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Suppression and Distortion of Non-Equilibrium Fluctuations by Transpiration

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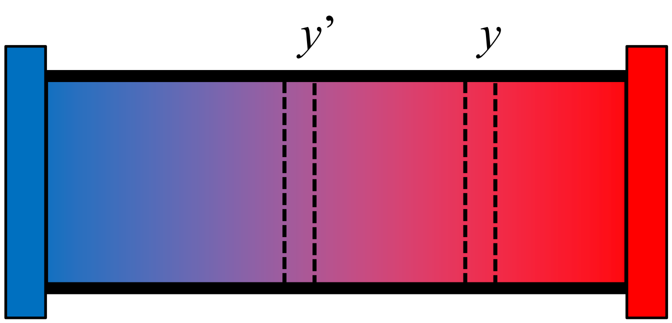
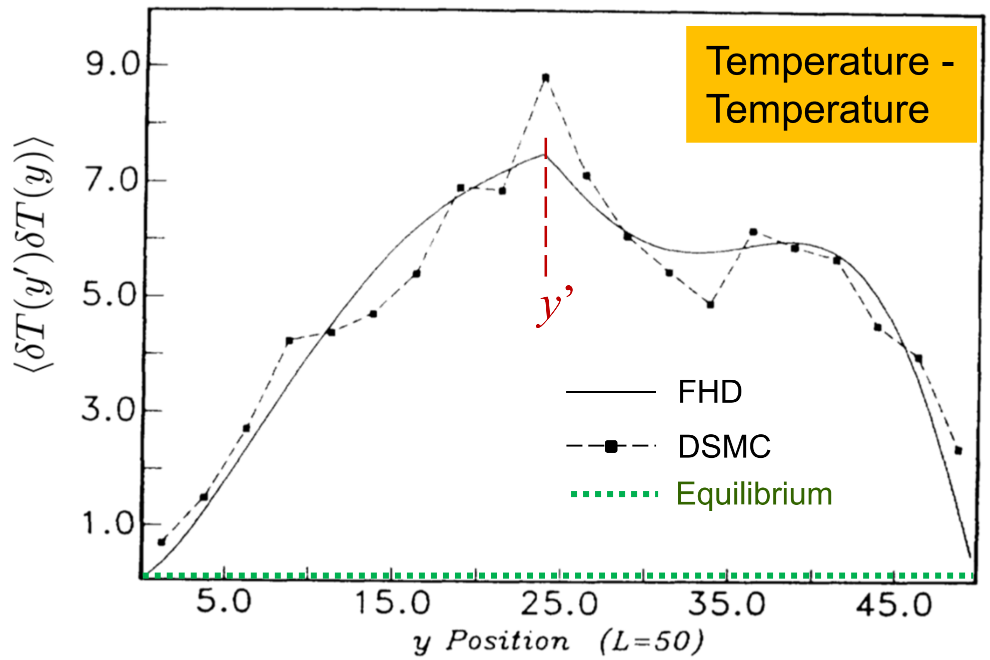
Non-equilibrium Fluctuations

Hydrodynamic fluctuations are long-ranged in a fluid held at a non-equilibrium steady state.

$$\langle \delta\rho(y')\delta v(y)\rangle \propto \nabla T$$
$$\langle \delta T(y')\delta T(y)\rangle \propto (\nabla T)^2$$





M. Malek Mansour, ALG, G. Lie and E. Clementi, *Phys. Rev. Lett.* **58** 874 (1987).

Fluctuating Hydrodynamics (FHD)

Landau and Lifshitz added stochastic flux terms into Navier-Stokes to model spontaneous fluctuations in fluids.

$$\frac{\partial}{\partial t}(\rho) = -\nabla\cdot(\rho\mathbf{u}),$$

$$\frac{\partial}{\partial t}(\rho\mathbf{u}) = -\nabla\cdot(\rho\mathbf{u}\otimes\mathbf{u}) - \nabla P - \nabla\cdot\left[\mathbf{\Pi}+\widetilde{\mathbf{\Pi}}\right],$$

$$\frac{\partial}{\partial t}(\rho E) = -\nabla\cdot(\rho\mathbf{u}E + P\mathbf{u}) - \nabla\cdot\left[\mathbf{Q}+\widetilde{\mathbf{Q}}\right] - \nabla\cdot\left(\left[\mathbf{\Pi}+\widetilde{\mathbf{\Pi}}\right]\cdot\mathbf{u}\right).$$

[Stress tensor]

[Heat flux]

L. D. Landau E. M. Lifshitz, *Fluid Mechanics*, 1st Ed., Pergamon (1959).
J. M. Ortiz de Zarate and J. V. Sengers, *Hydrodynamic Fluctuations in Fluids and Fluid Mixtures*, Elsevier (2006).

Dissipative Fluxes in FHD

The deterministic stress tensor and heat flux take their standard linear forms (Stokes and Fourier laws),

$$\Pi_{ij} = -\eta \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) + \delta_{ij} \left(\frac{2}{3} \eta \nabla \cdot \mathbf{u} \right), \qquad \text{and} \qquad \mathbf{Q} = -\kappa \nabla T$$

Stochastic stress tensor and heat flux are independent noises, white in space and time, with zero mean and variances,

$$\langle \widetilde{\Pi}_{ij}(\mathbf{r}, t) \widetilde{\Pi}_{kl}(\mathbf{r}', t') \rangle = 2 k_B \eta T \left[(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - \tfrac{1}{3}\delta_{ij}\delta_{kl} \right] \delta(t - t')\delta(\mathbf{r} - \mathbf{r}'),$$

$$\langle \widetilde{Q}_i(\mathbf{r}, t) \widetilde{Q}_j(\mathbf{r}', t') \rangle = 2 k_B \kappa T^2 \delta_{ij} \delta(t - t')\delta(\mathbf{r} - \mathbf{r}').$$

Finite Volume Stochastic PDE solver

Write the FHD equations as a stochastic PDE:

$$\frac{\partial}{\partial t}\mathbf{U} = -\nabla \cdot \mathbf{F} - \nabla \cdot \mathbf{D} - \nabla \cdot \tilde{\mathbf{S}}$$

Time integration: $\frac{\partial}{\partial t}\mathbf{U} = \frac{\partial}{\partial t}(\ \rho,\ \rho\mathbf{u},\ \rho E\ )$

Hyperbolic: $\mathbf{F} = (\ \rho\mathbf{u},\ \rho\mathbf{u}\mathbf{u} + P\mathbf{I},\ \rho\mathbf{u}E + \mathbf{u}P\ )$

Parabolic: $\mathbf{D} = (\ 0,\ \mathbf{\Pi},\ \mathbf{Q} + \mathbf{u} \cdot \mathbf{\Pi}\ )$

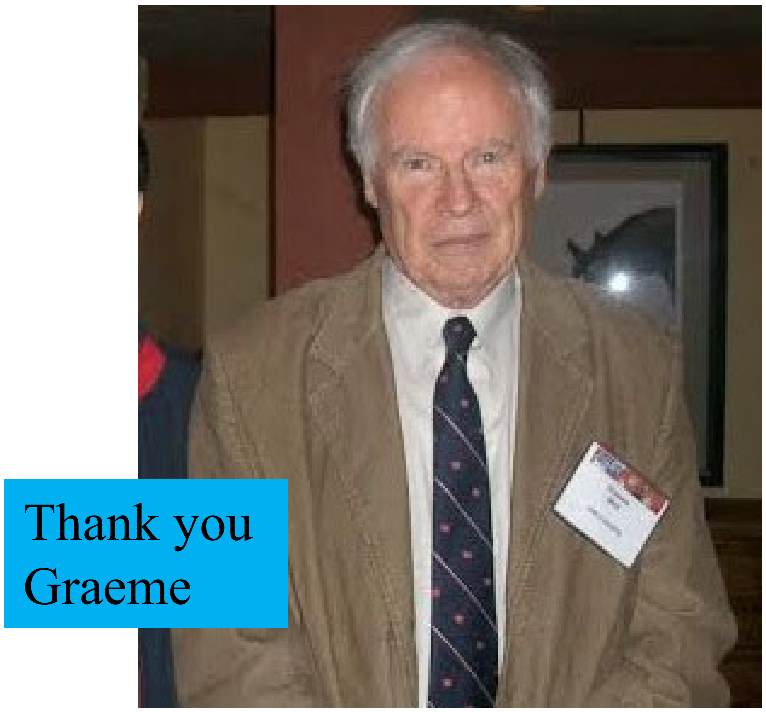
Stochastic: $\tilde{\mathbf{S}} = (\ 0,\ \tilde{\mathbf{\Pi}},\ \tilde{\mathbf{Q}} + \mathbf{u} \cdot \tilde{\mathbf{\Pi}}\ )$

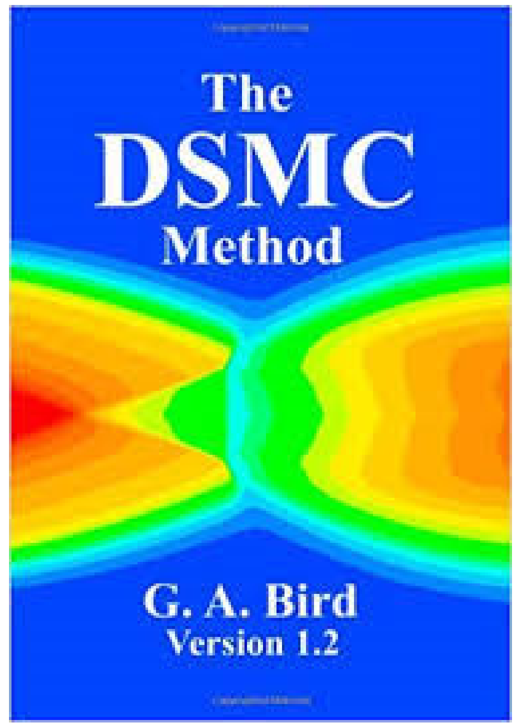
- Three-stage Runge-Kutta
- Four point centered
- Two point centered
- Weighted 2 point centered

J.B. Bell, ALG, and S. Williams, *Physical Review E* **76** 016708 (2007).
A. Donev, E. Vanden-Eijnden, ALG, and J.B. Bell, *Comm. Applied Math. Comp. Sci.* **5** 149–197 (2010).

Direct Simulation Monte Carlo (DSMC)

DSMC: You know it. You love it.





GRAEME A. BIRD COMMEMORATIVE ISSUE
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Guest Editor: M. A. Gallis

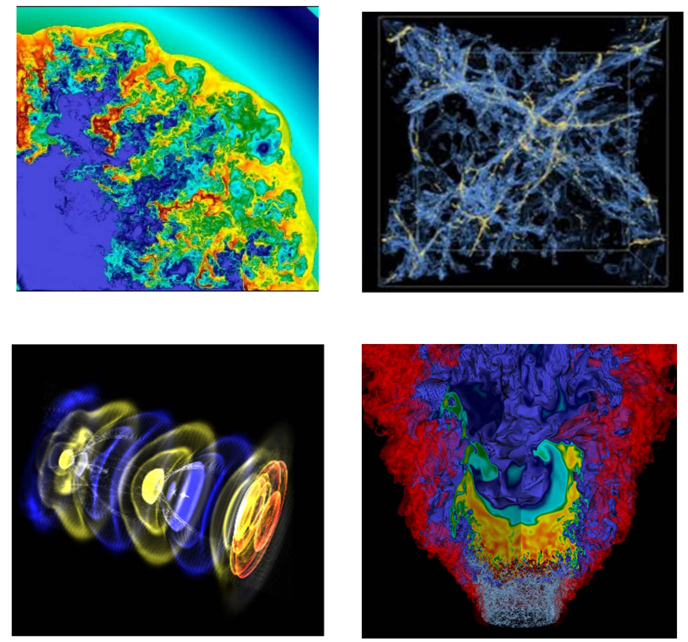
AMReX

Our FHD and DSMC codes are both written using AMReX, a block-structured adaptive mesh refinement framework.

Supports hierarchical mesh and particle data with embedded boundary capability.

AMReX is used for a wide range of applications including accelerator modeling, astrophysics, combustion, cosmology, multiphase flow, phase field modeling, etc.

Examples of AMReX applications

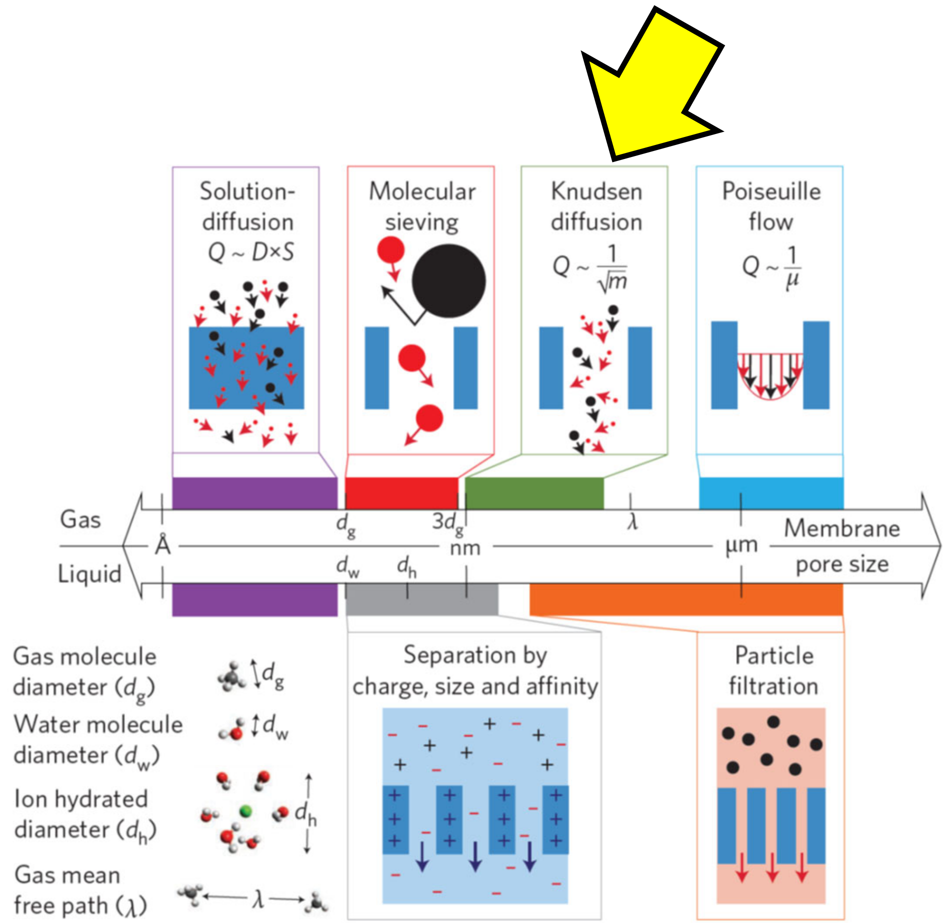


www.github.com/AMReX-Codes/amrex

Porous Membranes

Effusion (Knudsen diffusion) is gas transport through a membrane with pore sizes roughly between the mean free path and the molecule diameter.

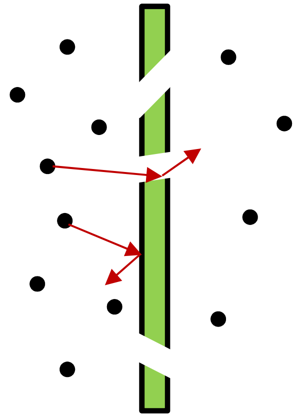
For standard conditions this range in pore size is between 100 nm to 2 nm.

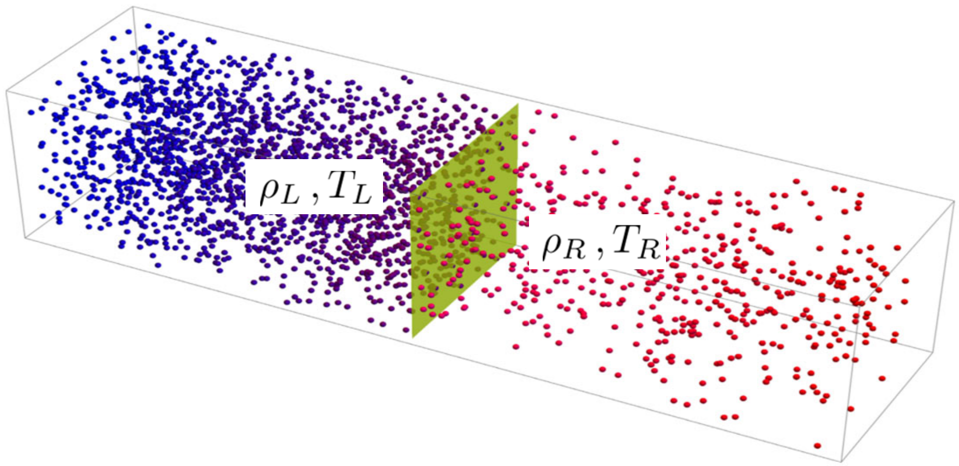


L. Wang, et al., *Nature Nanotechnology* **12** 509–522 (2017)

Transport by Effusion

Molecules reaching the interface cross it with probability f.





The mean fluxes of mass and energy are,

$$\langle J_M \rangle = \frac{f\,A\,k_B^{1/2}}{\sqrt{2\pi m}} \left(\rho_L T_L^{1/2} - \rho_R T_R^{1/2} \right), \qquad \langle J_{\mathcal{E}} \rangle = \frac{2f\,A\,k_B^{3/2}}{m\sqrt{2\pi m}} \left(\rho_L T_L^{3/2} - \rho_R T_R^{3/2} \right).$$

Langevin Model for Effusion Membrane

In FHD we model the mass and energy crossing the membrane with the Langevin equations,

$$\frac{d}{dt}M = \langle J_M \rangle + \widetilde{J}_M, \qquad \frac{d}{dt}\mathcal{E} = \langle J_\mathcal{E} \rangle + \widetilde{J}_\mathcal{E},$$

where the white noises have variances and covariances,

$$\langle \widetilde{J}_M(t)\widetilde{J}_M(t') \rangle = \frac{mfA\,k_B^{1/2}}{\sqrt{2\pi m}} \left(\rho_R T_R^{1/2} + \rho_L T_L^{1/2} \right) \delta(t-t')$$

$$\langle \widetilde{J}_\mathcal{E}(t)\widetilde{J}_\mathcal{E}(t') \rangle = \frac{6fA\,k_B^{5/2}}{m\sqrt{2\pi m}} \left(\rho_R T_R^{5/2} + \rho_L T_L^{5/2} \right) \delta(t-t'),$$

$$\langle \widetilde{J}_\mathcal{E}(t)\widetilde{J}_M(t') \rangle = \frac{2fA\,k_B^{3/2}}{\sqrt{2\pi m}} \left(\rho_R T_R^{3/2} + \rho_L T_L^{3/2} \right) \delta(t-t').$$

Note: Mass and energy noises are correlated

Langevin Fluxes in FHD Solver

Calculate mass and energy passing through membrane as:

- Calculate a flux correlation coefficient,

$$r = \frac{\langle J_{\delta\mathcal{E}\delta M}\rangle}{\sqrt{\langle J_{\delta\mathcal{E}\delta\mathcal{E}}\rangle\langle J_{\delta M\delta M}\rangle}}.$$

- Generate correlated Gaussian random variables,

$$\mathcal{G}_3 = \mathcal{G}_1 r + \sqrt{1-r^2}\mathcal{G}_2$$

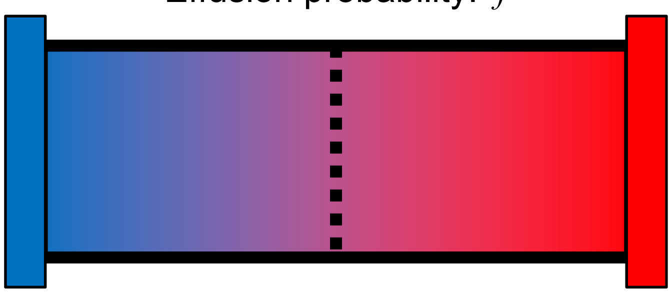
- Mass and energy crossing during a timestep are,

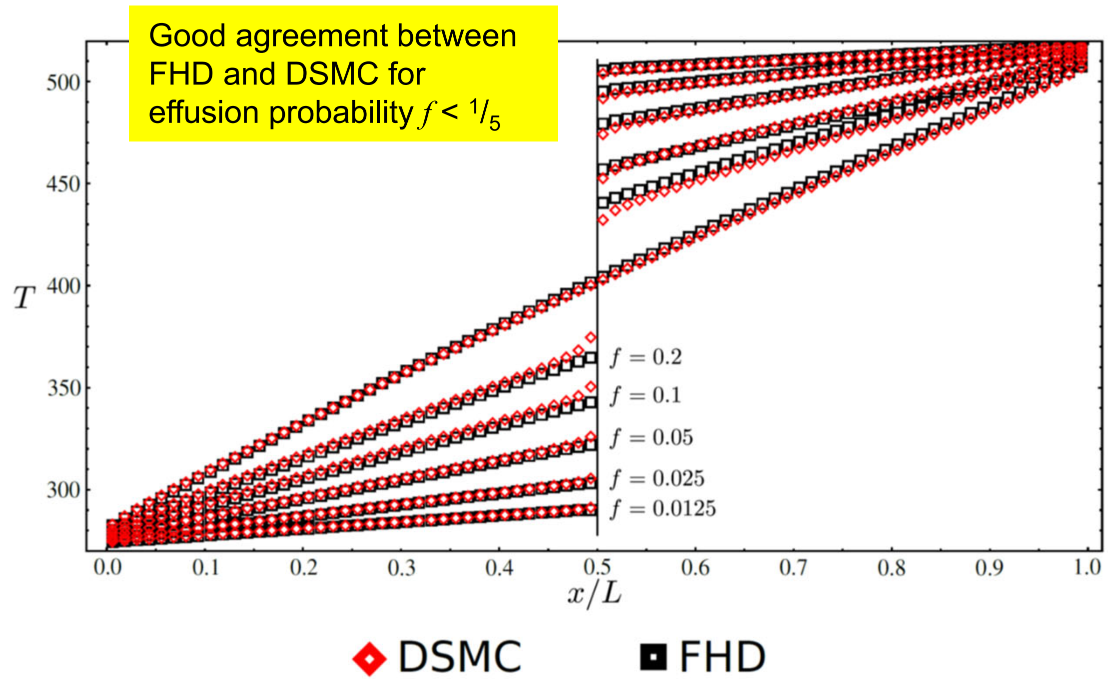
$$\begin{aligned} F_M &= \Delta t\langle J_M\rangle + \sqrt{\Delta t\langle J_{\delta M\delta M}\rangle}\ \mathcal{G}_1 \\ F_{\mathcal{E}} &= \Delta t\langle J_{\mathcal{E}}\rangle + \sqrt{\Delta t\langle J_{\delta\mathcal{E}\delta\mathcal{E}}\rangle}\ \mathcal{G}_3 \end{aligned}$$

Temperature Profiles

Simulated a dilute gas with a temperature gradient in a system bisected by a porous effusion membrane.

Porous Membrane
Effusion probability: f



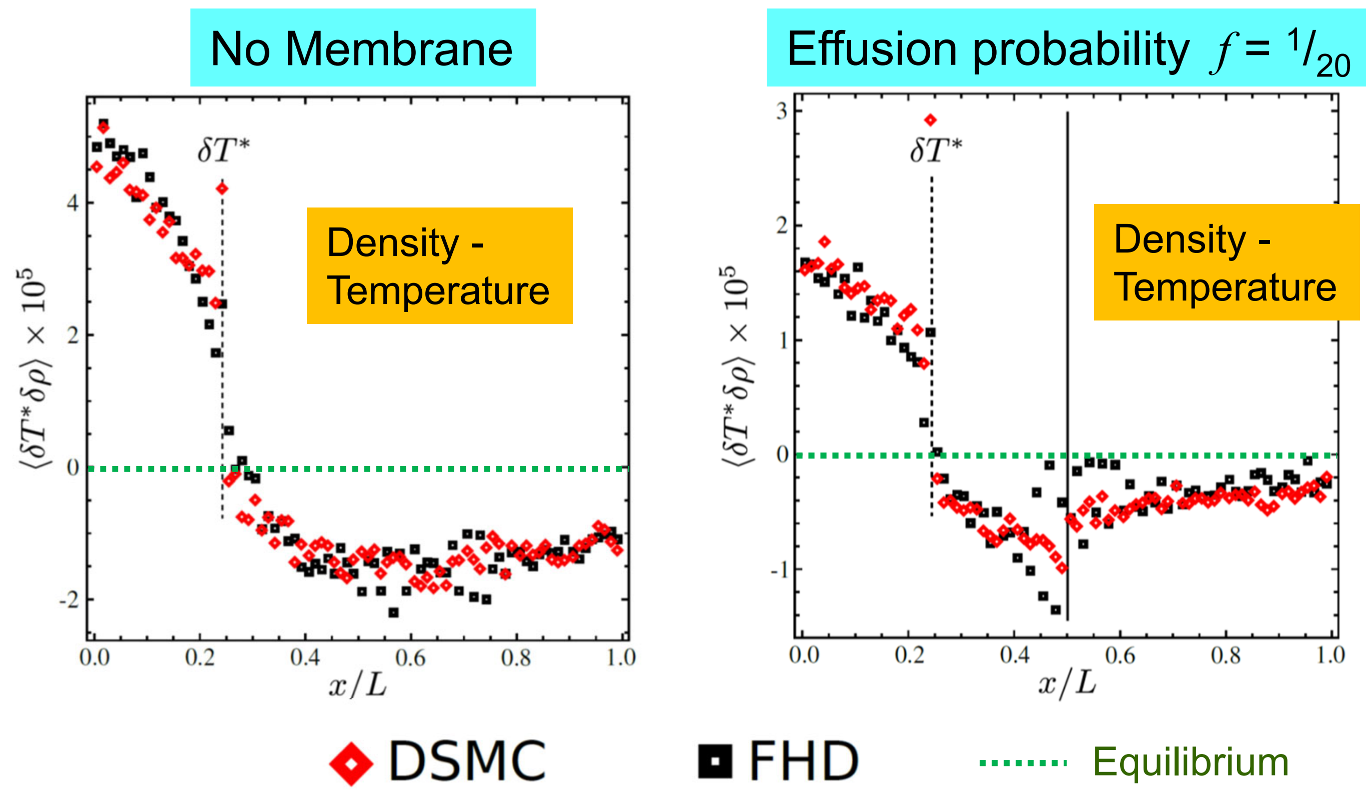


D. Ladiges, A. Nonaka, J.B. Bell, and ALG, *Physics of Fluids* **31**, 052002 (2019)

Correlations of Fluctuations

Temperature gradient is reduced by ~3 and so is density-temperature correlation

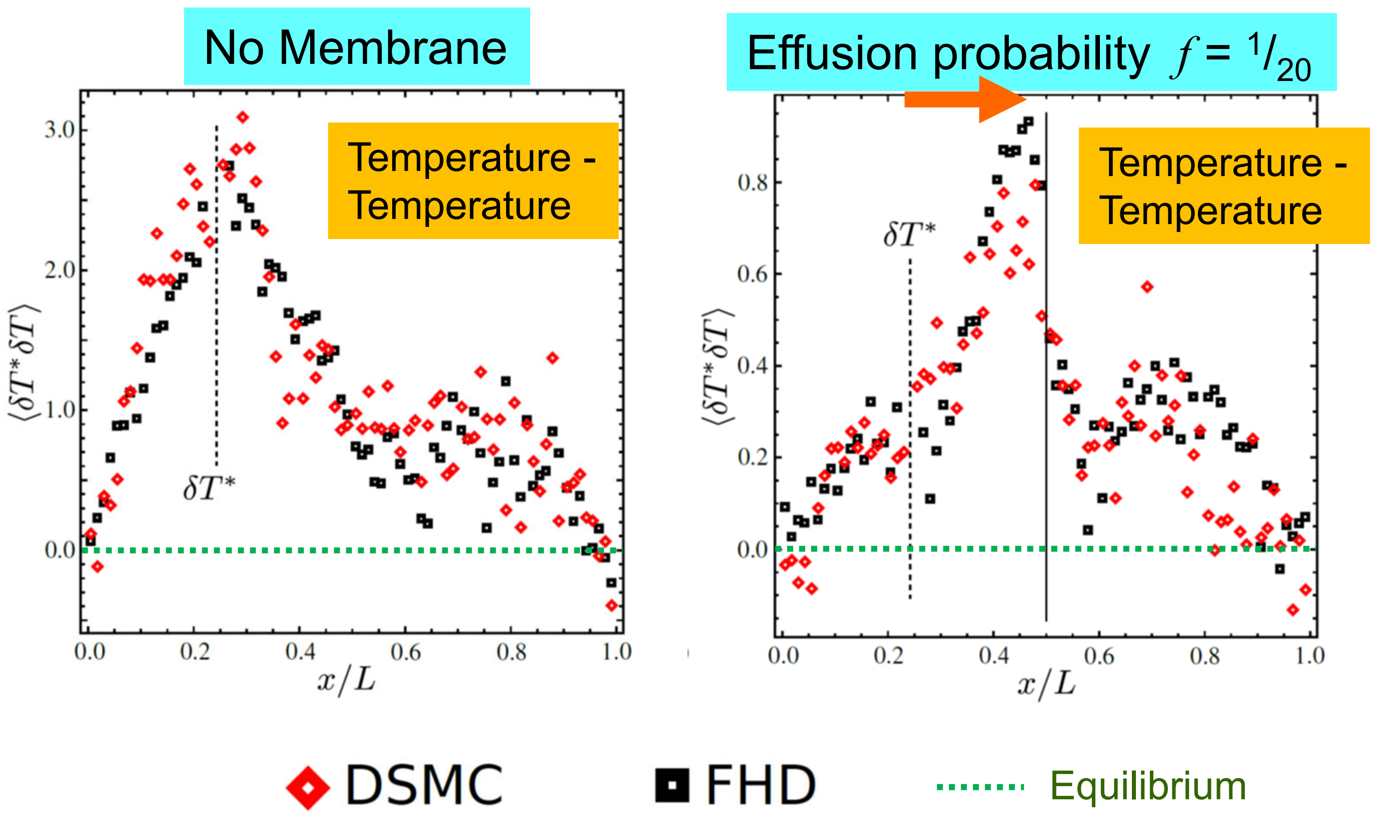
Correlation persists across the membrane



Correlations of Fluctuations

Temperature-temperature correlation is reduced but persists across the membrane

Correlation peak is significantly shifted towards the membrane



Master Equation Interface Model

Tested a Master equation formulation using Gillespie (SSA) algorithm in FHD.

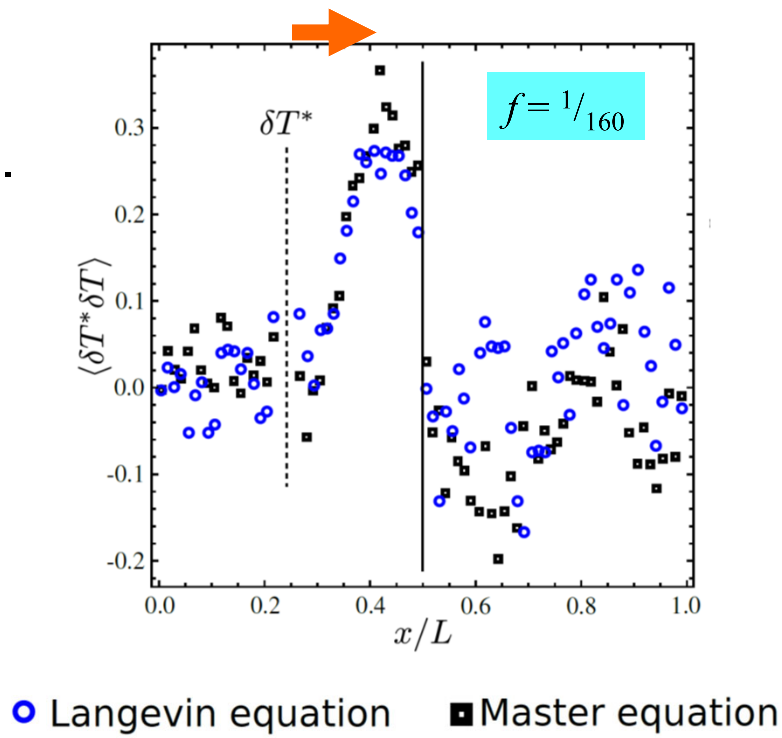
$$\tau_\mathrm{e} = \frac{m}{\rho f A}\sqrt{\frac{2\pi m}{k_B T}}$$

Mean waiting time for crossings

$$P_\epsilon(\epsilon) = \frac{\epsilon}{k_B^2 T^2}\, e^{-\epsilon/(k_B T)}$$

Distribution of molecule energies

The Langevin and Master equation models produced equivalent results.



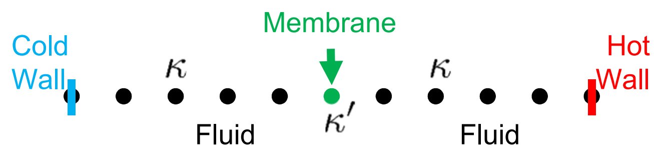
Stochastic Heat Equation

To investigate the shift in the peak we consider the linearized stochastic heat equation,

$$\rho c_v \frac{\partial}{\partial t}\delta T(y,t) = \frac{\partial}{\partial y}\kappa(y)\frac{\partial}{\partial y}\delta T - \frac{\partial}{\partial y}\widetilde{Q} \quad \text{with} \quad \begin{aligned}&\langle \widetilde{Q}(y,t)\widetilde{Q}(y',t')\rangle = \\ &\quad 2k_B\,\kappa(y)T_0(y)^2\;\delta(t-t')\delta(y-y').\end{aligned}$$

Discretizing in space, $\mathbf{U} = [\delta T_1, \ldots, \delta T_N]$

$$\frac{\partial}{\partial t}\mathbf{U} = -\mathbf{A}\mathbf{U} + \mathbf{B}\tilde{\boldsymbol{\xi}}$$



For this Ornstein-Uhlenbeck process we find the covariance $\sigma_{i,j} = \langle \delta T_i \delta T_j \rangle$ by solving

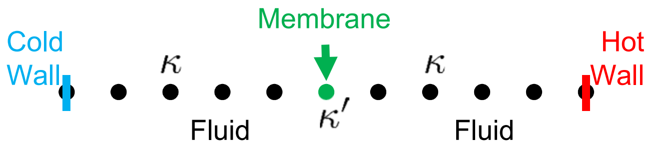
$$\mathbf{A}\boldsymbol{\sigma} + \boldsymbol{\sigma}\mathbf{A}^{\mathrm{T}} = \mathbf{B}\mathbf{B}^{\mathrm{T}}$$

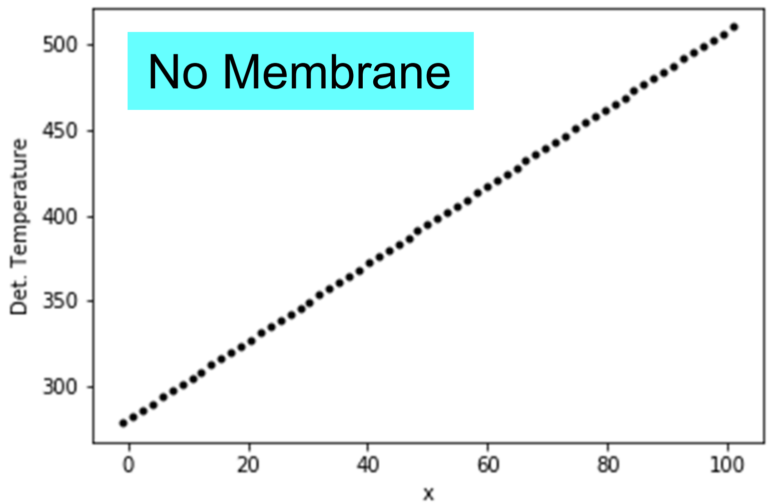
using linear algebra (e.g., by numerical relaxation)

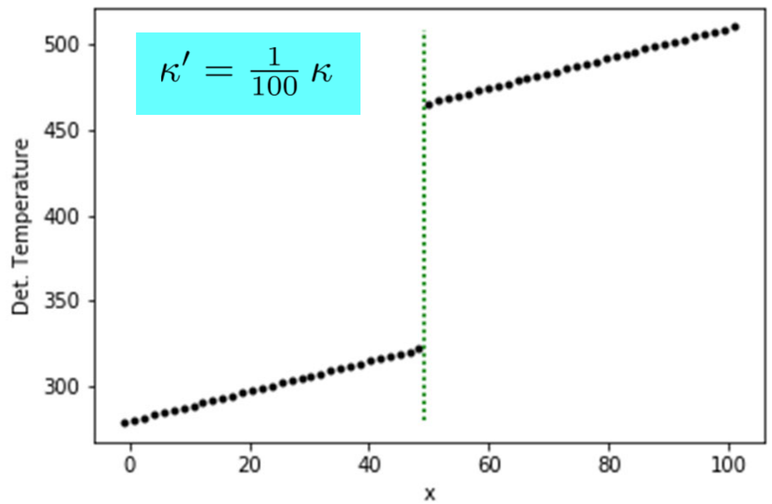
D. Ladiges, J.B. Bell, and ALG, in preparation (2019)

Temperature Profile

Deterministic temperature profiles are qualitatively similar to FHD and DSMC results

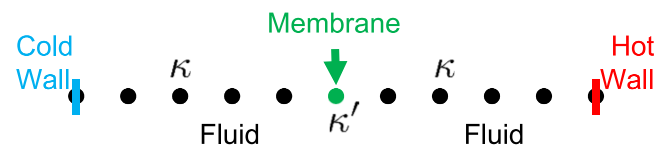


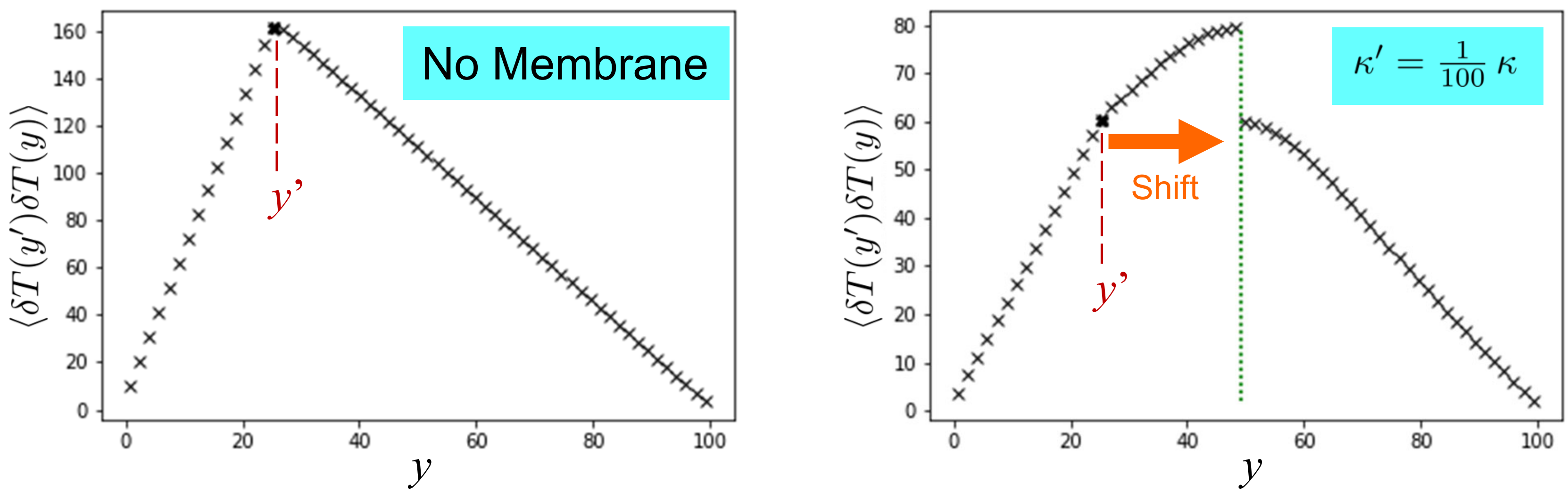




Correlations of Fluctuations

Temperature-temperature correlation is qualitatively similar to the FHD and DSMC result.





Summary & Future Work

Summary:

- Long range correlations persist through an effusive interface.
- Reduced magnitude largely due to change in ∇T.
- Distortion (peak shift) predicted by the stochastic heat equation.
- FHD can simulate gas transpiration; faster than DSMC.

Future work:

- Shear gradient; Concentration gradient
- Correlations parallel to the membrane
- Molecular sieving
- Ion transport in electrolytes
- Active transport (transport & chemistry)

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