Direct Simulation Monte Carlo & Hydrodynamic Fluctuations

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Hydrodynamic Fluctuations in Soft Matter Simulations

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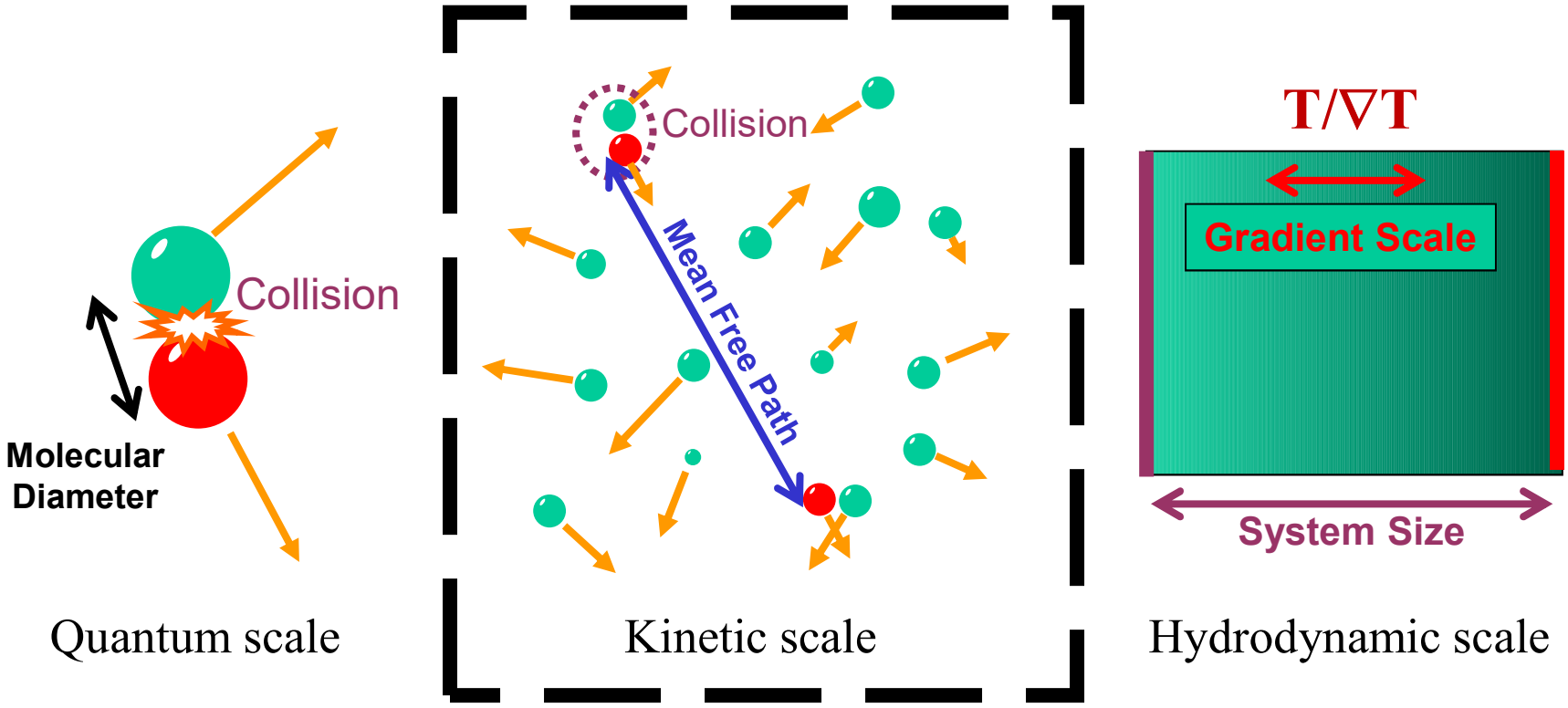
Part I
Direct Simulation Monte Carlo

Part II
Hydrodynamic Fluctuations

Physical Scales for Dilute Gases



DSMC is optimal for simulations at the kinetic scale

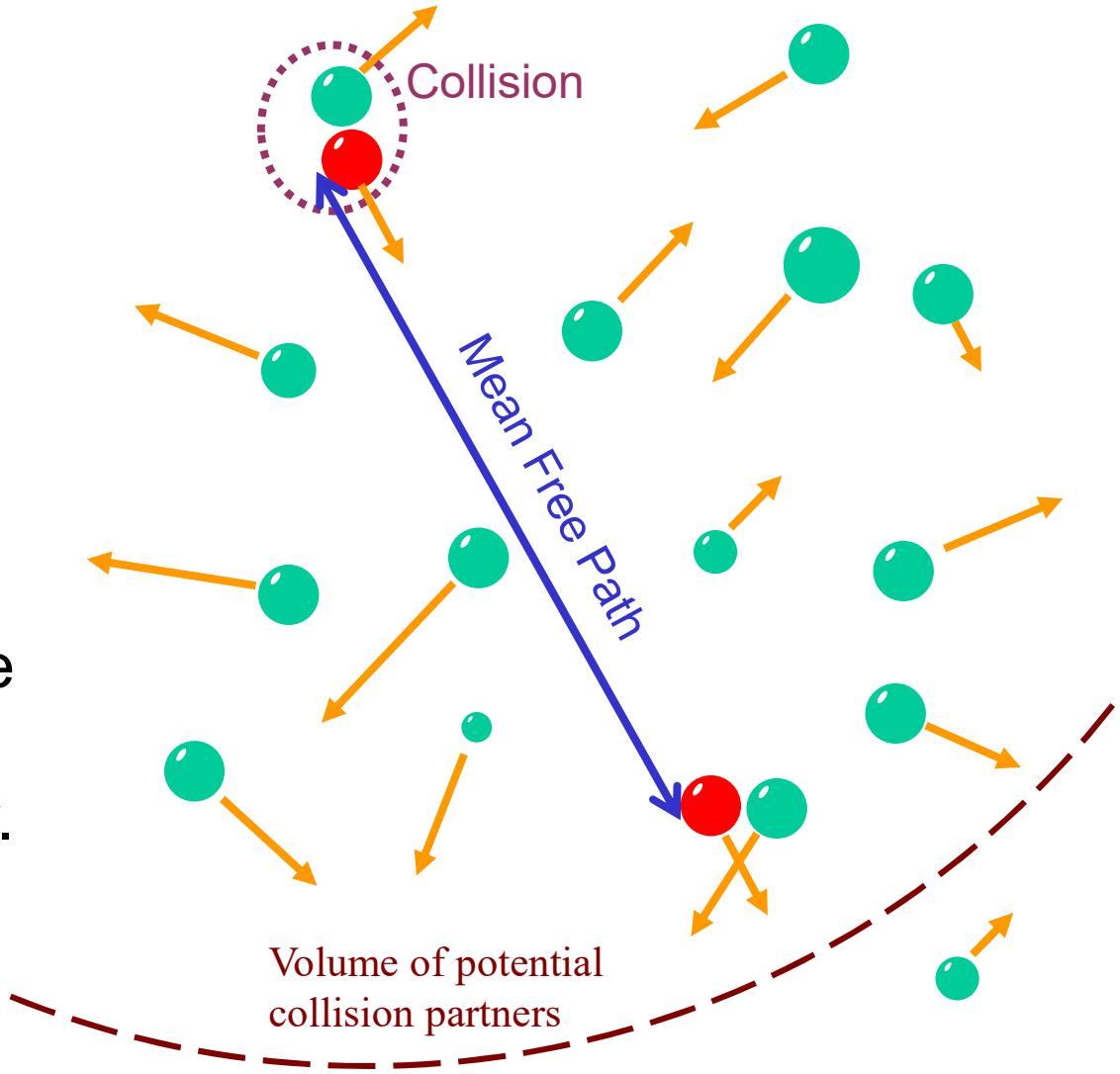
Molecular Dynamics & Dilute Gases

Molecular dynamics inefficient for simulating dilute gasses.

Relevant time scale is mean free time but MD time step limited by time of collision.

DSMC time step is large because collisions are evaluated stochastically.

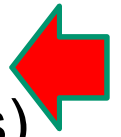
6000 molecules per cubic mean free path at STP



Direct Simulation Monte Carlo

Development of DSMC

- DSMC developed by Graeme Bird (late 60's)
- Popular in aerospace engineering (70's)
- Variants & improvements (early 80's)
- Applications in physics & chemistry (late 80's)
- Used for micro/nano-scale flows (early 90's)
- Extended to dense gases & liquids (late 90's)
- Used for granular gas simulations (early 00's)
- Multi-scale modeling of complex fluids (late 00's)

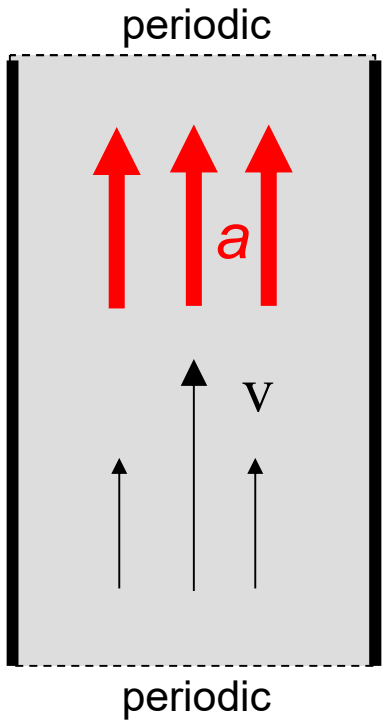


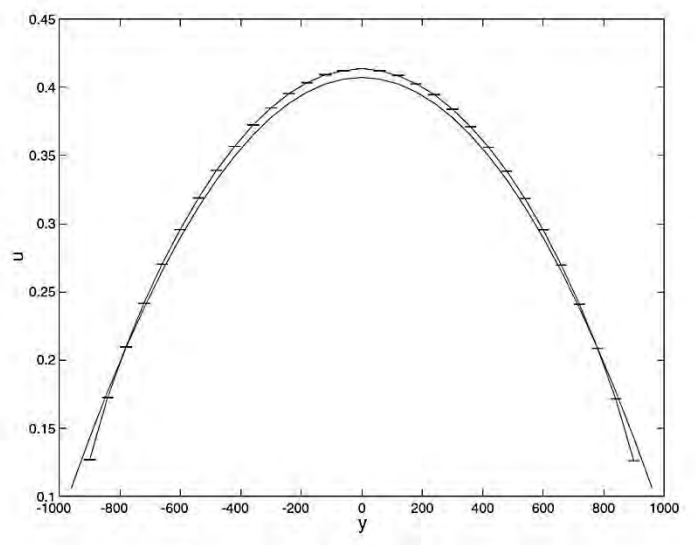
Hydrodynamic Fluctuations

DSMC is the dominant numerical method for molecular simulations of dilute gases

Plane Poiseuille Flow

Consider acceleration-driven Poiseuille flow in a narrow (10 mean free paths) channel with parallel walls.





Reynolds number is small (< 1) and Mach number is 0.1

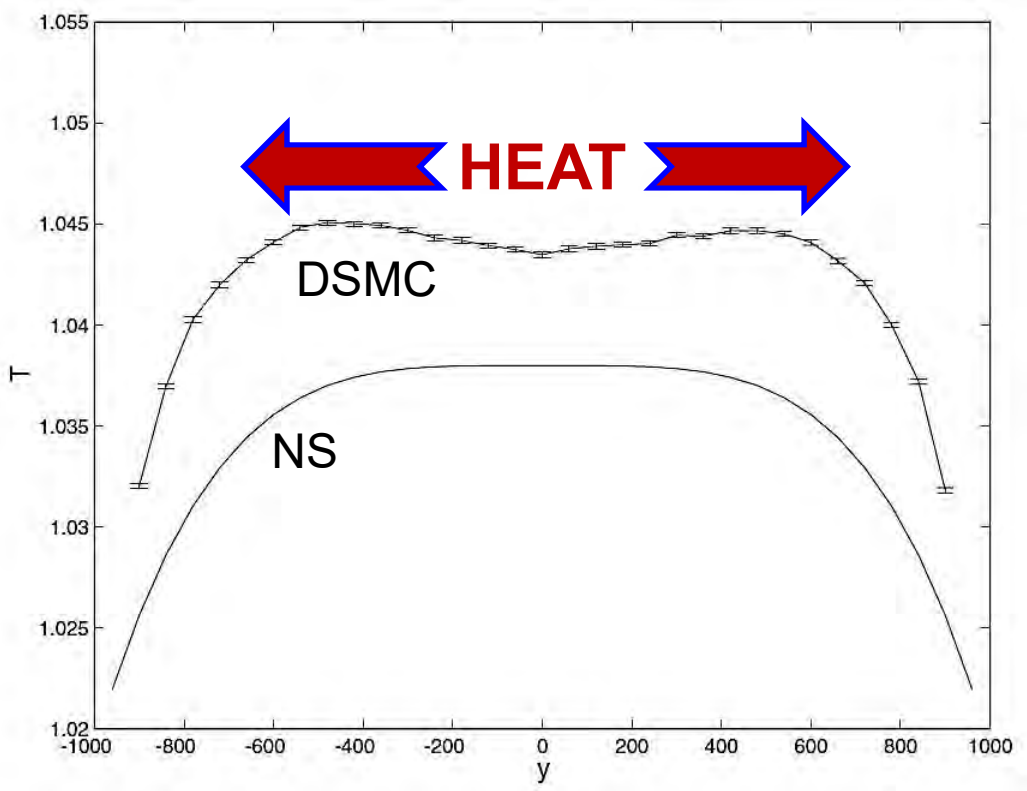
Velocity profile measured in DSMC is almost identical to that predicted by Navier Stokes equations

Heat Flux & Temperature Gradient

DSMC temperature profile has a *minimum* at the center of the channel.

Heat is generate inside the system by shearing and is removed at the walls so *heat is flowing from cold to hot*.

Results confirmed by super-Burnett eqns. and by molecular dynamics



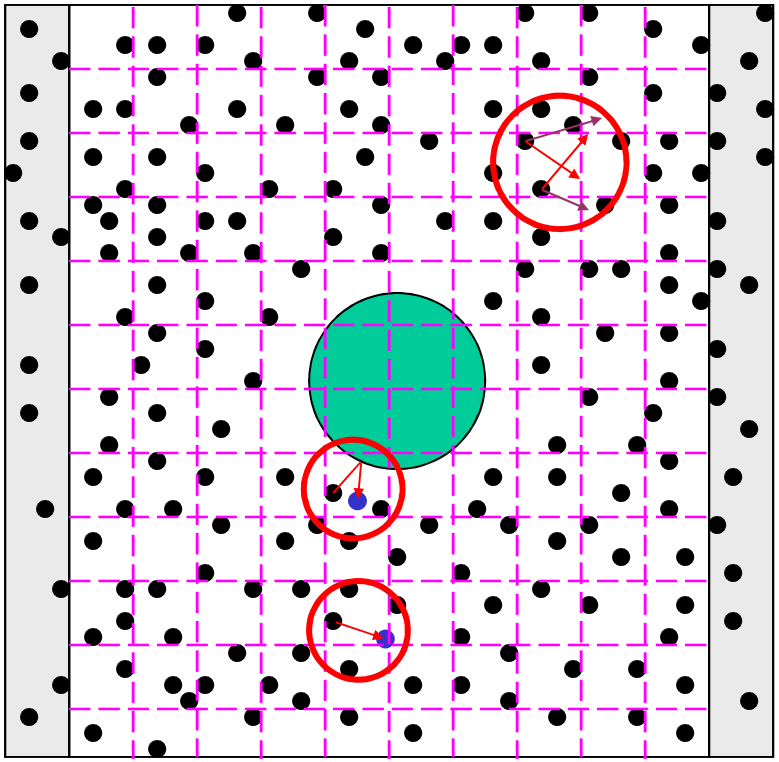
M. Malek Mansour, F. Baras and ALG, *Physica A* **240** 255 (1997)

DSMC Algorithm

DSMC resembles MD except in the evaluation of collisions

- Initialize system with particles
- Loop over time steps
  - Create particles at open boundaries
  - Move *all* the particles
  - Process any interactions of particle & boundaries
  - Sort particles into cells
  - Sample statistical values
  - Select and execute random collisions

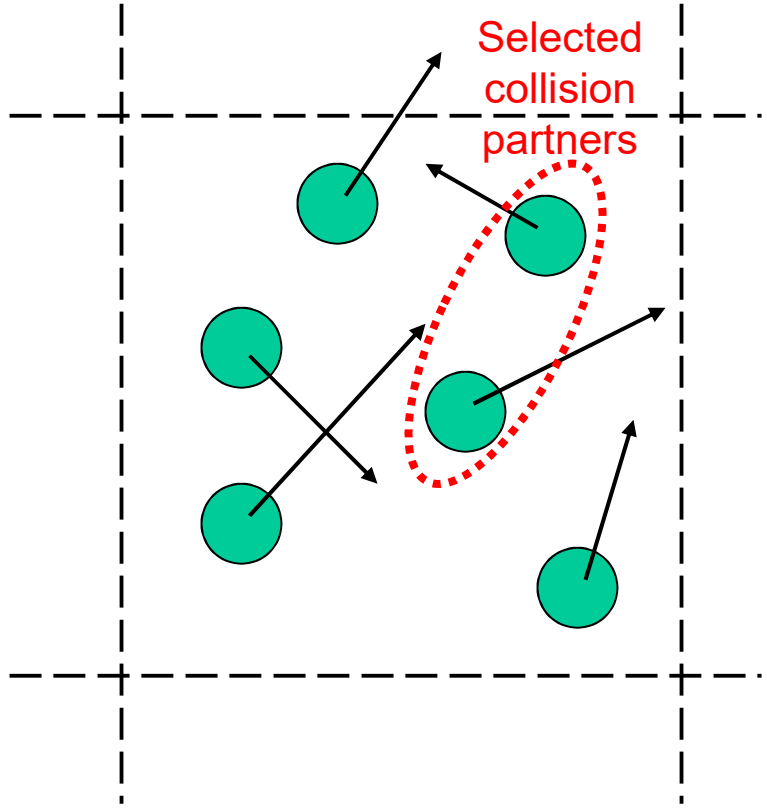
Example: Flow past a sphere



G.A. Bird, *The DSMC Method* (2013)
ALG, *Numerical Methods for Physics* (2015)

Traditional DSMC Collisions

- Sort particles into spatial collision cells
- Loop over collision cells
  - Compute collision frequency in a cell
  - Select random collision partners within cell
  - Process each collision



Collision pair with large relative velocity are more likely to collide but they do *not* have to be on a collision trajectory.

Collision Rate

Number of collisions that should occur during time step is

$$M_{coll} = \tau \, R\left(N, V, d, \langle v_r \rangle, \ldots\right)$$

where the functional form of the collision rate, R, depends on the intermolecular model.

For hard spheres of diameter d, the collision rate is,

$$R\left(N, V, d, \langle v_r \rangle\right) = \frac{N(N-1)\,\pi d^2 \langle v_r \rangle}{2V}$$

where is $\langle v_r \rangle$ the average relative velocity.

Early DSMC implementations used $N\,N_{\text{av}}$ instead of $N(N-1)$, where N_{av} is an estimated average value of N.

Collision Selections

To avoid having to compute the average relative velocity for all particle pairs in a cell, a larger number of attempted collisions are selected and some are rejected.

Number of collisions attempted during a time step is,

$$M_{cand} = \tau \, R\left(N, V, d, v_{\max}\right)$$

where $v_{\max} \geq \max\{v_r\}$ is estimated maximum relative velocity. An attempted collision is accepted with probability,

$$P(\mathbf{v}_i, \mathbf{v}_j) = \frac{R\left(N, V, d, v_r = \left|\mathbf{v}_i - \mathbf{v}_j\right|\right)}{R\left(N, V, d, v_{\max}\right)}$$

Early DSMC implementations used an exponentially distributed waiting time between collisions, similar to SSA (i.e., Gillespie's method for chemical reactions.

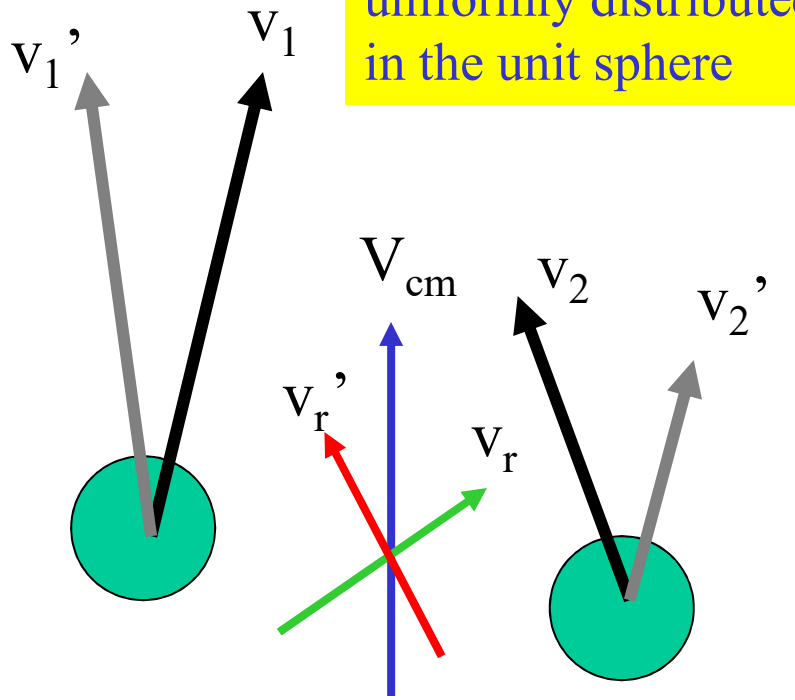
Post-Collision Velocities

Post-collision velocities

(6 variables) given by:

- Conservation of momentum (3 constraints)
- Conservation of energy (1 constraint)
- Random collision solid angle (2 choices)

For hard spheres the direction of v_r' is uniformly distributed in the unit sphere



Selection of the post-collision velocities must satisfy detailed balance.

Random Solid Angle

Post-collision relative velocity is,

$$\mathbf{v}_{\mathrm{r}}^{*} = v_{\mathrm{r}}[(\sin\theta\cos\phi)\hat{\mathbf{x}} + (\sin\theta\sin\phi)\hat{\mathbf{y}} + \cos\theta\hat{\mathbf{z}}]$$

The azimuthal angle is just,

$$\phi = 2\pi\Re_1$$

Polar angle distribution is,

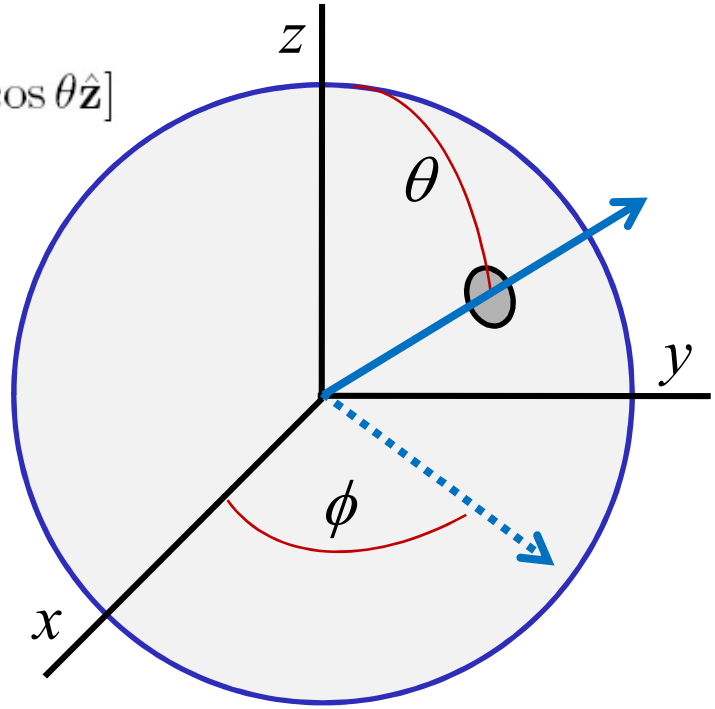
$$P_\theta(\theta)\,d\theta = \tfrac{1}{2}\sin\theta\,d\theta$$

But with change of variable,

$$P_q(q)\,dq = (\tfrac{1}{2})\,dq$$

So,

$$\begin{aligned} q &= 2\Re_2 - 1 \\ \cos\theta &= q \\ \sin\theta &= \sqrt{1-q^2} \end{aligned}$$



Generated by inversion method

Random Numbers

Need a high-quality random number generator for the uniform (0,1) distribution, such as Mersenne Twister.

Many distributions (e.g., Gaussian, exponential) may be generated by the inversion method:

Generate random value x with distribution $P(x)$ as $x = f(R)$ where R is uniformly distributed in (0,1).

Most other distributions are generated by the accept-reject method:

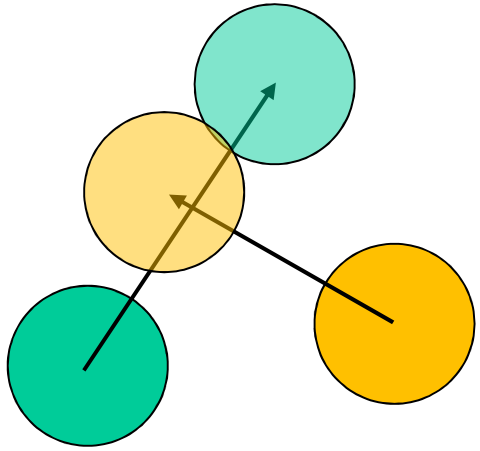
Draw x_{try} uniformly in the range of x;
Accept it if $P(x_{\text{try}}) > \max\{P(x)\}R$ else draw again.

Ballistic Motion

Particles motion is ballistic; during a time step, τ, particle positions are updated as,

$$\mathbf{r}(t + \Delta t) = \mathbf{r}(t) + \mathbf{v}(t)\,\tau$$

The particles move without interaction and can even overlap.

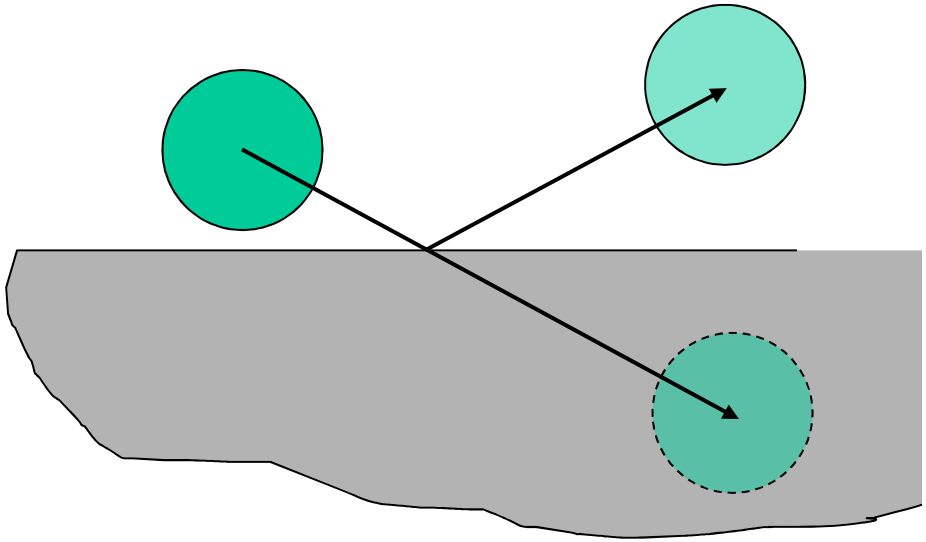


For transient flows, on the first time step use ½ τ (Strang splitting) to maintain accuracy. If measuring non-conserved variables (e.g., fluxes) then time-center the sampling (half move, sample, half move, collisions) for all steps.

Simple Boundaries

With periodic boundaries particles are re-introduced on the opposite side of the system.

Specular surfaces modeled by ballistic (mirror) reflection of point particles.



Be careful with corners.

Be careful with body forces.

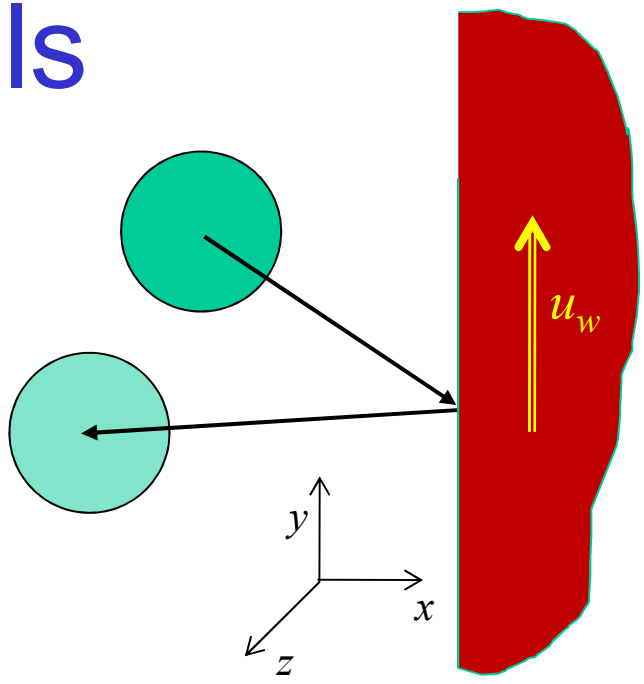
Thermal Walls

A more realistic treatment of a material surface is a thermal wall, which resets the velocity of a particle as a biased-Maxwellian distribution,

$$P_{v_x}(v_x) = \pm\frac{m}{kT_{\rm w}} v_x e^{-mv_x^2/2kT_{\rm w}}$$

$$P_{v_y}(v_y) = \sqrt{\frac{m}{2\pi kT_{\rm w}}} e^{-m(v_y-u_{\rm w})^2/2kT_{\rm w}}$$

$$P_{v_z}(v_z) = \sqrt{\frac{m}{2\pi kT_{\rm w}}} e^{-mv_z^2/2kT_{\rm w}}$$



These distributions (exponential and Gaussian) are simple to generate by inversion.

Walls can also be part-thermal, part-specular (accommodation).

Reservoir Boundary

Inflow/outflow boundary conditions commonly treated as a reservoir with given density, fluid velocity, temperature.

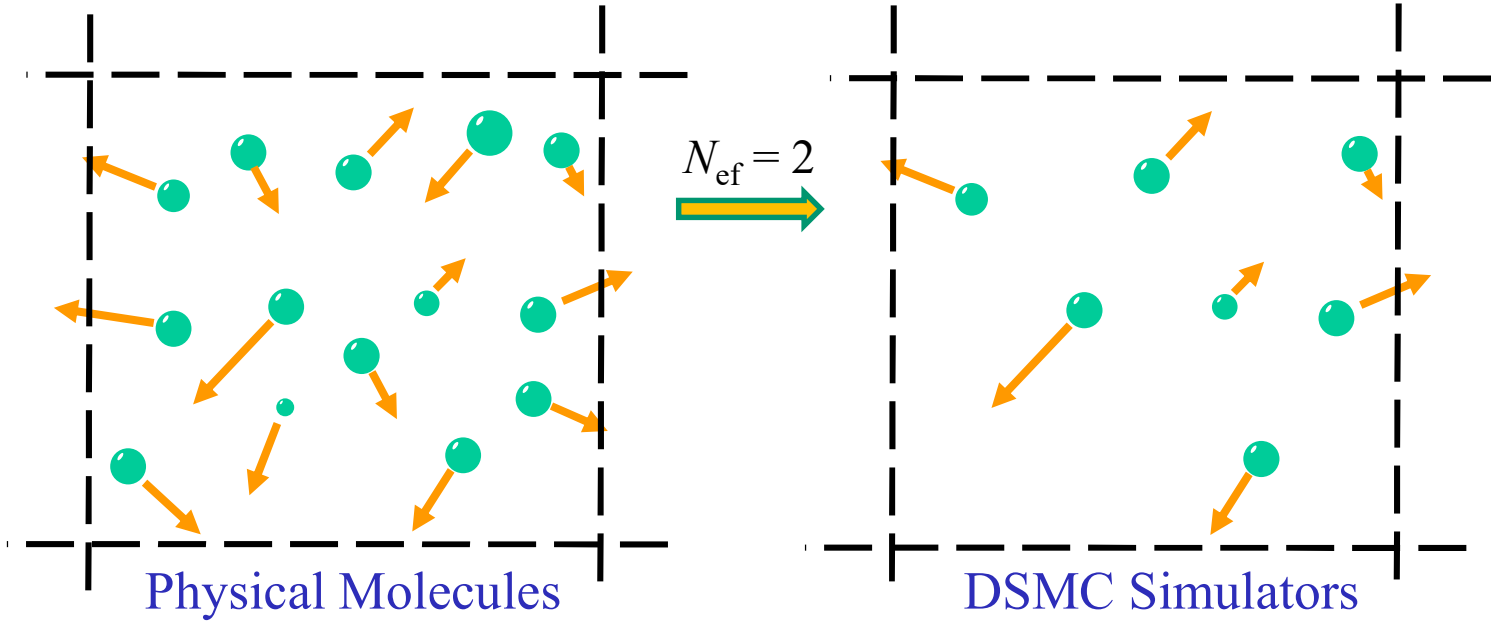
Particles in the main system are removed if they cross the boundary into the reservoir.

Particles injected from reservoir to main system by either:

- Surface generator: From number flux determines number to be injected; generate particle velocities from surface distribution (e.g., inflow Maxwellian).
- Volume generator: Initialize a "ghost cell" with particles before the ballistic move; discard any that do not cross the boundary into the main system during the move phase.

Molecules & “Simulators”

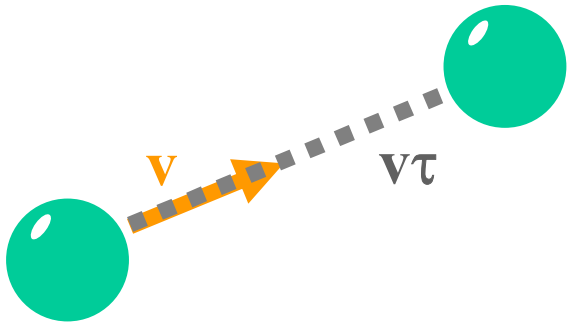
In DSMC the number of simulation particles (“simulators”) is typically a small fraction of the number physical molecules. Each simulator represents N_{ef} physical molecules.



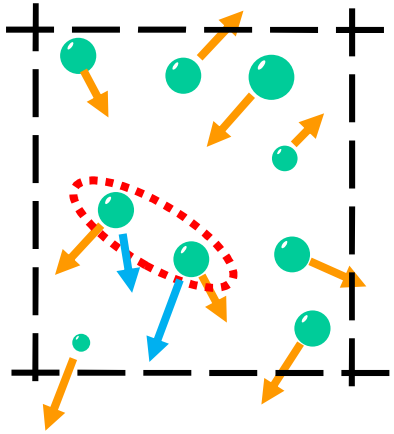
Accuracy of DSMC goes as $1/N$; for traditional DSMC about 10 particles per collision cell is the rule-of-thumb.

Ballistic & Collisional Transport

By their ballistic motion particles carry mass, momentum and energy. In a dilute gas, this is the *only* source of transport.

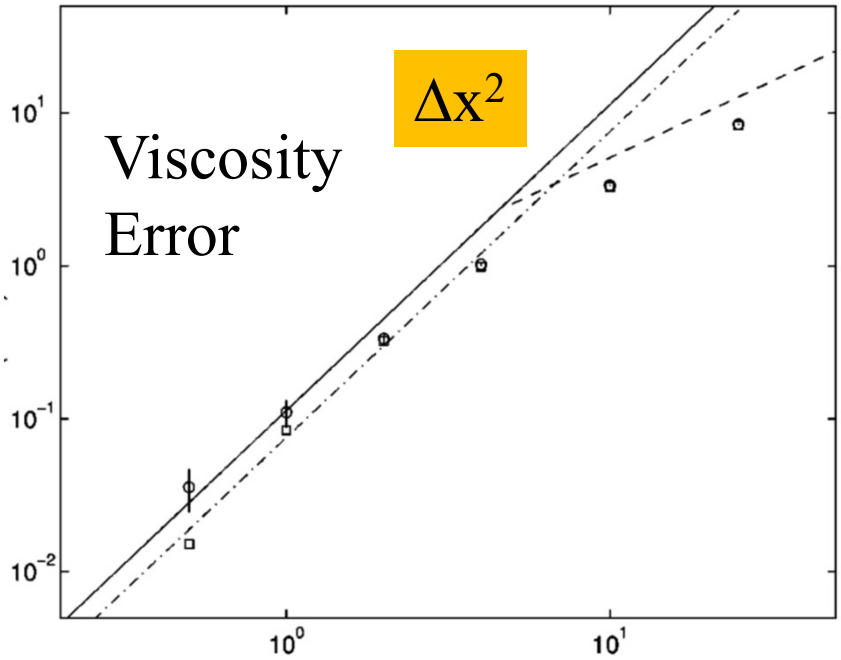


In DSMC, momentum and energy are also transported by the collisions. The larger the collision cell, the more collisional transport (greater average separation between particle pairs).

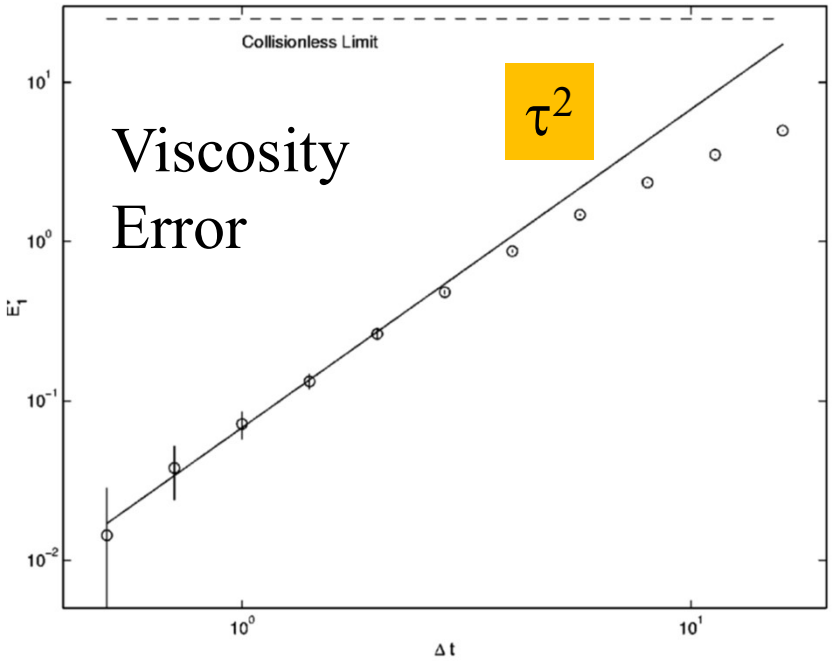


Cell Size and Time Step

Can calculate collisional transport by Green-Kubo theory.
The resulting error is quadratic in cell size and time step.



Cell size Δx



Time step τ

F. Alexander, ALG, and B. Alder, *Phys. Fluids*, **10** 1540 (1998)

ALG and W. Wagner, *Phys. Fluids* **12** 2621-33 (2000)

Cell Size and Time Step

Collisional transport is incorrect so to minimize it the cell size in DSMC is limited to a fraction of a mean free path.

For similar reasons, the time step is limited to a fraction of a mean collision time.

These two sources of error are the main limitations for large-scale DSMC simulations of macroscopic systems.

Due to symmetry, the collisional transport does not affect the pressure. However, if we restrict collisions to only particles moving towards each other then this symmetry is broken and DSMC has a non-ideal gas equation of state.

DSMC Variants for Dense Gases

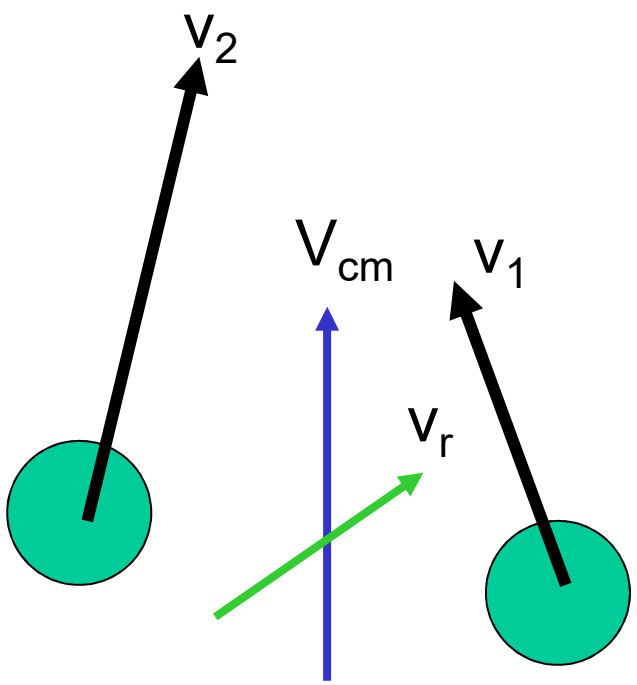
DSMC variants have been developed for dense gases:

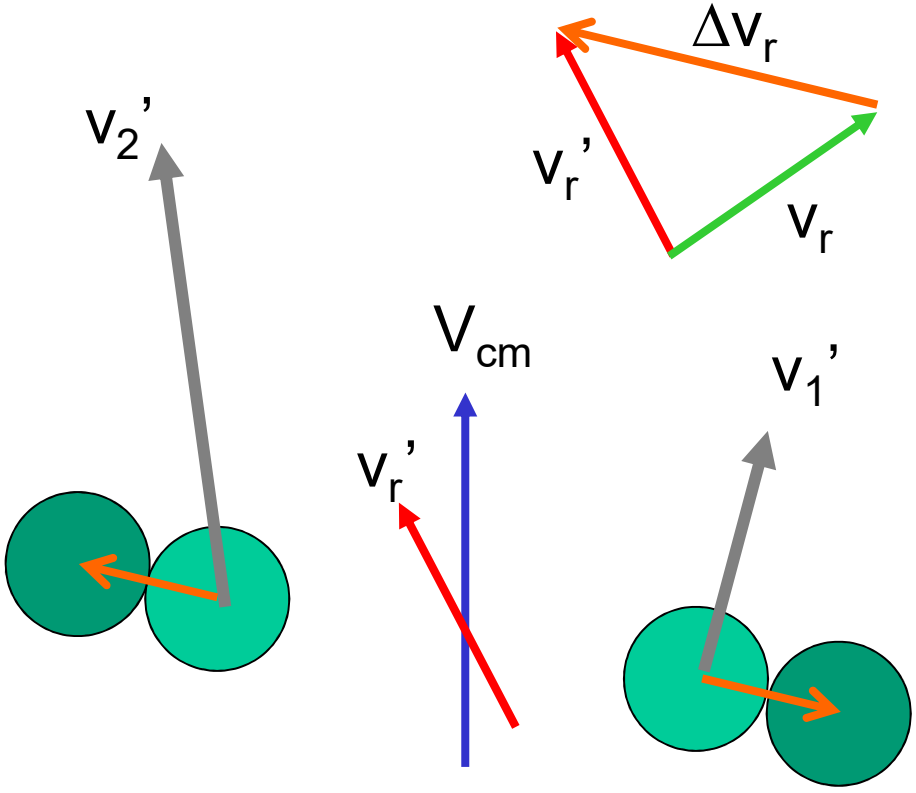
* Consistent Boltzmann Algorithm (CBA)
* Enskog-DSMC
* Consistent Universal Boltzmann Algorithm (CUBA)
* Stochastic Hard Sphere Dynamics (SHSD)

Basic idea is to modify the collision process so that the collisional transport produces the desired non-ideal equation of state.

Consistent Boltzmann Algorithm (CBA)

Pre-collision velocities





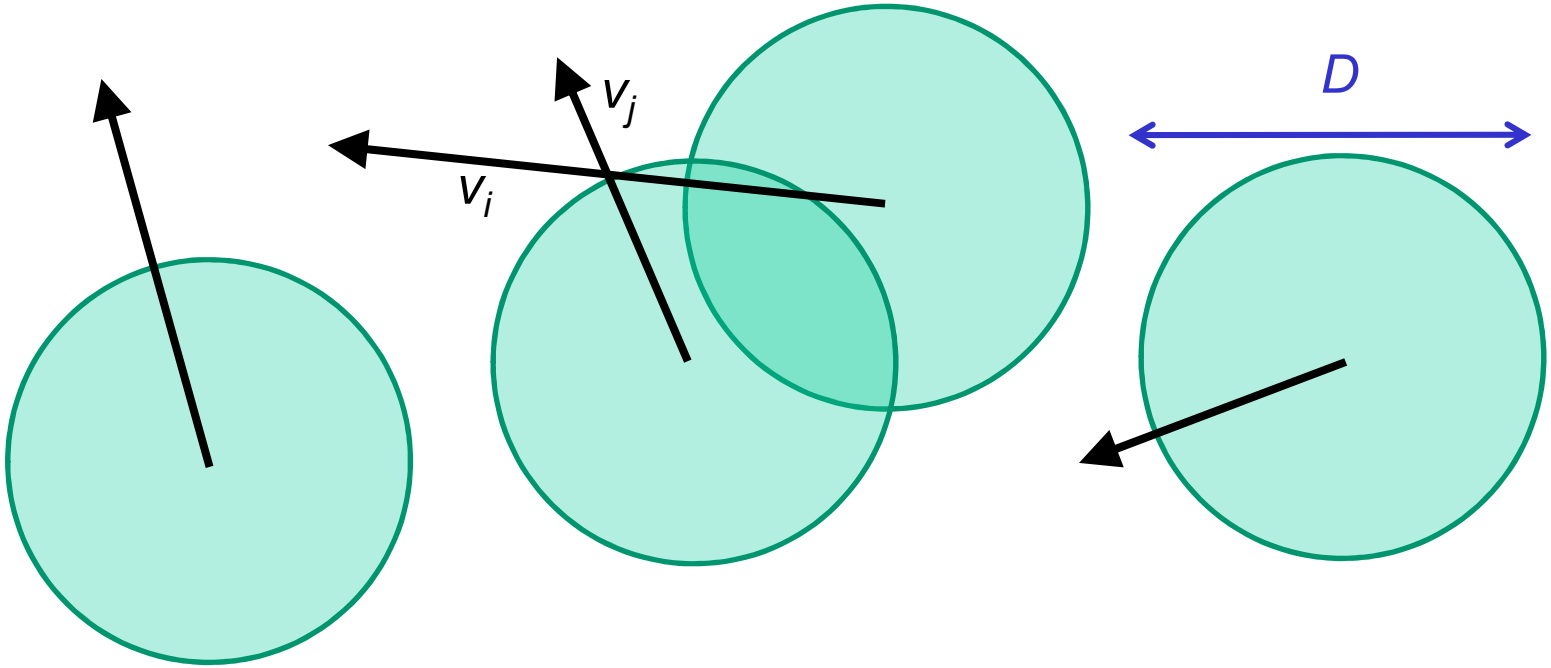
Post-collision velocities

Post-collision positions

F. Alexander, ALG, and B. Alder,
Phys. Rev. Lett. **74** 5212 (1995)

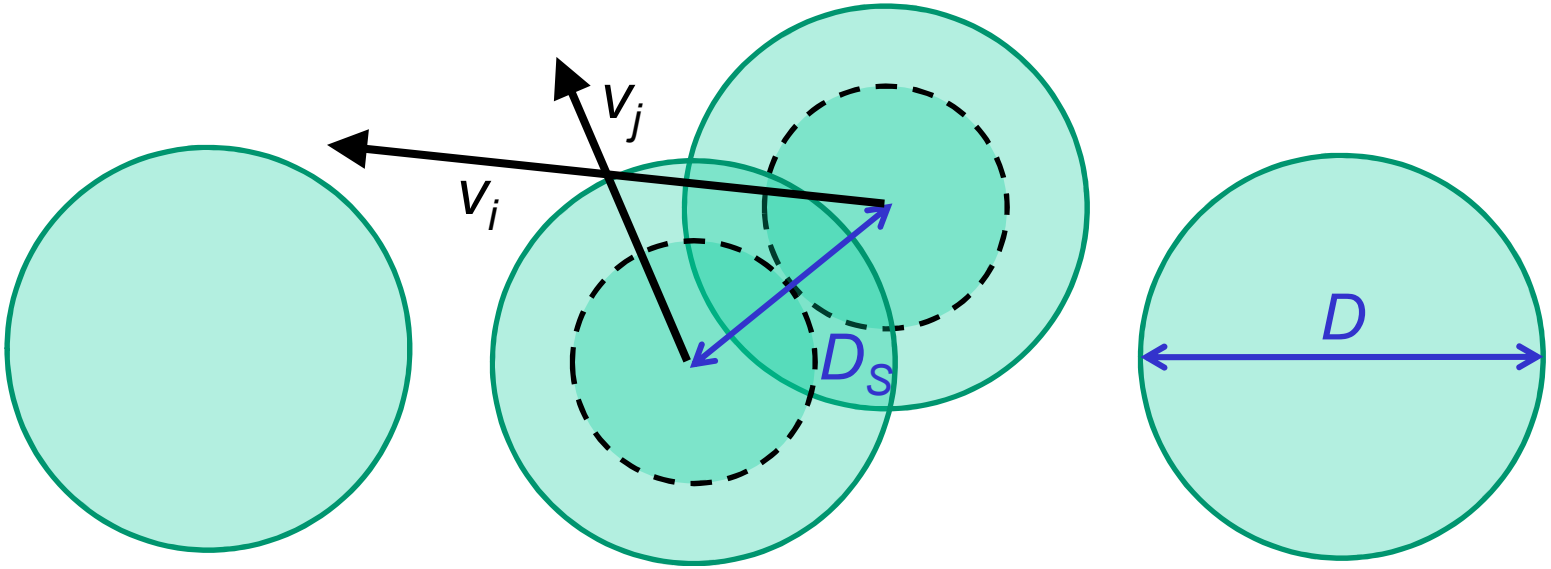
Isotropic (Cell-less) DSMC

Particles move ballistically during a time step and undergo random collisions if they are with an interaction range, D. Collision probability is a function of the relative velocity, v_{ij}.



A. Donev, B.J. Alder, and ALG, J. Statistical Mechanics P11008 (2009).

Stochastic Hard-Sphere Dynamics



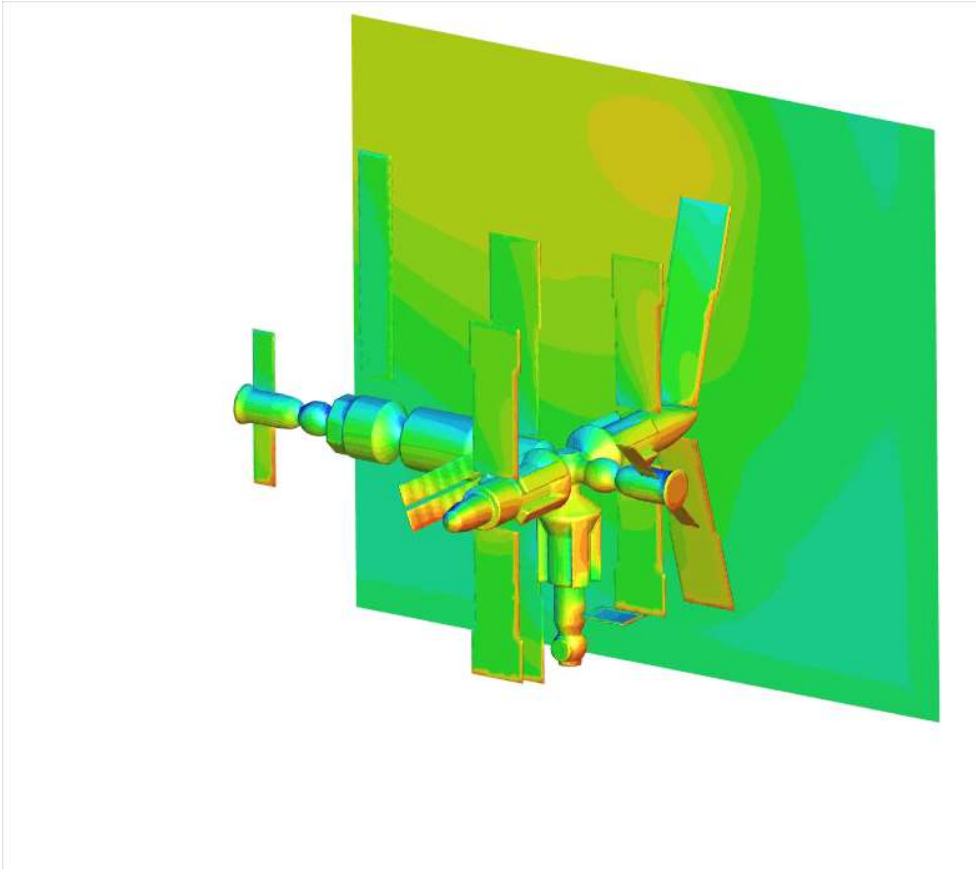
Stochastic Hard-Sphere Dynamics: Particles move ballistically in-between collisions. When two particles i and j are less than a diameter apart, $r_{ij} \leq D$, there is a probability rate $(3\chi/D)v_n\Theta(v_n)$ for them to collide as if they were elastic hard spheres with a variable diameter $D_S = r_{ij}$, where $v_n = -\mathbf{v}_{ij} \cdot \hat{\mathbf{r}}_{ij} > 0$.

A. Donev, B.J. Alder, and ALG, Phys. Rev. Lett. **101**, 075902 (2008).

SPARTA – Sandia DSMC Code

SPARTA is the open-source DSMC code developed at Sandia by Steve Plimpton and Michael Gallis.

SPARTA is similar to (and by the same authors) as the molecular dynamics code LAMMPS



Mir space station re-entry (temperature)
Courtesy of M. Gallis (Sandia)

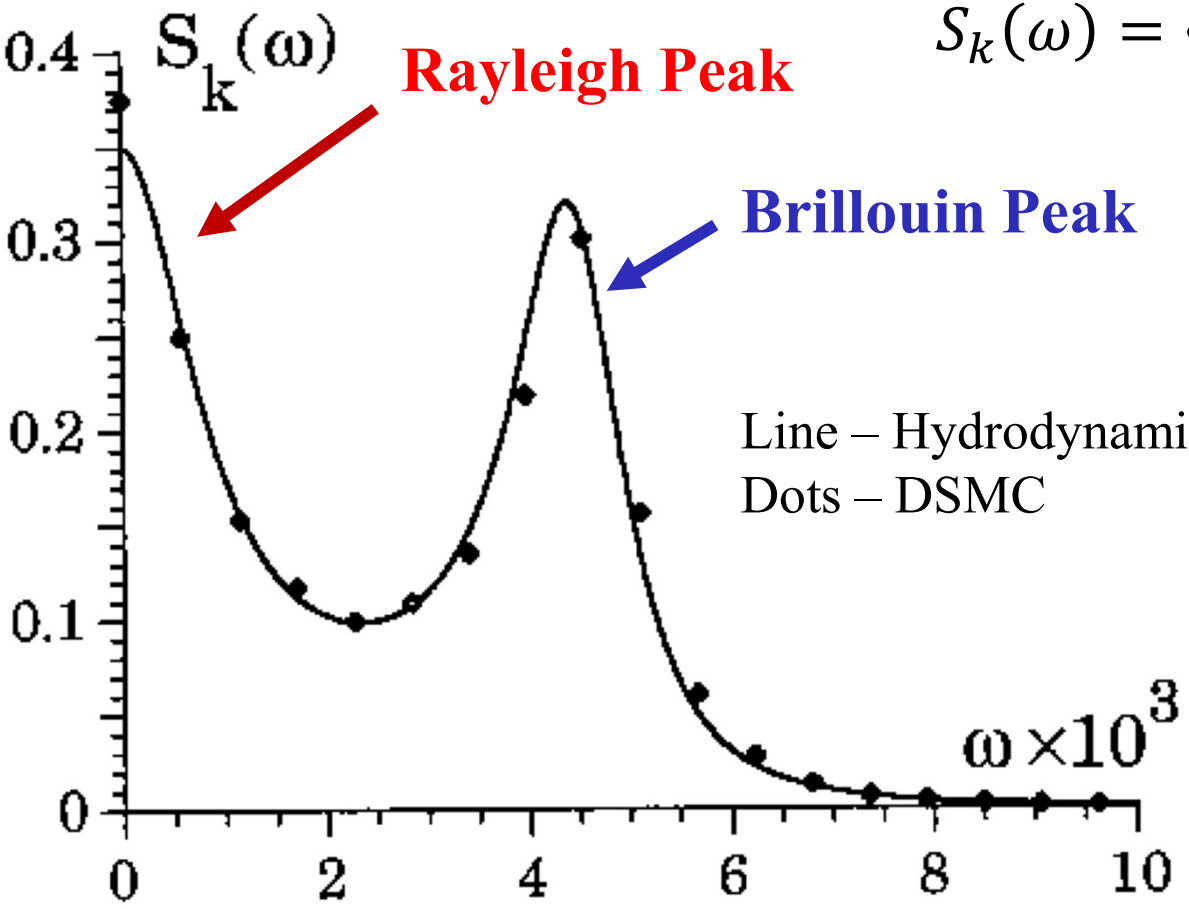
Part I
Direct Simulation
Monte Carlo

Part II
Hydrodynamic
Fluctuations

Fluctuations in DSMC

- Hydrodynamic fluctuations (density, temperature, etc.) have *nothing* to do with Monte Carlo aspect of DSMC.
- Variance of fluctuations in DSMC is exact at equilibrium (due to uniform distribution for position and Maxwell-Boltzmann for velocity).
- Time-correlations are correct, at least at hydrodynamic scales.
- Non-equilibrium fluctuations correct, at least at hydrodynamic scales.

Dynamic Structure Factor



$$S_k(\omega) = \langle \delta\rho(k,\omega)\delta\rho^*(k,\omega) \rangle$$

Line – Hydrodynamics
Dots – DSMC

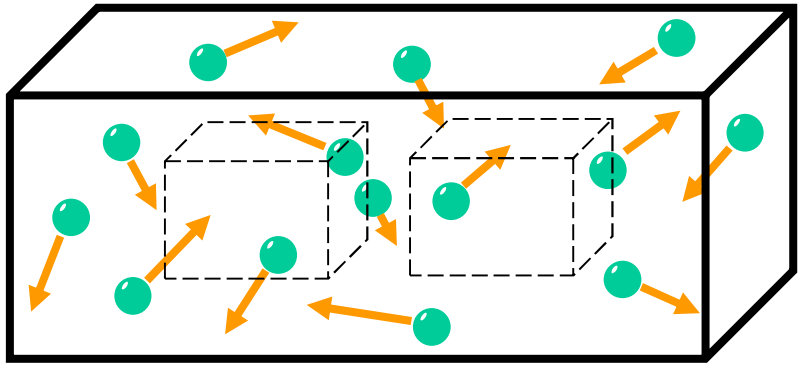
Experimentally measureable by light scattering

F. Baras, M. Malek Mansour, ALG and M. Mareschal, *J. Comp. Phys.* **119** 94 (1995).

Spatial Correlations

At equilibrium, fluctuations of conjugate hydrodynamic quantities are spatially uncorrelated. For example, density is spatially uncorrelated with fluid velocity,

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



Out of equilibrium, (e.g., gradient of temperature) long-ranged spatial correlations appear in a fluid.

Brillouin Peak Asymmetry

In the 1980's fluctuating hydrodynamics predicted and light scattering experiment confirmed that non-equilibrium fluctuations were correlated.

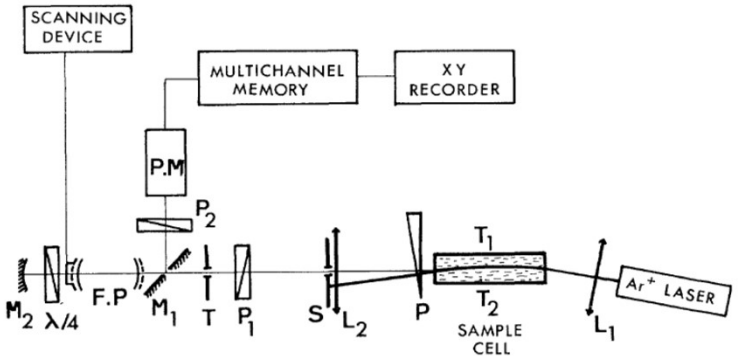
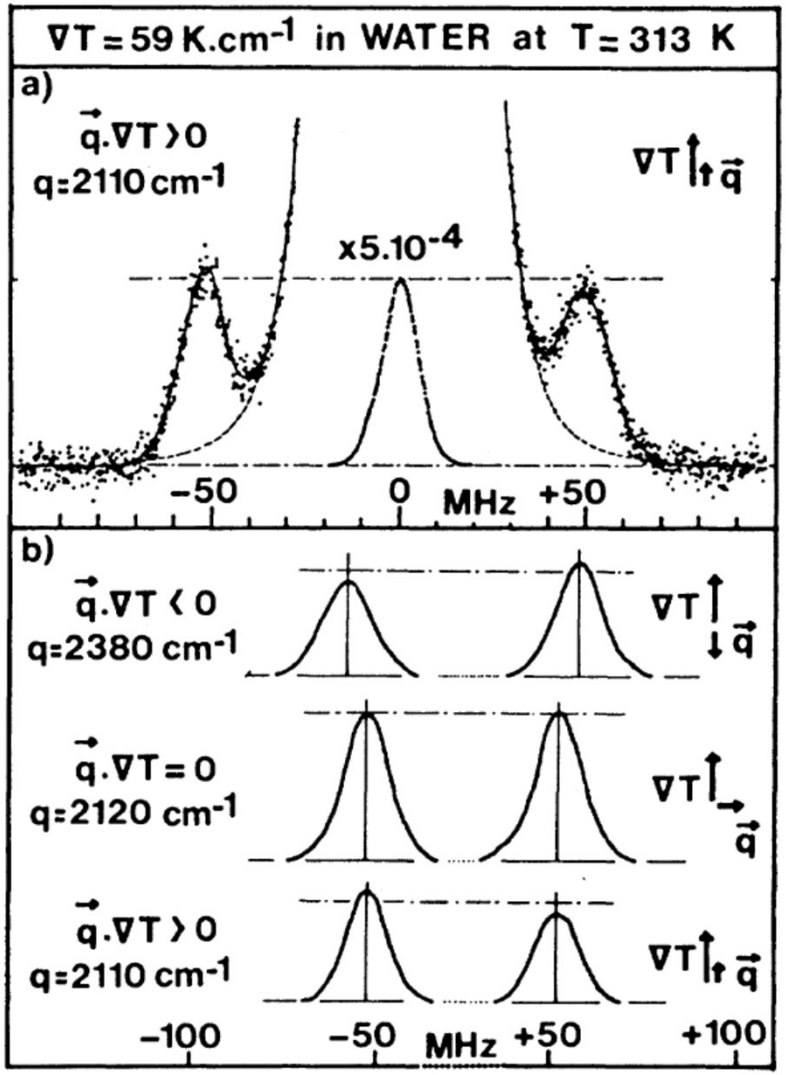


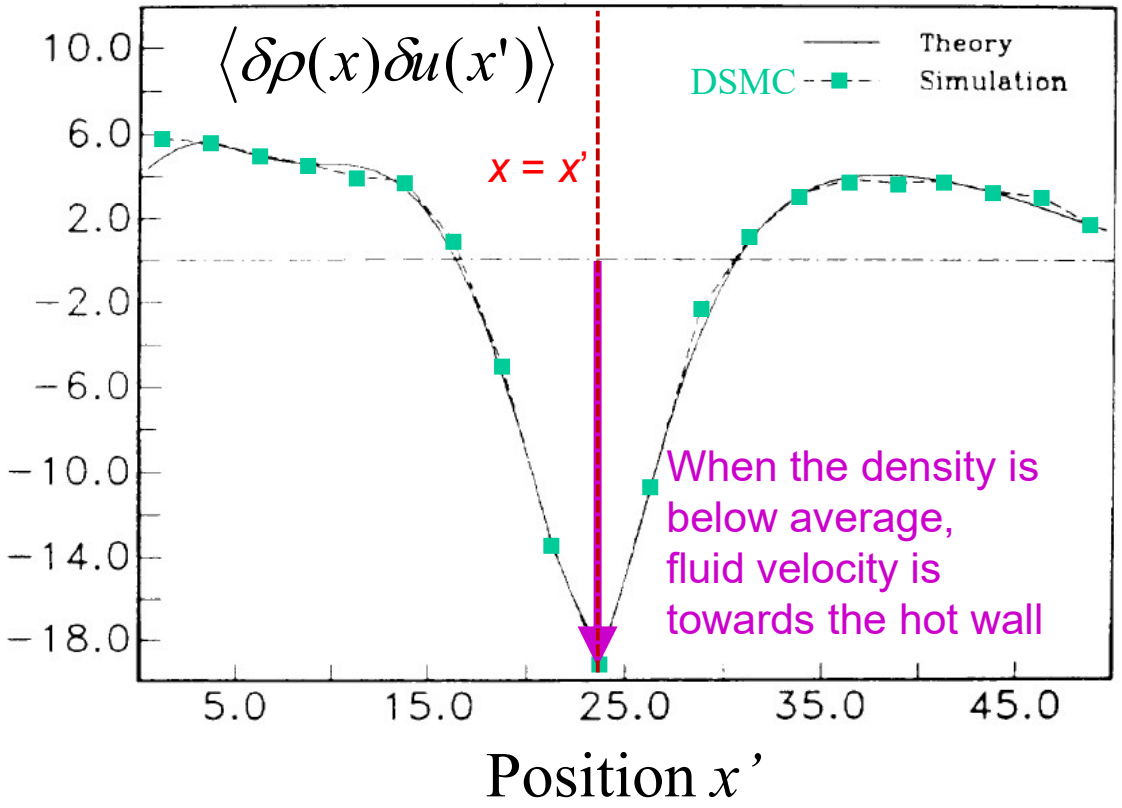
FIG. 1. Experimental setup: L_1 and L_2, lenses; P, prism; P_1 and P_2, polarizers; M_1 and M_2, mirrors; S, slit; T, pinhole.

D. Beysens, Y. Garrabos, and G. Zalczer, *Phys. Rev. Lett.* **45**, 403 (1980)

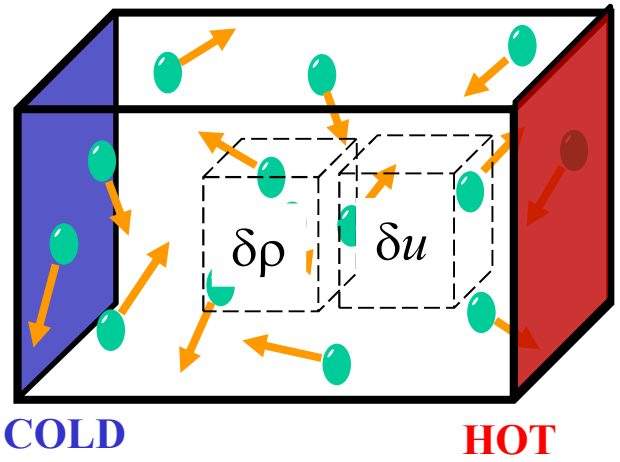


Density-Velocity Correlation

Correlation of density-velocity fluctuations under ∇T



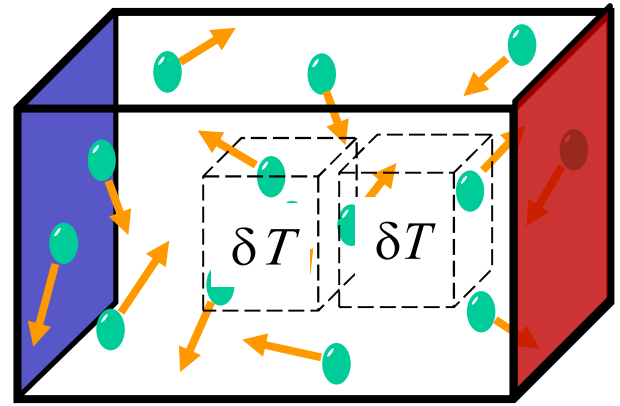
Theory is Landau-Lifshitz fluctuating hydrodynamics

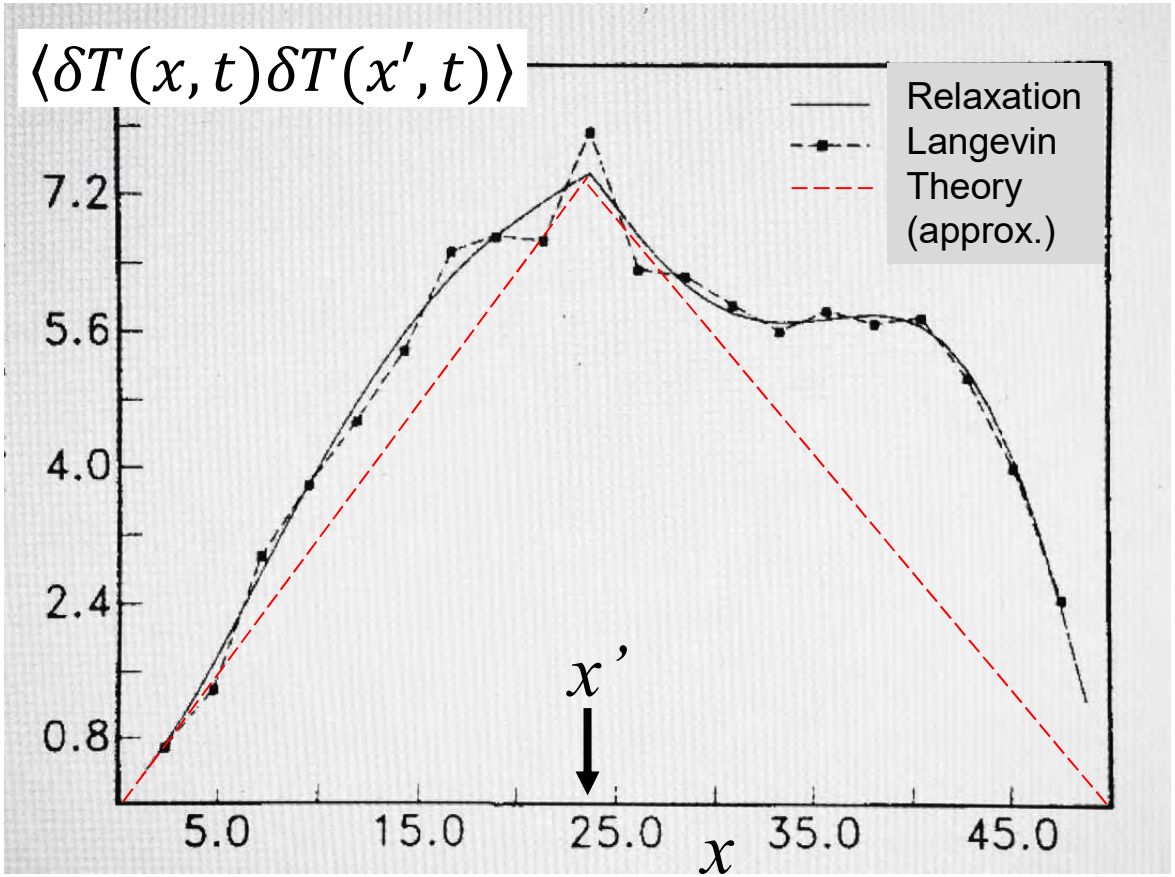


"Nonequilibrium Fluctuations studied by a Rarefied Gas Simulation", ALG, *Phys. Rev. A* **34** 1454 (1986)

Langevin Schemes for Fluctuating Hydrodynamics

Developed numerical Langevin schemes for the full fluctuating hydrodynamics eqns. in order to check our DSMC results.

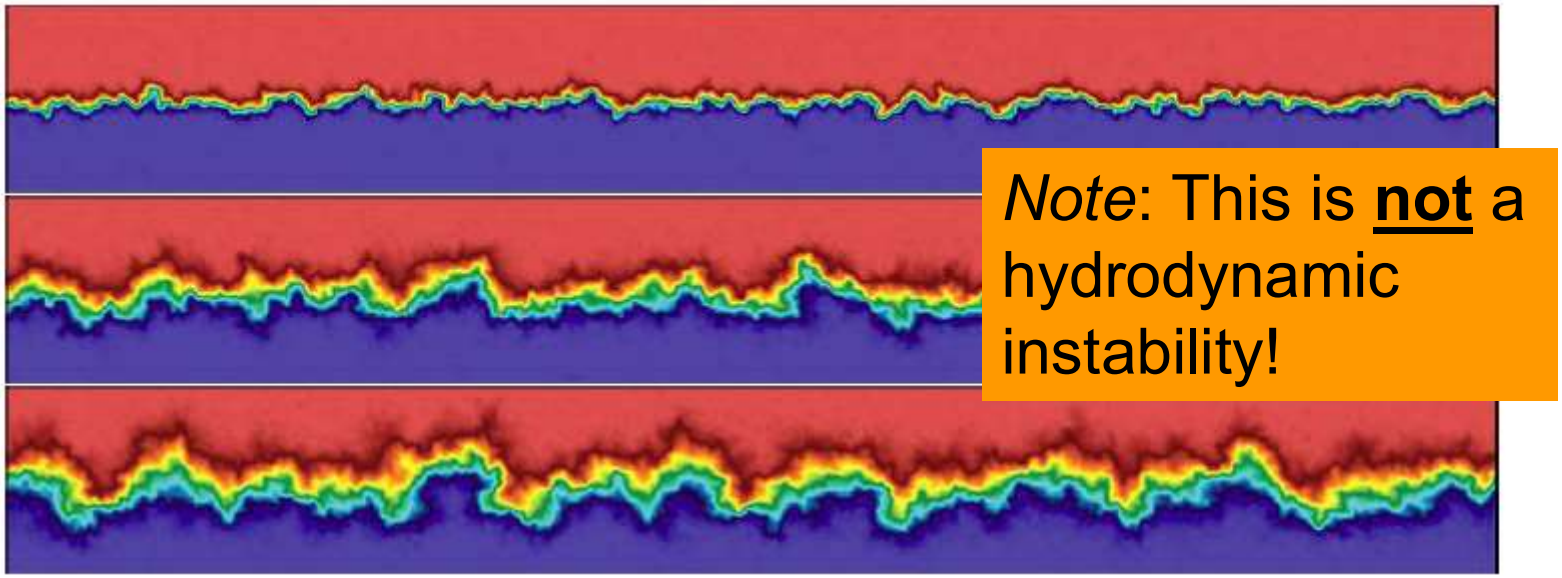




ALG, M. Malek Mansour, G. Lie and E. Clementi, *J. Stat. Phys.* **47** 209 (1987)

Giant Fluctuations in Mixing

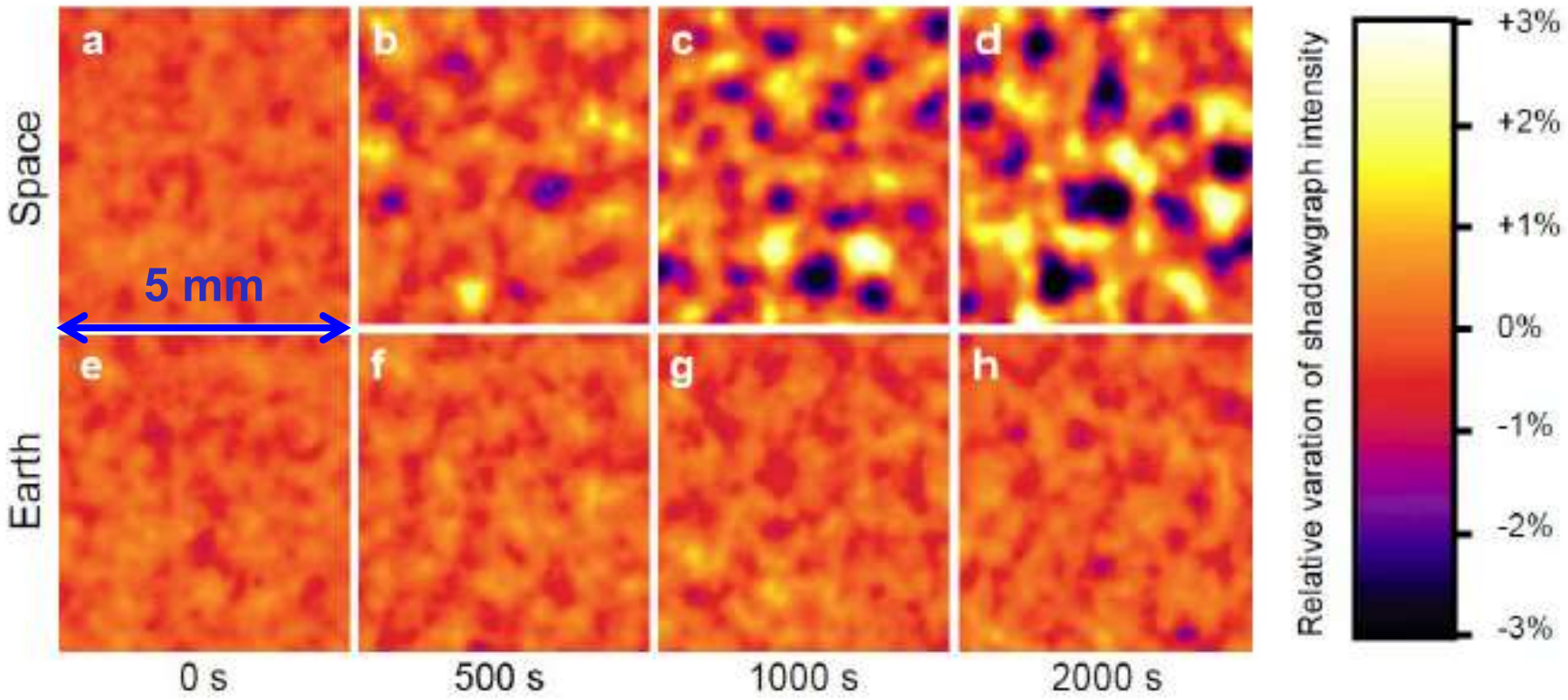
Fluctuations grow large during mixing even when the two species are identical (red & blue).



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 at $t = 0$.

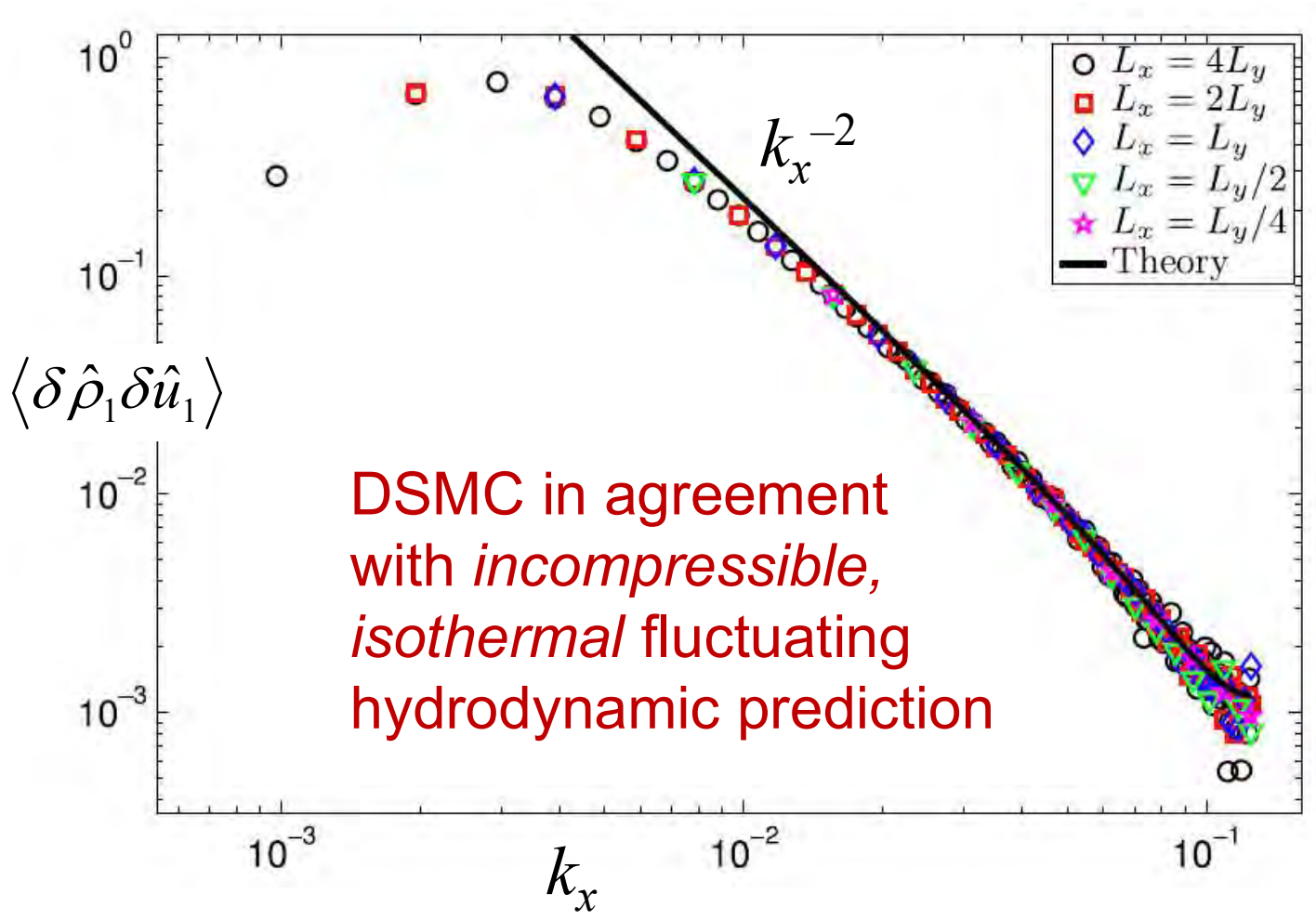
Experimental Observations (cont.)



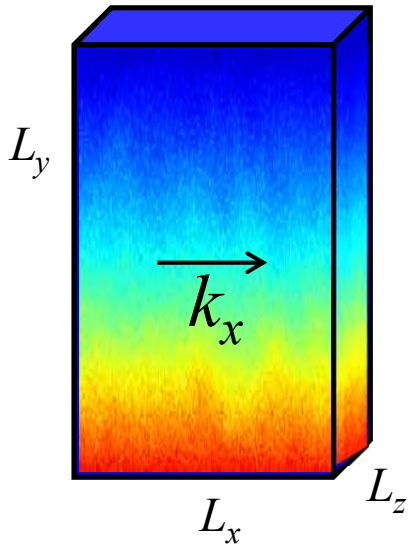
Experiments confirm that concentration fluctuations are reduced by gravity with a cut-off wavelength that goes as $1/g^{1/4}$.

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

Concentration-Velocity Correlation



Symbols are DSMC; $k_y = 0$

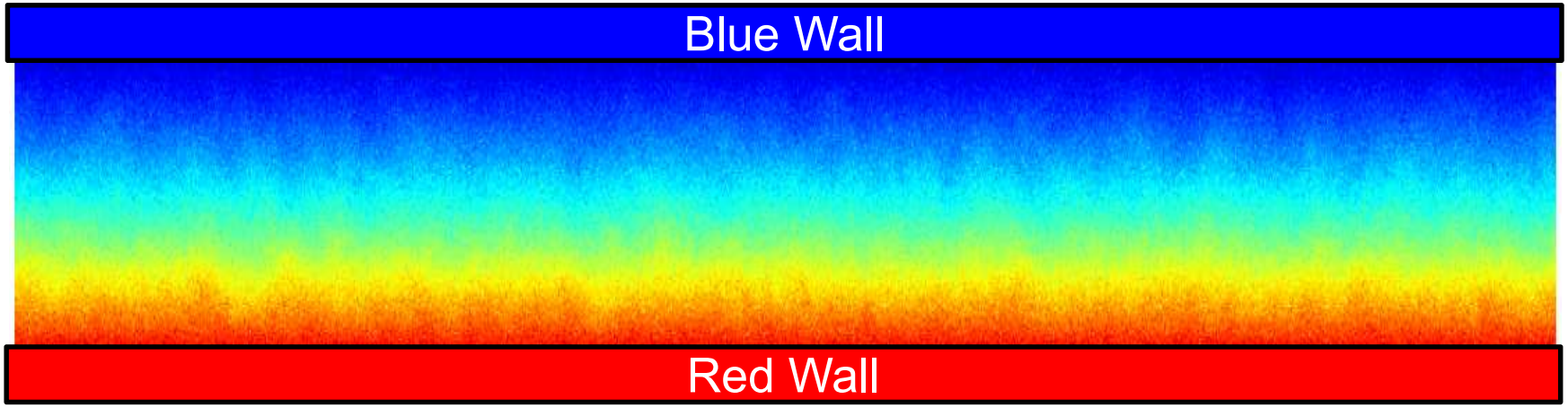


$L_y = 512\ \lambda; L_z = 2\ \lambda$

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

Diffusion Flux & Fluctuations

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



The non-equilibrium correlation $\left\langle \delta\hat{c}\,\delta\hat{u}_{\parallel} \right\rangle$ enhances the effective flux of concentration even at this steady state.

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

Fluctuation Enhanced Diffusion

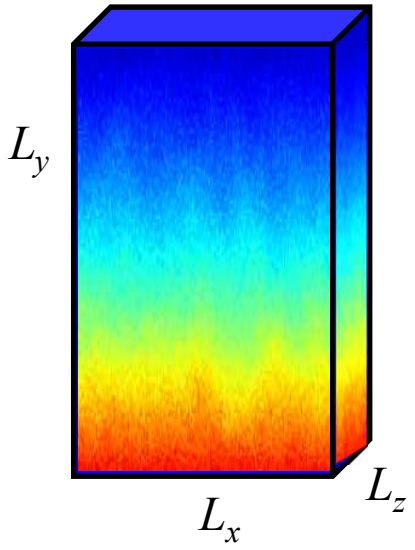
Can separate the contributions to the concentration flux as,

$$\langle \boldsymbol{F} \rangle = \underbrace{\langle \rho_1 \mathbf{u}_1 \rangle}_{D_{\text{eff}} \nabla c} = \underbrace{\langle \rho_1 \rangle \langle \mathbf{u}_1 \rangle}_{D_0 \nabla c} + \underbrace{\langle \delta\rho_1 \delta\mathbf{u}_1 \rangle}_{\Delta D \nabla c}$$

In DSMC we can easily measure

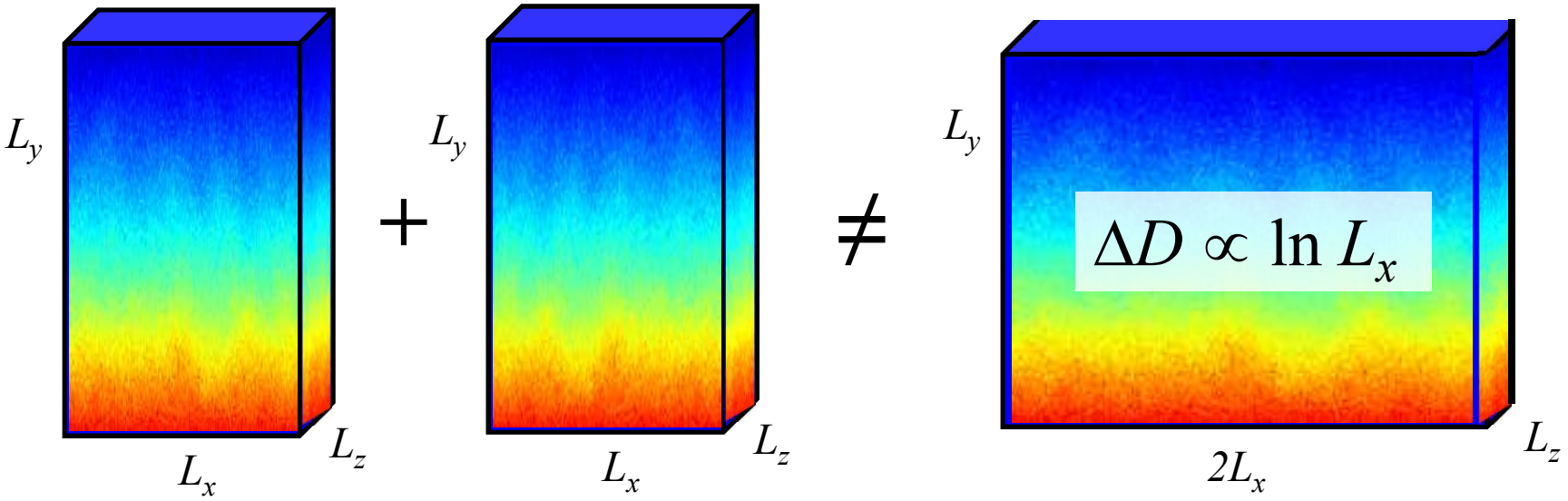
$$\langle \rho_1 \rangle, \langle \mathbf{u}_1 \rangle, \langle \rho_1 \mathbf{u}_1 \rangle \text{ and } \nabla c$$

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



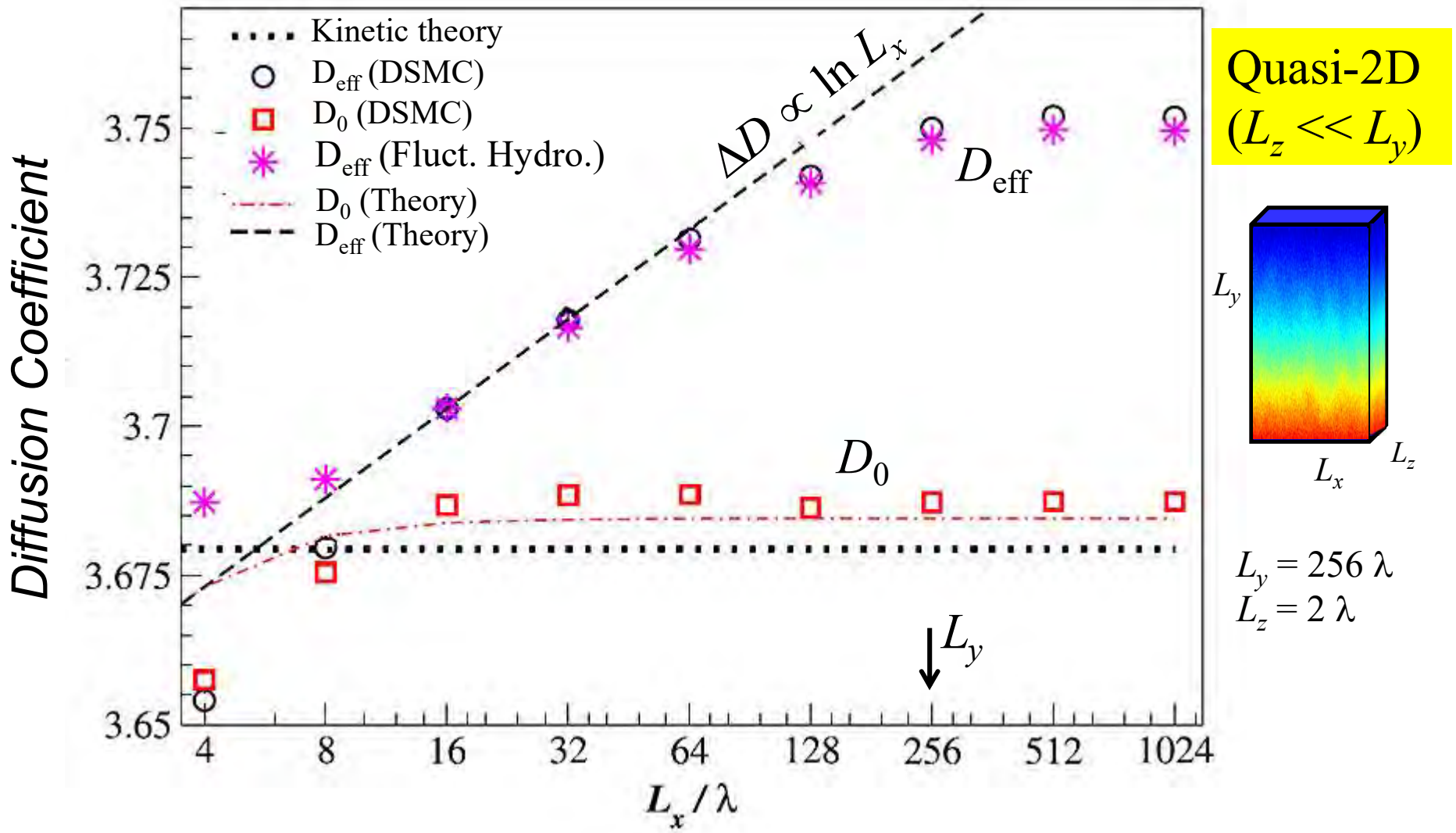
Diffusion and System Geometry

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 an enhanced diffusion rate.

DSMC and FNS Results (Quasi-2D)



Sampling and Fluctuations

The intuitive way to measure time-averaged fluid velocity from samples of particle velocities in a cell is,

Instantaneous Average

$$\langle u \rangle = \frac{1}{S} \sum_{j=1}^{S} \left(\frac{1}{N(t_j)} \sum_{i \in C}^{N(t_j)} v_i(t_j) \right) = \left\langle \frac{Momentum}{Mass} \right\rangle$$

An alternative measure is,

Cummulative Average

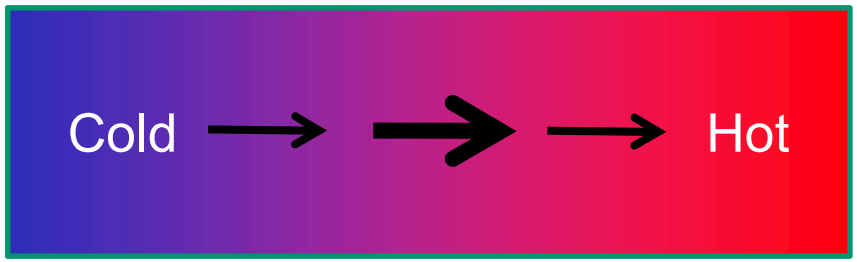
$$\langle u \rangle_* = \frac{\sum_j^S \sum_{i \in C}^{N(t_j)} v_i(t_j)}{\sum_j^S N(t_j)} = \frac{\langle Momentum \rangle}{\langle Mass \rangle}$$

These are not equivalent!

Anomalous Fluid Velocity

Mean instantaneous fluid velocity measurement gives an anomalous flow in a closed system at steady state with ∇T.

$$\langle u \rangle = \left\langle \frac{J}{M} \right\rangle \propto x(L-x)\nabla T$$



Using the cumulative mean, $\langle u \rangle_*$, gives the expected result of zero fluid velocity.

$$\langle u \rangle_* = \frac{\langle J \rangle}{\langle M \rangle} = 0$$

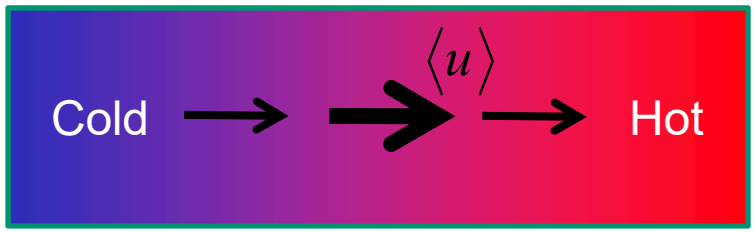
Fluid Velocity & Fluctuations

From the definitions,

$$\langle u \rangle \approx \langle u \rangle_* - \frac{\langle \delta\rho \delta u \rangle}{\langle \rho \rangle}$$

From correlation of non-equilibrium fluctuations,

$$\langle \delta\rho(x)\delta u(x) \rangle \propto -x(L-x)\nabla T$$



This effect is the origin of the anomalous fluid velocity.

M. Tysanner and ALG, *J. Comp. Phys.* **196** 173-83 (2004).

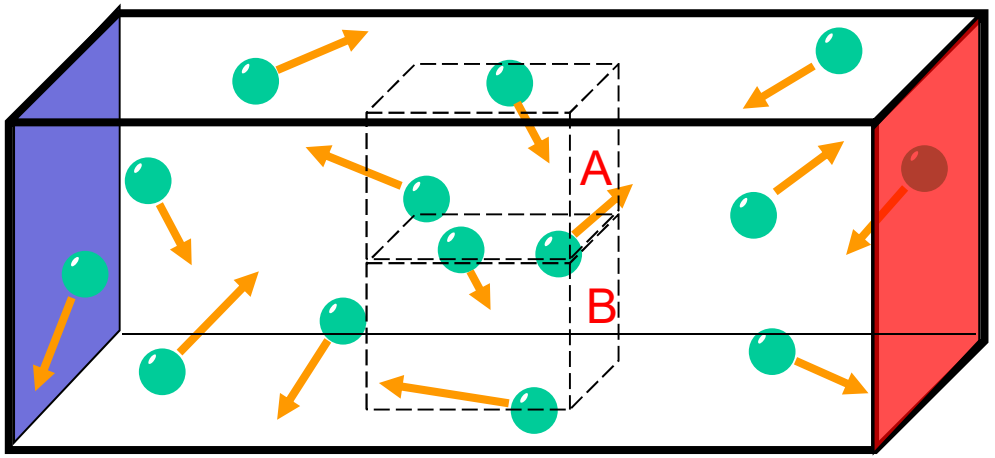
Instantaneous Temperature

Instantaneous temperature has a similar error due to density-temperature correlation of fluctuations.

Error goes as 1/*N*, so instantaneous temperature is *not* an intensive quantity.

Temperature of cell A = temperature of cell B but *not* equal to temperature of super-cell (A U B)

$$\langle T \rangle \approx \langle T \rangle_* - \frac{\langle \delta\rho\delta T \rangle}{\langle \rho \rangle}$$

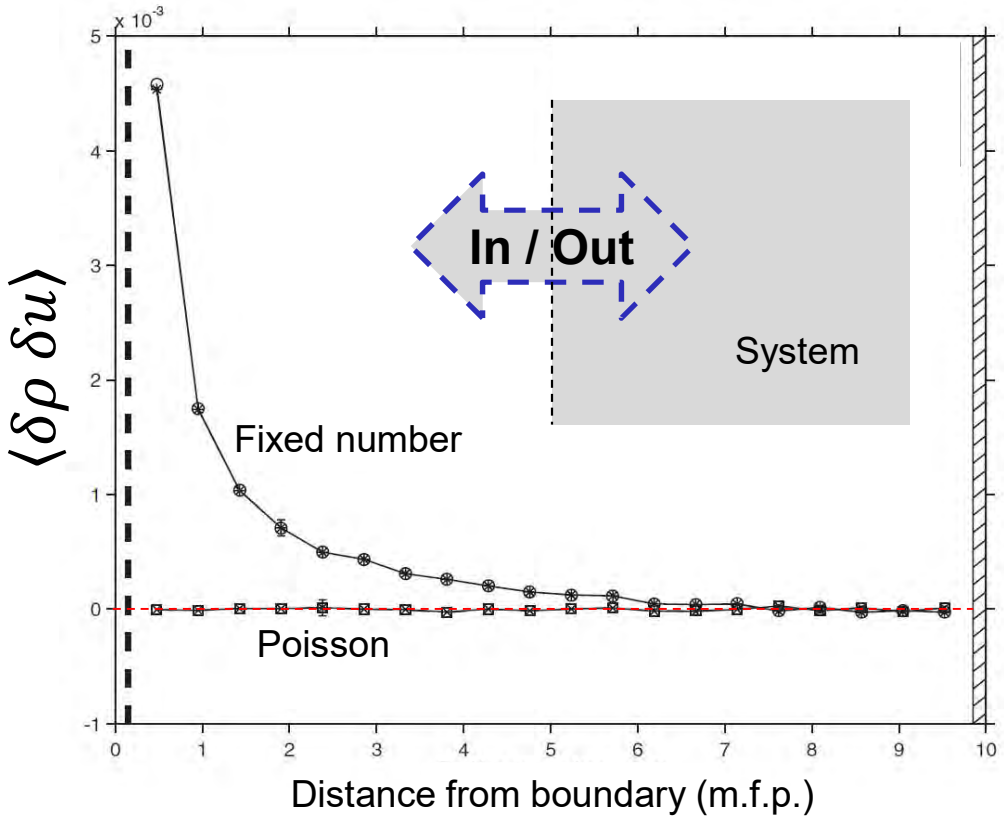


ALG, *Comm. App. Math. Comp. Sci.* **1** 53-78 (2006)

Inflow / Outflow Boundary

An inflow / outflow boundary models an infinite reservoir of particles with an equilibrium density and temperature.

Number of particles generated at the boundary should be Poisson distributed to match equilibrium.



M. Tysanner and ALG, *Int. J. Num. Meth. Fluids* **48** 1337-1349 (2005).

Brownian Motors

Heat engine is driven by thermal fluctuations.

Introduced by Smoluchowski and later popularized by Feynman.

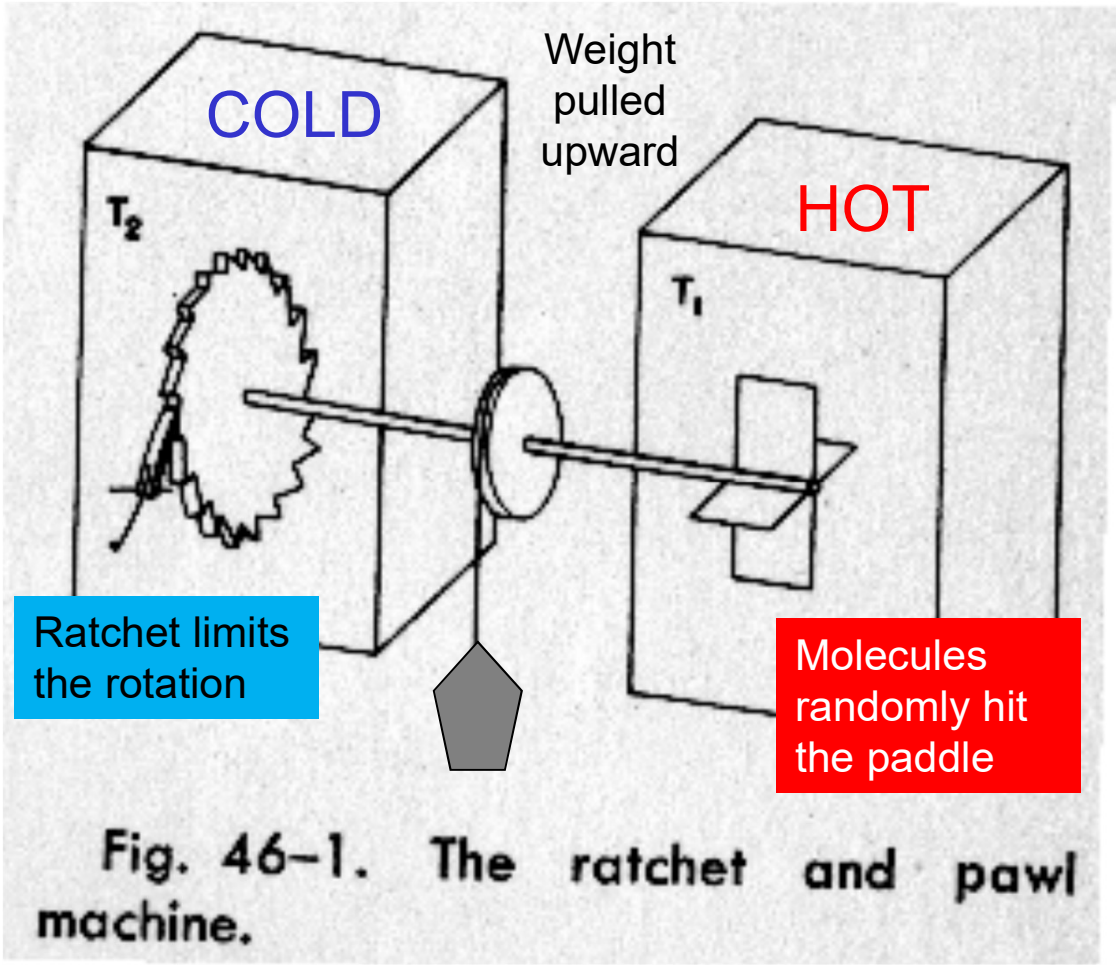
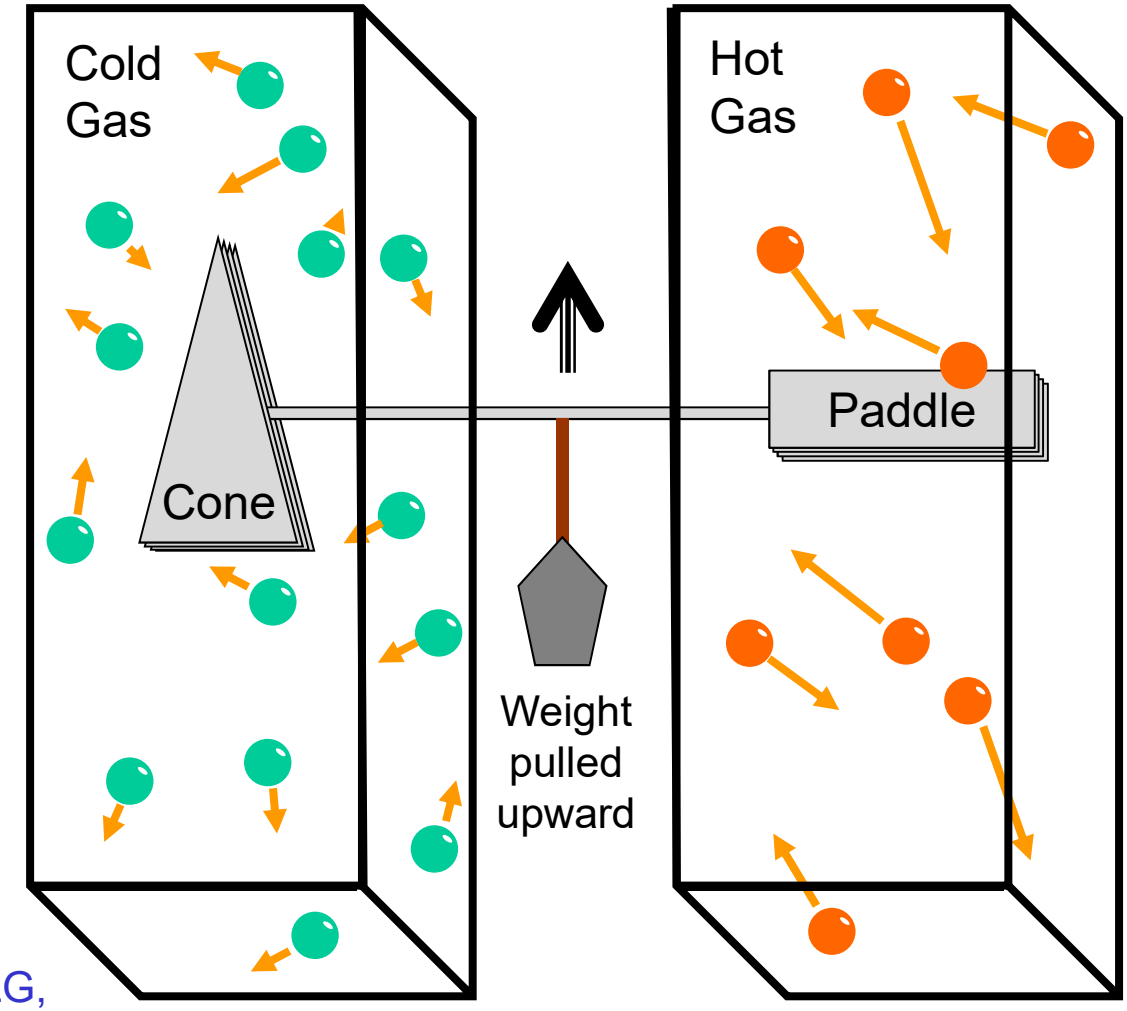


Fig. 46–1. The ratchet and pawl machine.

Triangula Brownian Motor

Feynman's complex ratchet and pawl mechanism is not necessary.

Heat engine can be made using simple, asymmetrically shaped Brownian objects, such as a triangular cone.

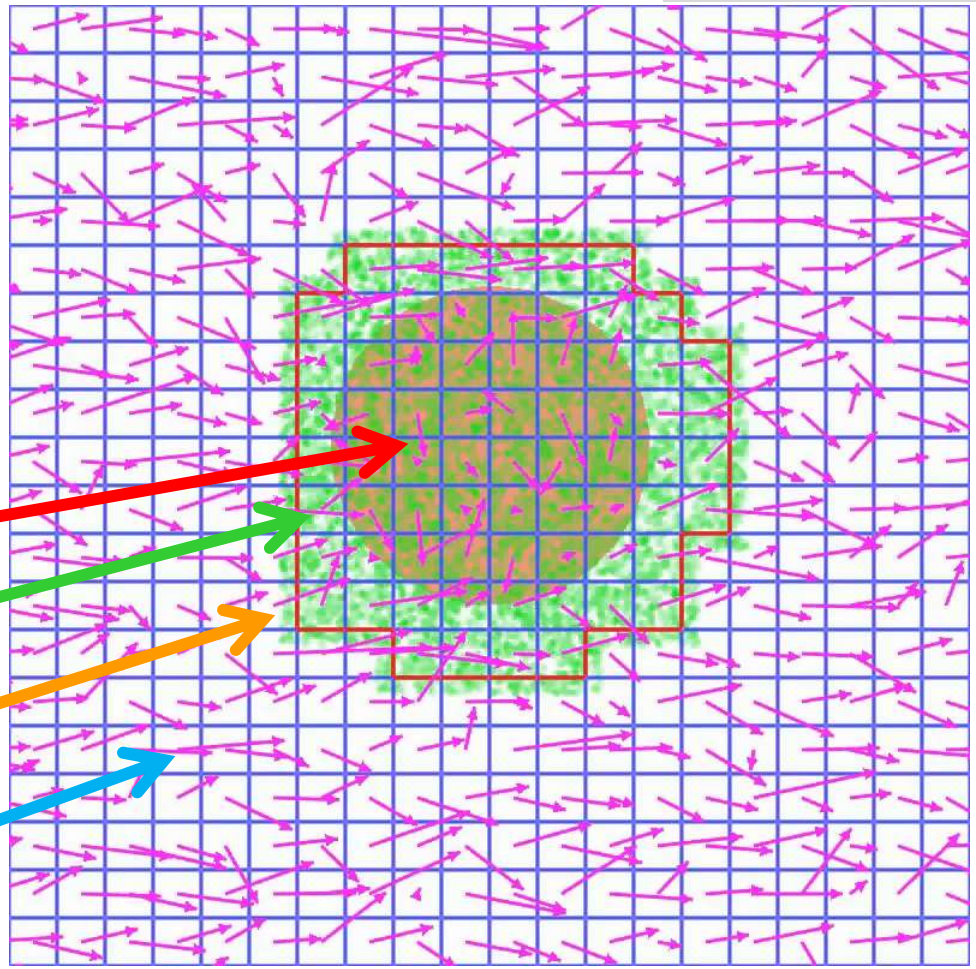


P. Meurs, C. Van den Broeck, and ALG, *Physical Review E* **70** 051109 (2004).

Particle/PDE Hybrid

Video courtesy of A. Donev

Particle/PDE Algorithm Refinement for flow past a sphere.



Stationary particle

Molecular simulation of solvent fluid

Interface

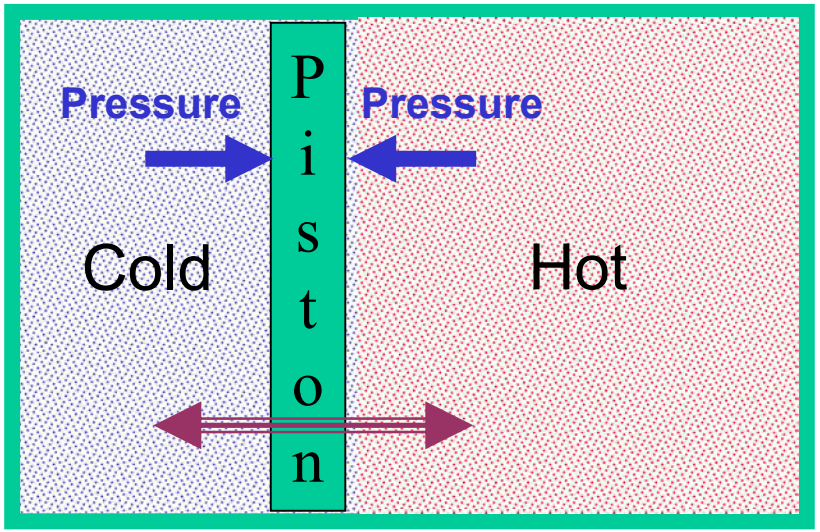
Continuum simulation of solvent fluid

Note: Continuum calculation done everywhere

Adiabatic Piston

Adiabatic piston is a classic problem in statistical mechanics.

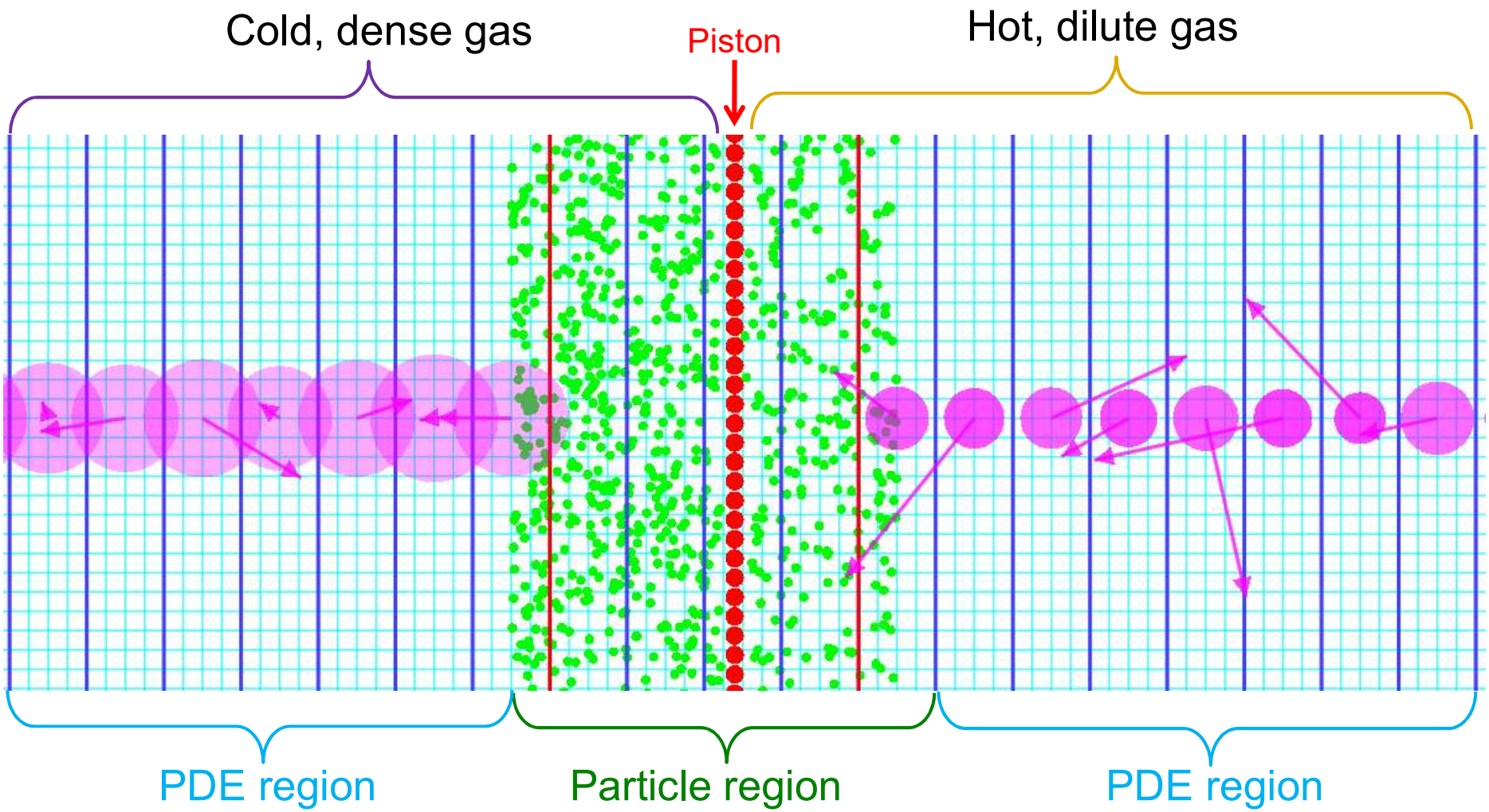
Chambers have gases at different temperatures but equal pressures.



Walls and piston are perfectly elastic to particle collisions yet the gases equilibrate to common temperature.

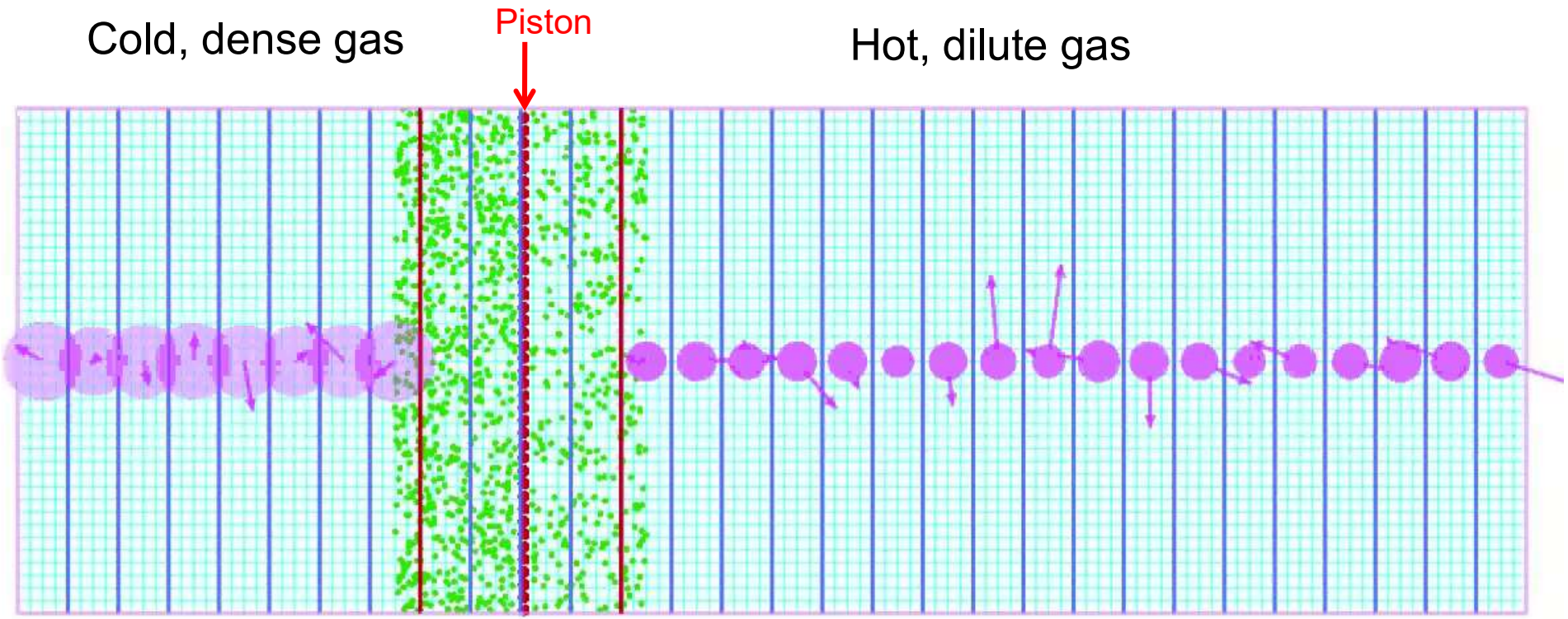
How? Heat is conducted between the chambers by the Brownian motion of the piston.

Adiabatic Piston



Initially the gas pressure is equal on both sides of the piston.

Sample Run of Adiabatic Piston

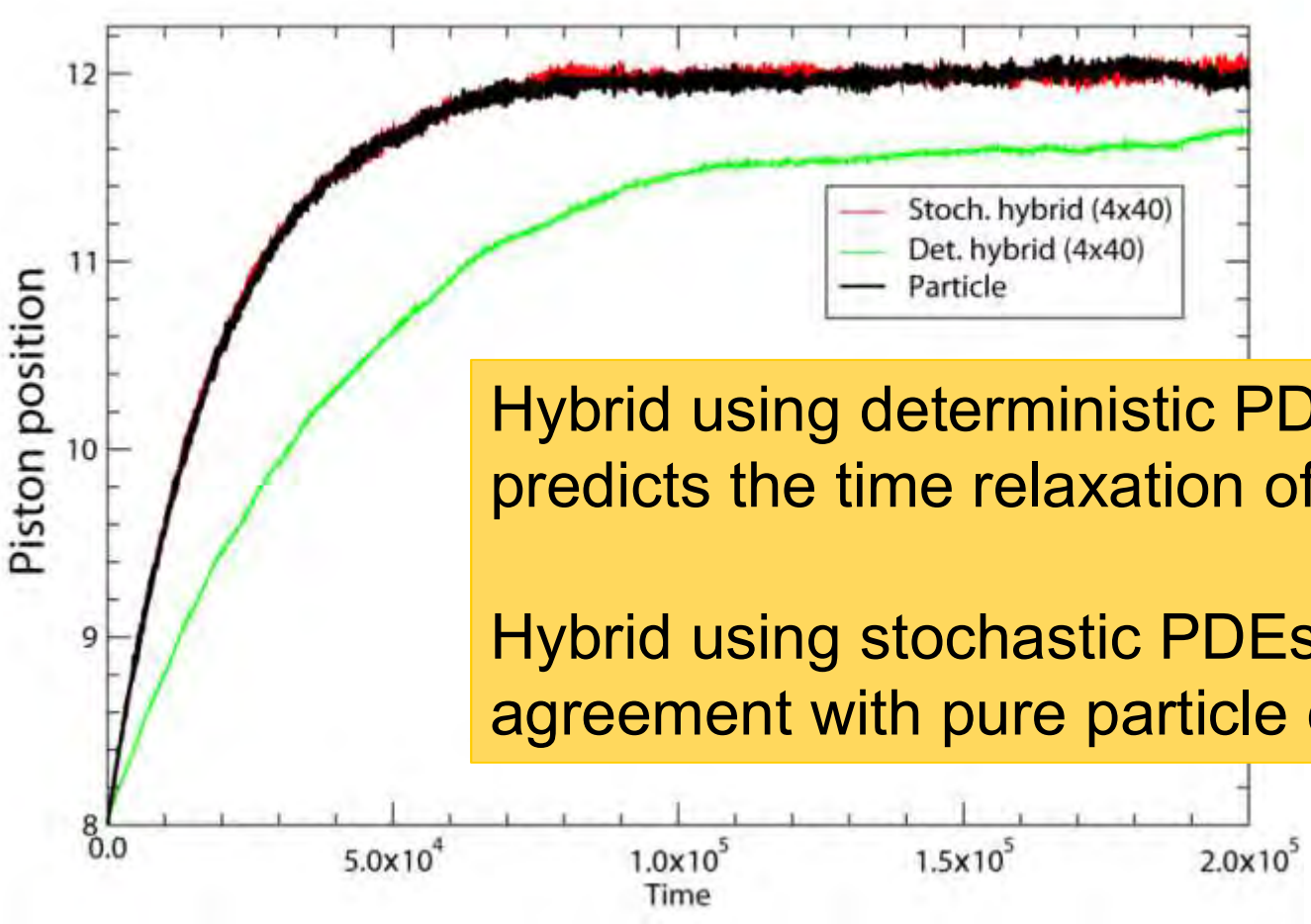


$\rho = {}^{4}/_{3}\ \rho_{eq}$
$T = {}^{3}/_{4}\ T_{eq}$

$\rho = {}^{3}/_{4}\ \rho_{eq}$
$T = {}^{4}/_{3}\ T_{eq}$

Video courtesy of A. Donev

Time Relaxation of the Piston

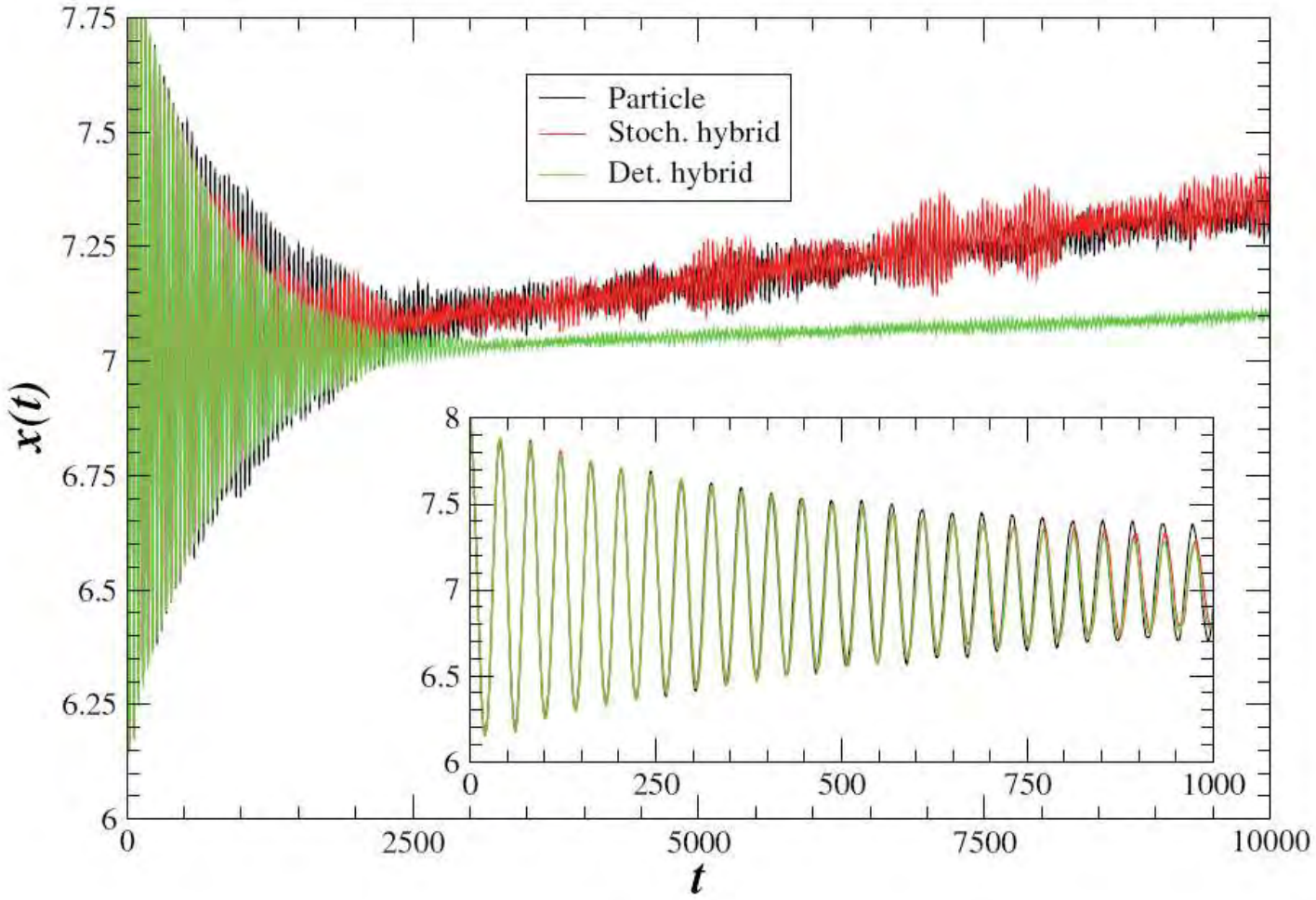


Hybrid using deterministic PDEs under-predicts the time relaxation of the piston.

Hybrid using stochastic PDEs in excellent agreement with pure particle calculations.

A. Donev, J.B. Bell, ALG, and B. Alder, *SIAM Multiscale Mod. Sim.* **8** 871-911 (2010).

Relaxation from Mechanical Non-Equilibrium



Summary & Remarks

Here are some closing thoughts:

- DSMC was developed for kinetic theory (Boltzmann equation) problems
- DSMC is useful for studying problems in fluctuating hydrodynamics
- As with all molecular methods, statistical measurements require careful attention
- Particle / Continuum hybrids are useful for specialized problems
- Thank you for your attention