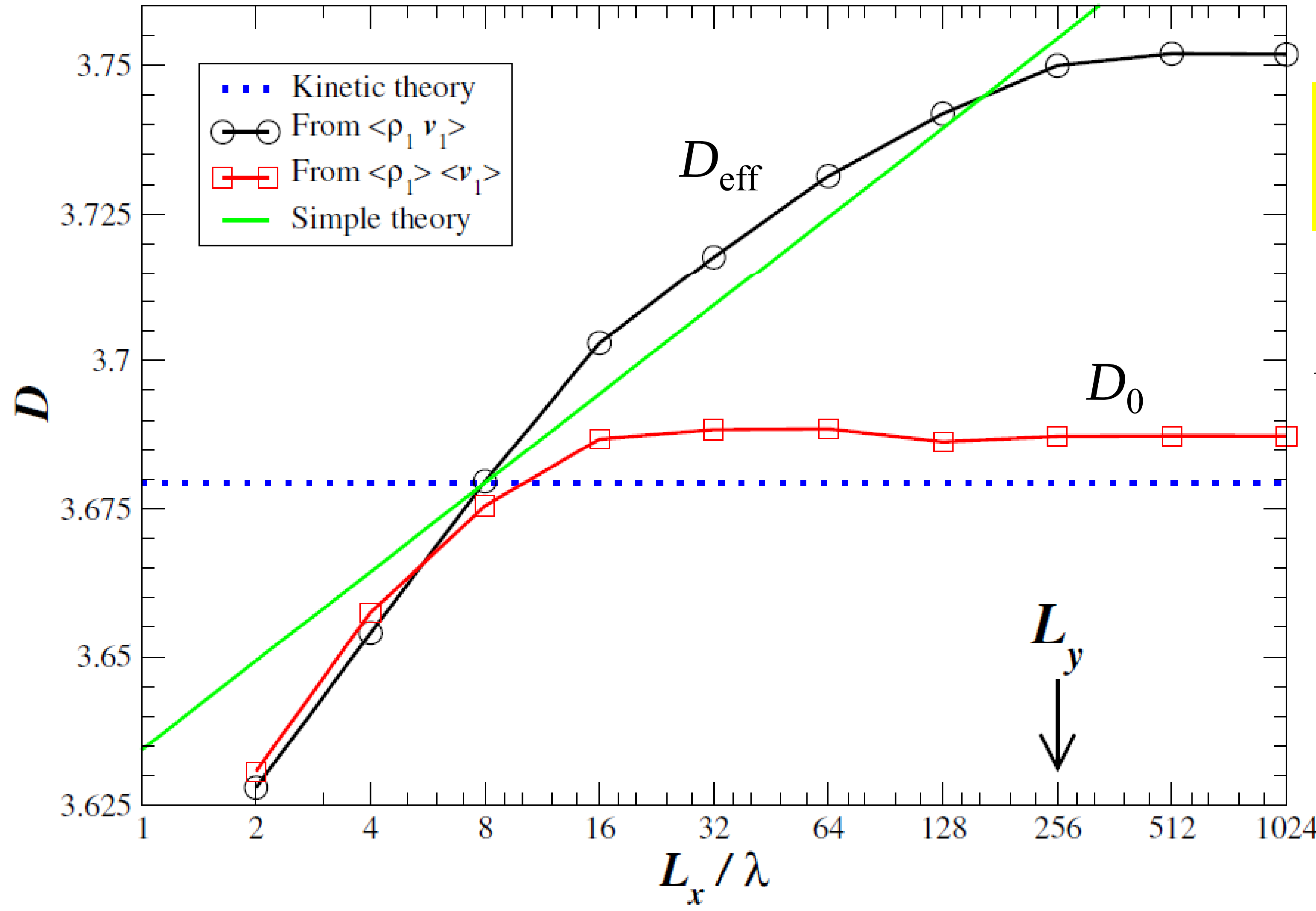
In DSMC we can easily measure

$$\langle \rho_1 \rangle \quad \langle v_{1,y} \rangle \quad \langle \rho_1 v_{1,y} \rangle \quad \text{and} \quad \nabla c$$

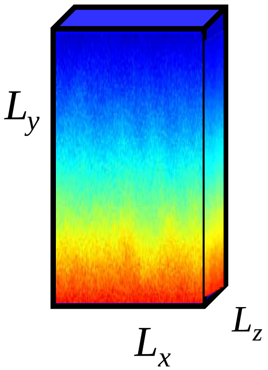


and find the bare diffusion coefficient D_0 and the total effective diffusion coefficient D_{eff}

DSMC Results for D_{eff} and D_0



Quasi-2D
($L_z << L_y$)



$L_y = 256\ \lambda$
$L_z = 2\ \lambda$

Fluctuating Hydrodynamic Solvers

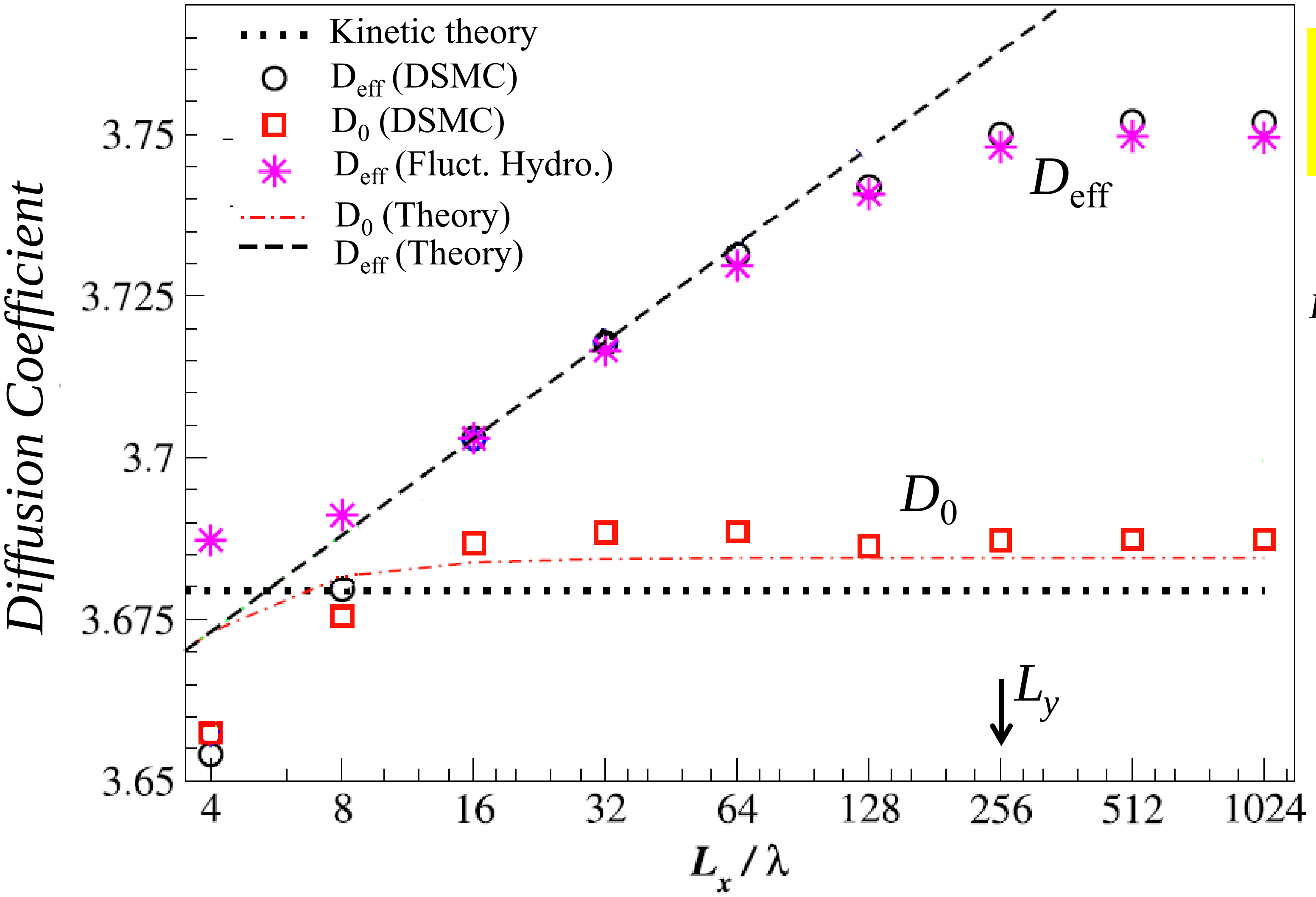
We have developed stochastic CFD schemes for the full hydrodynamic equations and verified them using DSMC simulations.

Using these CFD schemes we can simulate our system and include effects neglected in the simple theory, such as compressibility and temperature fluctuations.

Bell, ALG, and Williams, Phys. Rev. E 76 016708 (2007)

Donev, Vanden-Eijnden, ALG, and Bell, Comm. Applied Math. Comp. Sci., 5 149–197 (2010).

Fluctuating Hydro. Solver Results



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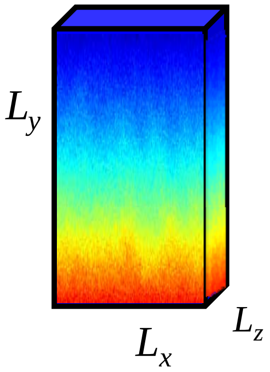

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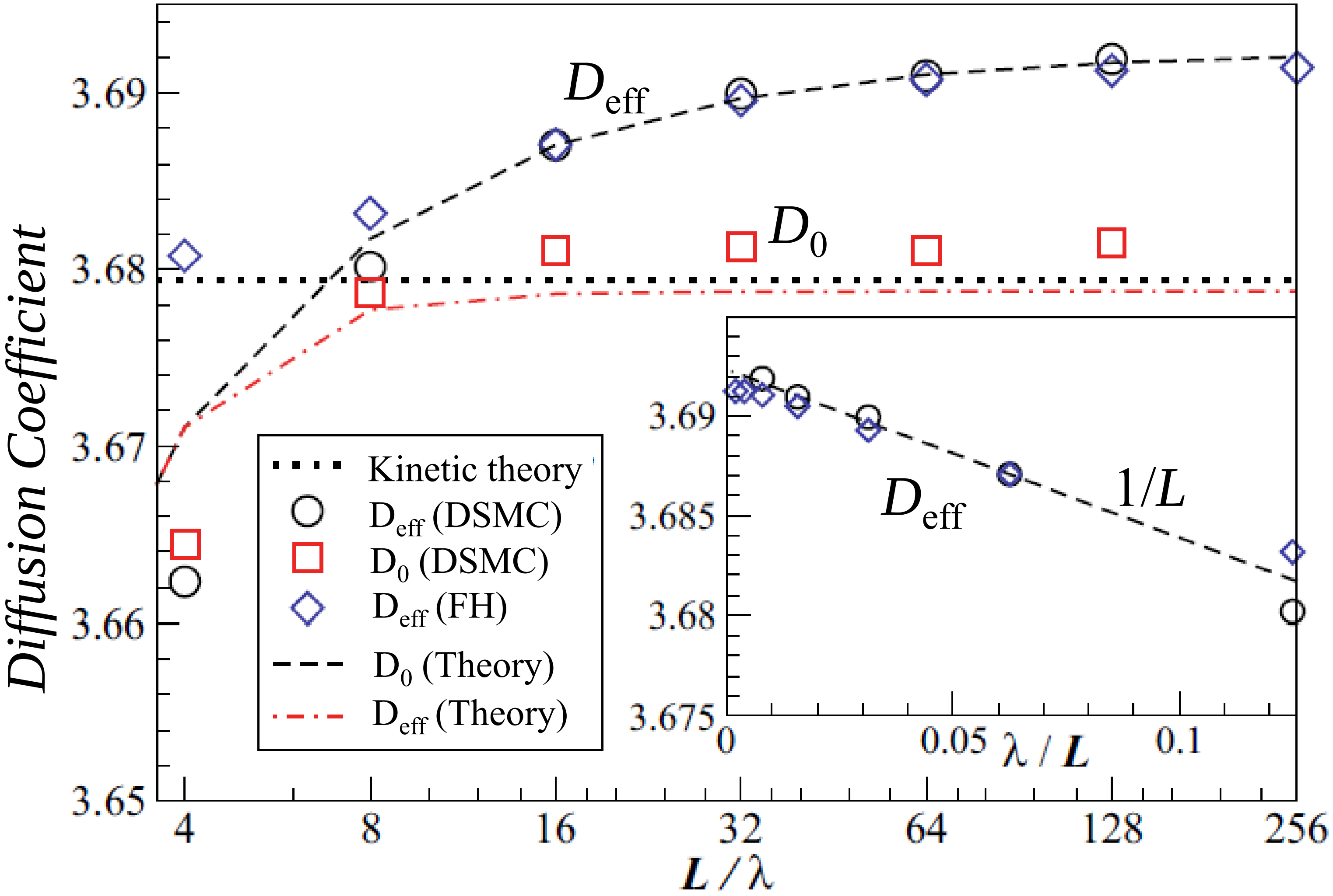
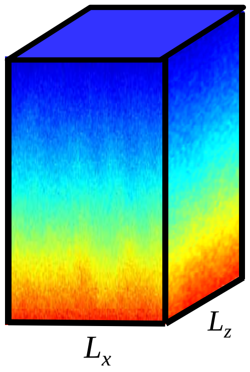
Full 3D Systems



Full 3D ($L_x = L_z$)



$L_y = L_z = L$

ΔD goes as $1/L_0 - 1/L$

Concluding Remarks

- Diffusion enhancement is *very* small in a typical DSMC applications.
- Effect is stronger in liquids and should be possible to measure in MD simulations.
- Enhancement also occurs for viscosity and for thermal conductivity.
- We studied “red/blue” mixture but enhancement occurs in the general case.
- Enhancement is closely related to the long-time tail effect from kinetic theory.

Thank you for your attention and for your participation at ***DSMC11***