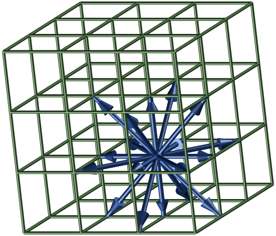
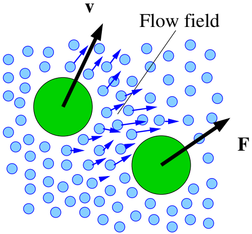
"Simulating Soft Matter with ESPResSo, ESPResSo++ and VOTCA"
ESPResSo Summer School 2012 @ ICP, Stuttgart University

Coupling Molecular Dynamics and Lattice Boltzmann

to simulate Hydrodynamics and Brownian motion





Ulf D. Schiller
u.schiller@fz-juelich.de

Institute of Complex Systems, Theoretical Soft Matter and Biophysics
Forschungszentrum Jülich

Overview

Scope of this lecture:

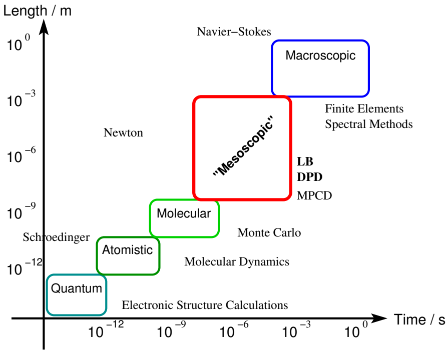
- Hydrodynamic interactions in soft matter
- Mesoscopic modeling
- Thermal fluctuations and Brownian motion

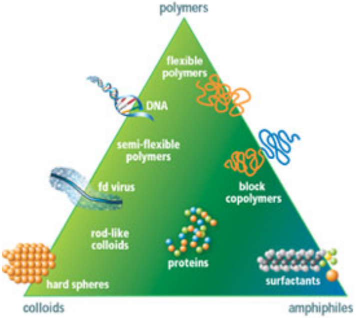
Method:

- Fluctuating lattice Boltzmann (FLB)

[B. Dünweg, UDS, A. J. C. Ladd, PRE 76, 036704 (2007)]
[B. Dünweg, UDS, A. J. C. Ladd, Comp. Phys. Comm. 180, 605 (2009)]

Time and length scales of (soft) matter





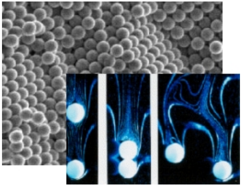
Source: IFF, FZ Jülich

- Mesoscopic scale bridges between microscopic and macroscopic scales
- Microhydrodynamics links between Newton and Navier-Stokes

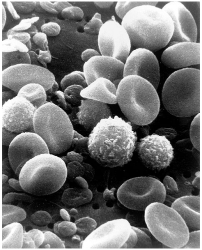
Complex fluids: Multiphase systems

Source: Wikipedia, GFDL

Source: Universiteit Utrecht



Source: Emory University

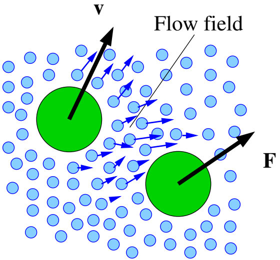


Source: Wikimedia

- Solutions, suspensions, emulsions: “contain” multiple length scales

$\longrightarrow$ Motion of the solutes and flow of the solvent are both important

Hydrodynamic interactions (HI)



Correlations:

$$\langle \Delta \mathbf{r}_i \otimes \Delta \mathbf{r}_j \rangle = 2\,\mathsf{D}_{ij}(\mathbf{r})\Delta t$$

Without HI:

$$\mathbf{v}_i = \frac{D_0}{k_B T}\mathbf{F}_i$$

With HI:

$$\mathbf{v}_i = \frac{1}{k_B T}\sum_{j \neq i} \mathsf{D}_{ij}(\mathbf{r})\mathbf{F}_j$$

Oseen tensor:

$$\mathsf{D}_{ij}(\mathbf{r}) = \frac{k_B T}{8\pi\eta\, r}\left(1 + \frac{\mathbf{r}\otimes\mathbf{r}}{r^2}\right)$$

→ Hydrodynamic interactions are long-ranged!

Do we need to include hydrodynamic interactions?

- Does a sailboat need sails?
- Hydrodynamics make a fluid a fluid!
- In many cases, long-range correlations due to HI can not be neglected. (Unless HI are screened.)
- There is no reason to neglect them in order to save computing time. (Algorithms have become reasonably fast.)

HI at microscopic level (Newton)

- equation of motion in the overdamped limit (neglect inertia)

$$\mathbf{r}_i(t+\Delta t) = \mathbf{r}_i(t) + \frac{\Delta t}{k_B T} \sum_{j \neq i} \mathrm{D}_{ij} \mathbf{F}_j(t) + \Delta \mathbf{r}_i$$

- correlation matrix

$$\left\langle \Delta \mathbf{r}_i \otimes \Delta \mathbf{r}_j \right\rangle = 2 \mathrm{D}_{ij} \Delta t$$

$\longrightarrow$ Brownian Dynamics (BD)

- difficulty: $\Delta \mathbf{r}_i$ requires matrix decomposition
- Cholesky: $\mathcal{O}(N^3)$, Chebychev expansion: $\mathcal{O}(N^{2.25})$, "P3M": $\mathcal{O}(N^{1.25} \ln N)$
- does not describe *explicit* momentum transport (often desired)

HI at macroscopic level (Navier-Stokes)

- Continuity equation

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0$$

- Navier-Stokes equation

$$\frac{\partial (\rho \mathbf{u})}{\partial t} + \nabla \cdot \Pi = \rho \mathbf{f}$$

- Stress tensor

$$\Pi = \underbrace{\rho c_s^2 1 + \frac{\mathbf{j} \otimes \mathbf{j}}{\rho}}_{\Pi^{\text{eq}}} + \underbrace{\eta : \left(\nabla \otimes \frac{\mathbf{j}}{\rho} \right)}_{\Pi^{\text{visc}}} + \Pi^{\text{fluct}}$$

- nonlinear partial differential equation

Low Reynolds number: Stokes flow

- incompressible Navier-Stokes equation (dimensionless form)

$$Re\left(\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v}\cdot\nabla)\mathbf{v}\right) = -\nabla p + \nabla^2 \mathbf{v} + \mathbf{f}$$

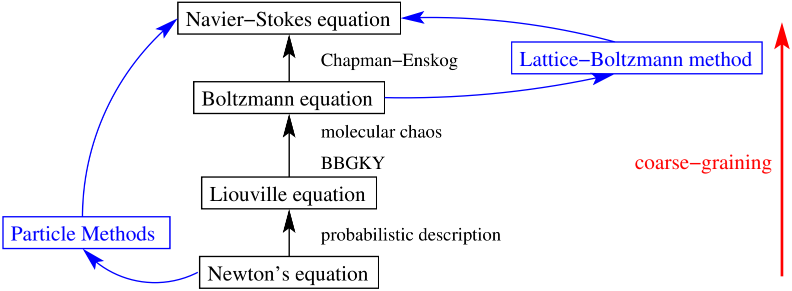
- $Re = \rho v L/\eta$ small $\rightarrow$ neglect substantial derivative (inertia)

$\rightarrow$ Stokes equation (dimensions reintroduced)

$$\begin{aligned} \nabla\cdot\sigma &= -\nabla p + \eta\nabla^2\mathbf{v} = -\rho\mathbf{f} \\ \nabla\cdot\mathbf{v} &= 0 \end{aligned}$$

- boundary conditions $\rightarrow$ hard to solve for complex fluids

From Newton to Navier-Stokes



→ Reduce the number of degrees of freedom by *eliminating fast variables*

Mesoscopic modeling for hydrodynamics

- hydrodynamic interactions: require conservation of mass and momentum
- properties of the solvent: diffusion coefficient, viscosity, temperature,...
- correct thermodynamics: required at least in equilibrium

Overview of methods

- Brownian dynamics (BD)
- Direct simulation Monte Carlo (DSMC)
- Multi-particle collision dynamics (MPC)
- Dissipative particle dynamics (DPD)
- Lattice gas automata (LGA)
- Lattice Boltzmann (LB)

Implicit solvent (BD) vs. explicit solvent (LB)

BD	LB
$Sc = \infty$	$Sc \gg 1$
$Ma = 0$	$Ma \ll 1$
$Re = 0$	$Re \ll 1$
$Bo > 0$	$Bo > 0$

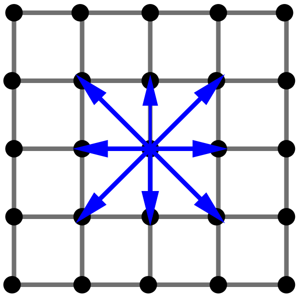
- Schmidt number $Sc = \nu/D$ (diffusive momentum transport vs. diffusive mass transport)
- Mach number $Ma = v/c$ (flow velocity vs. speed of sound; importance of fluid compressibility)
- Reynolds number $Re = vL/\nu$ (convective vs. diffusive momentum transport)
- "Boltzmann number" Bo: $\Delta x/x$ (thermal fluctuation vs. mean value, on the scale of an effective degree of freedom – depends on the degree of coarse-graining!)
- Remark: For particle methods, $Bo = O(1)$; not so for discretized field theories!

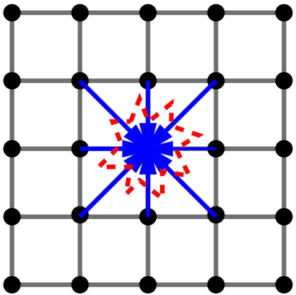
Lattice Boltzmann

- Hardy, Pomeau, de Pazzis (1973): 2D lattice gas model (HPP)
- Frisch, Hasslacher, Pomeau (1986): **lattice gas automaton (FHP)**
- d'Humières, Lallemand, Frisch (1986): 3D lattice gas automaton
- McNamara and Zanetti (1988): **lattice Boltzmann**
- Higuera and Jimenez (1989): linear collision operator
- Koelman (1991): lattice BGK
- Qian (1992): DnQm models
- d'Humières, Luo and coworkers (1992-): multi-relaxation time models
- Karlin and coworkers (1998-): entropic lattice Boltzmann
- Ladd and coworkers (1993-): fluctuating lattice Boltzmann
- ...

Lattice Boltzmann

Historic origin: lattice gas automaton





Kinetic approach: The Boltzmann equation

- evolution equation for the (one-)particle distribution function

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\cdot\frac{\partial}{\partial \mathbf{r}} + \frac{\mathbf{F}}{m}\cdot\frac{\partial}{\partial \mathbf{v}}\right) f(\mathbf{r},\mathbf{v},t) = \mathscr{C}[f]$$

- Boltzmann collision operator

$$\mathscr{C}[f] = \int d\mathbf{v}_1 \int d\Omega\, \sigma(v_{\mathrm{rel}},\Omega)\, v_{\mathrm{rel}} \left[f(\mathbf{r},\mathbf{v}',t) f(\mathbf{r},\mathbf{v}_1',t) - f(\mathbf{r},\mathbf{v},t) f(\mathbf{r},\mathbf{v}_1,t)\right]$$

- Detailed balance

$$f(\mathbf{r},\mathbf{v}_1',t) f(\mathbf{r},\mathbf{v}_2',t) = f(\mathbf{r},\mathbf{v}_1,t) f(\mathbf{r},\mathbf{v}_2,t)$$

$\longrightarrow$ Equilibrium distribution (Maxwell-Boltzmann) $f = f^{\mathrm{eq}} + f^{\mathrm{neq}}$

$$\ln f^{\mathrm{eq}} = \gamma_0 + \gamma\mathbf{v} + \gamma_4\mathbf{v}^2$$

Macroscopic moments

- "average" of polynomials $\psi(\mathbf{v})$ in components of $\mathbf{v}$

$$m_\psi(\mathbf{r},t) = \int \psi(\mathbf{v}) f(\mathbf{r},\mathbf{v},t) d\mathbf{v}$$

- density, momentum density, stress tensor

$$\begin{aligned}
\rho(\mathbf{r},t) &= m \int f d\mathbf{v} \\
\mathbf{j}(\mathbf{r},t) &= m \int \mathbf{v} f d\mathbf{v} \\
\Pi(\mathbf{r},t) &= m \int \mathbf{v} \otimes \mathbf{v} f d\mathbf{v}
\end{aligned}$$

Separation of scales

- Observation: not all m_ψ show up in the macroscopic equations of motion
- ρ, $\mathbf{j}$ (and e) are collisional invariants

$$\int d\mathbf{r} d\mathbf{v} \frac{\delta m_{\rho,\mathbf{j},e}(f)}{\delta f} \mathscr{C}[f] = 0$$

- local equilibrium (Maxwell-Boltzmann) $f^{\mathrm{eq}}(\rho, \mathbf{j}, e)$
- Hydrodynamics describes variation of ρ and $\mathbf{j}$ (and e) through *transport* (over a macroscopic distance $\sim L$)
- all other variables relax rapidly through *collisions* ($\sim \lambda$ mean free path)

$\rightarrow$ scale separation: $\varepsilon \sim Kn = \frac{\lambda}{L} \ll 1$ Knudsen number $Kn = \frac{\lambda}{L}$

How can we exploit the scale separation?

- we are only interested in the dynamics of the slow variables up to a certain order
- the dynamics of the fast variables beyond that order is unimportant
- any set of fast variables that leaves the slow dynamics unchanged will do

→ the number of degrees of freedom can be greatly reduced!

- Caveat: imperfect scale separation → fast variables can couple to slow dynamics

⏩ skip derivation

Discretization à la Grad

- Truncated Hermite expansion

$$f^N(\mathbf{r},\mathbf{v},t) = \omega(\mathbf{v}) \sum_{n=0}^{N} \frac{1}{n!} \mathrm{a}^{(n)}(\mathbf{r},t)\mathscr{H}^{(n)}(\mathbf{v})$$

$$(a^{(0)} = \rho,\ a^{(1)} = \mathbf{j},\ a^{(2)} = \Pi - \rho 1,\ \ldots)$$

- Gauss-Hermite quadrature

$$\mathrm{a}^{(n)} = \int \mathscr{H}^{(n)}(\mathbf{v}) f^N(\mathbf{r},\mathbf{v},t)\, d\mathbf{v} = \sum_i w_i \frac{\mathscr{H}^{(n)}(\mathbf{c}_i) f^N(\mathbf{r},\mathbf{c}_i,t)}{\omega(\mathbf{c}_i)}$$

$$= \sum \mathscr{H}^{(n)}(\mathbf{c}_i) f_i(\mathbf{r},t)$$

$\longrightarrow$ Discrete velocity machine (DVM)

$$\partial_t f_i + c_{i\alpha}\partial_\alpha f_i = -\lambda(f_i - f_i^{\mathrm{eq}}).$$

Space-time discretization

$$\frac{df_i}{dt} + \lambda f_i = \lambda f_i^{\mathrm{eq}}.$$

- Integration

$$f_i(\mathbf{r} + \tau \mathbf{c}_i, t + \tau) = e^{-\lambda \tau} f_i(\mathbf{r}, t) + \lambda e^{-\lambda \tau} \int_0^{\tau} e^{\lambda t'} f_i^{\mathrm{eq}}(\mathbf{r} + t' \mathbf{c}_i, t + t')\, dt'$$

- Expansion

$$f_i^{\mathrm{eq}}(\mathbf{r} + t' \mathbf{c}_i, t + t') = f_i^{\mathrm{eq}}(\mathbf{r}, t) + t' \frac{f_i^{\mathrm{eq}}(\mathbf{r} + \tau \mathbf{c}_i, t + \tau) - f_i^{\mathrm{eq}}(\mathbf{r}, t)}{\tau} + \mathscr{O}(\tau^2)$$

$\longrightarrow$ Fully discretized Boltzmann equation = Lattice Boltzmann

$$f_i(\mathbf{r} + \tau \mathbf{c}_i, t + \tau) = f_i(\mathbf{r}, t) - \lambda \left[f_i(\mathbf{r}, t) - f_i^{\mathrm{eq}}(\mathbf{r}, t) \right]$$

Quadratures

Quadrature	LB model	q	b_q	w_q	$\mathbf{c}_q$
$E_{1,5}^{3}$	D1Q3	0	1	$\frac{2}{3}$	0
		1	2	$\frac{1}{6}$	$\pm\sqrt{3}$
$E_{2,5}^{9}$	D2Q9	0	1	$\frac{4}{9}$	$(0,0)$
		1	4	$\frac{1}{9}$	$(\pm\sqrt{3},0),(0,\pm\sqrt{3})$
		2	4	$\frac{1}{36}$	$(\pm\sqrt{3},\pm\sqrt{3})$
$E_{3,5}^{15}$	D3Q15	0	1	$\frac{2}{9}$	$(0,0,0)$
		1	6	$\frac{1}{9}$	$(\pm\sqrt{3},0,0),(0,\pm\sqrt{3},0),(0,0,\sqrt{3})$
		3	8	$\frac{1}{72}$	$(\pm\sqrt{3},\pm\sqrt{3},\pm\sqrt{3})$
$E_{3,5}^{19}$	D3Q19	0	1	$\frac{1}{3}$	$(0,0,0)$
		1	6	$\frac{1}{18}$	$(\pm\sqrt{3},0,0),(0,\pm\sqrt{3},0),(0,0,\sqrt{3})$
		2	12	$\frac{1}{36}$	$(\pm\sqrt{3},\pm\sqrt{3},0),(\pm\sqrt{3},0,\pm\sqrt{3}),(0,\pm\sqrt{3},\pm\sqrt{3})$
$E_{3,5}^{27}$	D3Q27	0	1	$\frac{8}{27}$	$(0,0,0)$
		1	6	$\frac{2}{27}$	$(\pm\sqrt{3},0,0),(0,\pm\sqrt{3},0),(0,0,\sqrt{3})$
		2	12	$\frac{1}{54}$	$(\pm\sqrt{3},\pm\sqrt{3},0),(\pm\sqrt{3},0,\pm\sqrt{3}),(0,\pm\sqrt{3},\pm\sqrt{3})$
		3	8	$\frac{1}{216}$	$(\pm\sqrt{3},\pm\sqrt{3},\pm\sqrt{3})$

Notation $E_{D,d}^{n}$: D dimensions, d degree, n abscissae

q: neighbor shell, b_q: number of neighbors, w_q weights, $\mathbf{c}_q$ velocities

$$\mathrm{T}^{(n)} = \sum_i w_i \mathbf{c}_i \ldots \mathbf{c}_i = \begin{cases} 0 & n \text{ odd} \\ \delta^{(n)} & n \text{ even} \end{cases}, \quad \forall n \leq d.$$

Models with polynomial equilibrium

- Ansatz: expansion in the velocities **u** (Euler stress is quadratic in **u**)

$$f_i^{\text{eq}}(\rho, \mathbf{u}) = w_i \rho \left[1 + A\mathbf{u}\cdot\mathbf{c}_i + B(\mathbf{u}\cdot\mathbf{c}_i)^2 + C\mathbf{u}^2\right]$$

- cubic symmetry of lattice tensors $\mathrm{T}^{(n)}$

$$\sum_i w_i = 1 \qquad \sum_i w_i c_{i\alpha} = 0$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} = \sigma_2\, \delta_{\alpha\beta} \qquad \sum_i w_i c_{i\alpha} c_{i\beta} c_{i\gamma} = 0$$

$$\sum_i w_i c_{i\alpha} c_{i\beta} c_{i\gamma} c_{i\delta} = \kappa_4\, \delta_{\alpha\beta\gamma\delta} + \sigma_4 \left(\delta_{\alpha\beta}\delta_{\gamma\delta} + \delta_{\alpha\gamma}\delta_{\beta\delta} + \delta_{\alpha\delta}\delta_{\beta\gamma}\right)$$

$\longrightarrow$ at least three shells required to satisfy the conditions

$$\sum_i w_i = 1 \qquad \kappa_4 = 0 \qquad \sigma_4 = \sigma_2^2 \qquad c_s^2 = \sigma_2$$

The lattice Boltzmann equation

- recall the linear Boltzmann equation

$$\left(\frac{\partial}{\partial t} + \mathbf{v}\cdot\frac{\partial}{\partial \mathbf{r}}\right) f(\mathbf{r},\mathbf{v},t) = \mathscr{L}\left[f(\mathbf{r},\mathbf{v},t) - f^{\mathrm{eq}}(\mathbf{v})\right]$$

$f(\mathbf{r},\mathbf{v},t)$: distribution function
$f^{\mathrm{eq}}(\mathbf{v})$: Maxwell-Boltzmann distribution
$\mathscr{L}$: linear collision operator

- systematic discretization → lattice Boltzmann equation

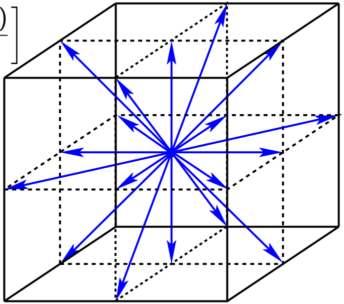
$$f_i(\mathbf{r}+\tau\mathbf{c}_i, t+\tau) = f_i^*(\mathbf{r},t) = f_i(\mathbf{r},t) + \sum_j \mathscr{L}_{ij}\left[f_j(\mathbf{r},t) - f_j^{\mathrm{eq}}(\rho,\mathbf{u})\right]$$

$f_i(\mathbf{r},t)$: population number
$\mathbf{r}$: discrete lattice point
$f_i^{\mathrm{eq}}(\rho,\mathbf{u})$: equilibrium distribution
τ: discrete time step
$\mathbf{c}_i$: discrete velocity vector
$\mathscr{L}_{ij}$: collision matrix

The D3Q19 model

Equilibrium distribution:

$$f_i^{\text{eq}}(\rho,\mathbf{u}) = w_i\rho\left[1+\frac{\mathbf{u}\cdot\mathbf{c}_i}{c_s^2}+\frac{\mathbf{uu}:(\mathbf{c}_i\mathbf{c}_i-c_s^2 1)}{2c_s^4}\right]$$



Moments:

$$\sum_i f_i^{\text{eq}} = \rho$$

$$\sum_i f_i^{\text{eq}}\mathbf{c}_i = \rho\mathbf{u}$$

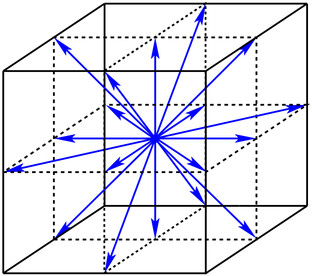
$$\sum_i f_i^{\text{eq}}\mathbf{c}_i\mathbf{c}_i = \rho c_s^2 1 + \rho\mathbf{uu}$$

Weight coefficients:

$w_i = 1/3$ for $|\mathbf{c}_i| = 0$, $w_i = 1/18$ for $|\mathbf{c}_i| = 1$, $w_i = 1/36$ for $|\mathbf{c}_i| = \sqrt{2}$

Speed of sound: $c_s = \frac{1}{\sqrt{3}}\left(\frac{a}{\tau}\right)$

The LB algorithm


D3Q19 lattice

1. streaming step: move $f_i^*(\mathbf{r},t)$ along $\mathbf{c}_i$ to the next lattice site, increment t by τ

$$f_i(\mathbf{r}+\tau\mathbf{c}_i, t+\tau) = f_i^*(\mathbf{r},t)$$

2. collision step: apply $\mathscr{L}_{ij}$ and compute the post-collisional $f_i^*(\mathbf{r},t)$ on every lattice site

$$f_i^*(\mathbf{r},t) = f(\mathbf{r},t) + \sum_j \mathscr{L}_{ij}\left[f_j(\mathbf{r},t) - f_j^{\mathrm{eq}}(\rho,\mathbf{u})\right]$$

Hydrodynamic moments in lattice Boltzmann

- hydrodynamic fields are velocity moments of the populations

$$\rho = \sum_i f_i \qquad \rho\mathbf{u} = \sum_i f_i \mathbf{c}_i \qquad \Pi = \sum_i f_i \mathbf{c}_i \otimes \mathbf{c}_i$$

- construct orthogonal basis e_{ki} for moments (recall $\psi(\mathbf{v})$ and m_ψ)

$$m_k = \sum_i e_{ki} f_i$$

$0 \leq k \leq 9$: hydrodynamic modes (slow), $k \geq 10$: kinetic modes (fast)

- collision matrix is diagonal in mode space

$$\mathscr{L}(\mathbf{f} - \mathbf{f}^{\mathrm{eq}}) = \mathrm{M}^{-1}\left(\mathrm{M}\mathscr{L}\mathrm{M}^{-1}\right)\mathrm{M}(\mathbf{f} - \mathbf{f}^{\mathrm{eq}}) = \mathrm{M}^{-1}\hat{\mathscr{L}}(\mathbf{m} - \mathbf{m}^{\mathrm{eq}})$$

$\rightarrow$ **MRT model**

$$(m_k - m_k^{\mathrm{eq}})^* = \gamma_k (m_k - m_k^{\mathrm{eq}})$$

Choice of the moment basis

$m_0 = \rho = \sum_i f_i$	mass
$m_1 = j_x = \sum_i f_i c_{ix}$	momentum x
$m_2 = j_y = \sum_i f_i c_{iy}$	momentum y
$m_3 = j_z = \sum_i f_i c_{iz}$	momentum z
$m_4 = \mathrm{tr}(\Pi)$	bulk stress
$m_5, \ldots, m_9 \simeq \overline{\Pi}$	shear stresses
$m_{10}, \ldots, m_{18}$	“kinetic modes”, “ghost modes”

Multiple relaxation time model (MRT)

$\gamma_0 = \gamma_1 = \gamma_2 = \gamma_3 = 0$ mass and momentum conservation

$\gamma_4 = \gamma_b$ bulk stress

$\gamma_5 = \ldots = \gamma_9 = \gamma_s$ shear stress

$\gamma_{10} = \ldots = \gamma_{18} = 0$ simplest choice, careful with boundaries!

- Remark: we could also relax the populations directly:

$$f_i^{\text{neq}*} = \sum_j \mathscr{L}_{ij} f_j^{\text{neq}}$$

- simplest choice $\mathscr{L}_{ij} = \lambda^{-1}\delta_{ij} \rightarrow$ lattice BGK
- not a particularly good choice (less stable, less accurate)

Viscous stress relaxation

$$\Pi = \overline{\Pi} + \frac{1}{3}\mathrm{tr}(\Pi)1$$

- recall: collision step applies linear relaxation to the moments

$$\begin{aligned} \overline{\Pi}^{*\mathrm{neq}} &= \gamma_s \overline{\Pi}^{\mathrm{neq}} \\ \mathrm{tr}(\Pi^{*\mathrm{neq}}) &= \gamma_b \mathrm{tr}(\Pi^{\mathrm{neq}}) \end{aligned}$$

- Chapman-Enskog expansion leads to

▸ Chapman-Enskog

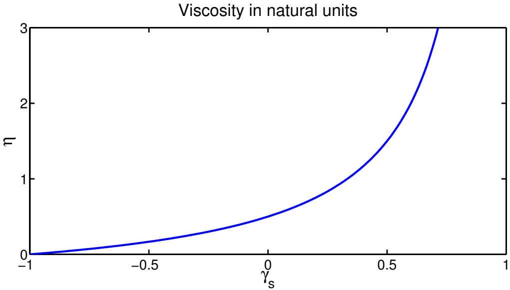
$$-\frac{1}{2}(\Pi^{*\mathrm{neq}} + \Pi^{\mathrm{neq}}) = \sigma = \eta\left(\nabla\mathbf{u} + (\nabla\mathbf{u})^t\right) + \left(\eta_b - \frac{2}{3}\eta\right)(\nabla\cdot\mathbf{u})1$$

→ shear and bulk viscosities are determined by the relaxation parameters

$$\eta = \frac{\rho c_s^2 \tau}{2}\frac{1+\gamma_s}{1-\gamma_s} \qquad \eta_b = \frac{\rho c_s^2 \tau}{3}\frac{1+\gamma_b}{1-\gamma_b}$$

Viscosity of the lattice Boltzmann fluid

- incompressible Navier-Stokes equation is recovered



- $-1 \leq \gamma_s \leq 1 \Leftrightarrow$ positive viscosities

$\longrightarrow$ any viscosity value is accessible

Units in LB

- grid spacing a, time step τ, particle mass m_p
- these merely control the *accuracy* and *stability* of LB!
- physically relevant: mass density ρ, viscosity η, temperature $k_B T$
- recall:

$$c_s^2 = \frac{1}{3}\frac{a^2}{\tau^2} = \hat{c}_s^2 \frac{a^2}{\tau^2}$$

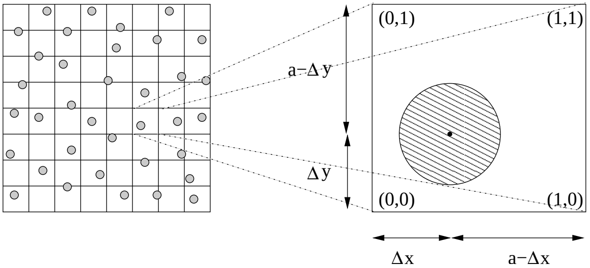
$$\eta = \frac{\rho c_s^2 \tau}{2} \frac{1+\gamma_s}{1-\gamma_s} = \hat{\rho}\hat{c}_s^2 \hat{\eta} \frac{m_p}{a\tau}$$

$$k_B T = m_p c_s^2 = m_p \hat{c}_s^2 \frac{a^2}{\tau^2}$$

→ always make sure you are simulating the right *physics*!

→ for comparison with experiments: match dimensionless numbers!
(Ma, Re, Pe, Sc, Kn, Pr, Wi, De, ...)

Coupling of particles and fluid



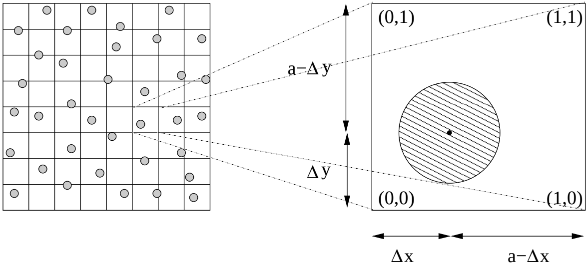
[Ahlrichs and Dünweg, J. Chem. Phys. 111, 8225 (1999)]

- Idea: treat monomers as point particles and apply Stokesian drag

$$\mathbf{F} = -\zeta\,[\mathbf{V} - \mathbf{u}(\mathbf{R}, t)] + \mathbf{f}_{\text{stoch}}$$

- ensure momentum conservation by transferring momentum to the fluid
- dissipative force
  → add stochastic force to fulfill fluctuation-dissipation relation

Coupling of particles and fluid



[Ahlrichs and Dünweg, J. Chem. Phys. 111, 8225 (1999)]

- interpolation scheme

$$\mathbf{u}(\mathbf{R}, t) = \sum_{\mathbf{x}\in\text{Cell}} \delta_{\mathbf{x}} \mathbf{u}(\mathbf{x}, t)$$

- momentum transfer

$$-\frac{\Delta t}{a^3}\mathbf{F} = \Delta\mathbf{j} = \frac{\mu}{a^2\tau}\sum_{\mathbf{x}\in\text{Cell}}\sum_i \Delta f_i(\mathbf{x}, t)\mathbf{c}_i$$

"Bare" vs. effective friction constant

- the input friction ζ_{bare} is not the real friction

- $D_0 > k_B T/\zeta_{\text{bare}}$ (due to long time tail)

$$\mathbf{V} = \frac{1}{\zeta_{\text{bare}}}\mathbf{F} + \mathbf{u}_{av} \qquad \mathbf{u} \approx \frac{1}{8\pi\eta r}(1 + \hat{r}\otimes\hat{r})\mathbf{F} \qquad \mathbf{u}_{av} = \frac{1}{g\eta a}\mathbf{F}$$

$$\frac{1}{\zeta_{\text{eff}}} = \frac{1}{\zeta_{\text{bare}}} + \frac{1}{g\eta a}$$

- Stokes contribution from interpolation with range a

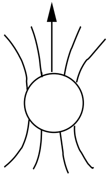
$\longrightarrow$ this *regularizes* the theory (no point particles in nature!)

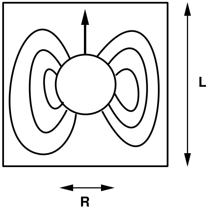
- ζ_{bare} has no physical meaning!

Finite size effects

Study diffusion / sedimentation of a single object

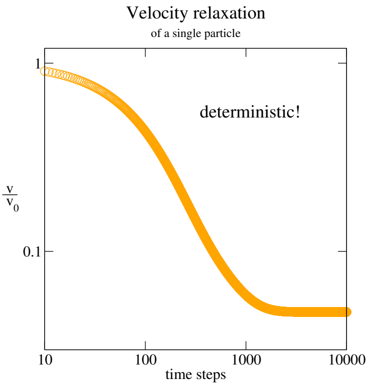
- $L = \infty$: $u(r) \sim 1/r$
- $F \sim \eta R v = \eta R^2 (v/R)$
- area R^2, shear gradient v/R
- backflow due to momentum conservation
- additional shear gradient v/L
- additional force $\eta R^2 (v/L) = \eta R v (R/L)$
- **finite size effect** $\sim R/L$

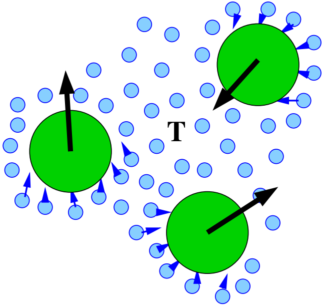




Thermal fluctuations

- so far the LB model is athermal and entirely deterministic
- for Brownian motion, we need fluctuations!





Do we need fluctuations?

If you go to the beach, do you bring a swimsuit?

- Ideal gas, temp. T, particle mass m_p, sound speed c_s:

$$k_B T = m_p c_s^2$$

- $c_s \sim a/h$ (a lattice spacing, h time step)
- c_s as small as possible

Example (water):
mass density $\rho = 10^3 kg/m^3$
sound speed realistic: $1.5 \times 10^3 m/s$
sound speed artificial: $c_s = 10^2 m/s$
temperature $T \approx 300K$, $k_B T = 4 \times 10^{-21}$
particle mass: $m_p = 4 \times 10^{-25} kg$

	macroscopic scale	molecular scale
lattice spacing	$a = 1mm$	$a = 1nm$
time step	$h = 10^{-5} s$	$h = 10^{-11} s$
mass of a site	$m_a = 10^{-6} kg$	$m_a = 10^{-24} kg$
"Boltzmann number"	$Bo = (m_p/m_a)^{1/2} = 6 \times 10^{-10}$	$Bo = (m_p/m_a)^{1/2} = 0.6$

Low Mach number physics

- LB requires $u \ll c_i$ hence $u \ll c_s$

$\longrightarrow$ low Mach number $Ma = u/c_s \ll 1$ $\rightarrow$ compressibility does not matter

$\longrightarrow$ equation of state does not matter $\rightarrow$ choose ideal gas!

m_p particle mass

$$p = \frac{\rho}{m_p} k_B T$$

$$c_s^2 = \frac{\partial p}{\partial \rho} = \frac{1}{m_p} k_B T$$

$$p = \rho c_s^2$$

$$k_B T = m_p c_s^2$$

Generalized lattice gas model (GLG)

- consider integer population numbers (m_p mass of an LB particle)

$$\nu_i = \frac{f_i}{\mu} \qquad \mu = \frac{m_p}{a^3} \qquad \boxed{\mu \nu_i = w_i \rho}$$

- each lattice site in contact with a heat bath

- Possion + constraints

$$P(\{\nu_i\}) \propto \prod_i \frac{\bar{\nu}_i^{\nu_i}}{\nu_i!} e^{-\bar{\nu}_i} \delta\left(\mu \sum_i \nu_i - \rho\right) \delta\left(\mu \sum_i \nu_i \mathbf{c}_i - \mathbf{j}\right)$$

[B. Dünweg, UDS, A. J. C. Ladd, PRE 76, 036704 (2007)]

Entropy

- associated entropy

$$P \propto \exp\left[S\left(\{\nu_i\}\right)\right] \delta\left(\mu \sum_i \nu_i - \rho\right) \delta\left(\mu \sum_i \nu_i \mathbf{c}_i - \mathbf{j}\right)$$

- Stirling: $\nu_i! = \exp\left(\nu_i \ln \nu_i - \nu_i\right)$

$$\begin{aligned} S\left(\{\nu_i\}\right) &= -\sum_i \left(\nu_i \ln \nu_i - \nu_i - \nu_i \ln \bar{\nu}_i + \bar{\nu}_i\right) \\ &= \frac{1}{\mu} \sum_i \rho w_i \left(\frac{f_i}{\rho w_i} - \frac{f_i}{\rho w_i} \ln \frac{f_i}{\rho w_i} - 1\right) \end{aligned}$$

$\rightarrow$ μ controls the mean square fluctuations (degree of coarse-graining)

Maximum entropy principle

- maximize entropy S subject to constraints for mass and momentum conservation

$$\frac{\partial S}{\partial \nu_i} + \chi + \lambda \cdot \mathbf{c}_i = 0 \qquad \mu \sum_i \nu_i - \rho = 0 \qquad \mu \sum_i \nu_i \mathbf{c}_i - \mathbf{j} = 0$$

- formal solution

$$f_i^{\text{eq}} = w_i \, \rho \exp\left(\chi + \lambda \cdot \mathbf{c}_i\right)$$

- expansion up to $\mathcal{O}(u^2)$

$$f_i^{\text{eq}}(\rho, \mathbf{u}) = w_i \rho \left[1 + \frac{\mathbf{u} \cdot \mathbf{c}_i}{c_s^2} + \frac{\mathbf{uu} \colon (\mathbf{c}_i \mathbf{c}_i - c_s^2 1)}{2 c_s^4} \right]$$

Fluctuations around equilibrium

- Gauss distribution for non-equilibrium part

$$P \propto \exp\left(-\sum_i \frac{\left(f_i^{\mathrm{neq}}\right)^2}{2\mu\rho w_i}\right) \delta\left(\sum_i f_i^{\mathrm{neq}}\right) \delta\left(\sum_i \mathbf{c}_i f_i^{\mathrm{neq}}\right)$$

- transform to the modes ($b_k = \sum_i w_i e_{ki}^2$, Basis e_{ki})

$$P\left(\left\{m_k^{\mathrm{neq}}\right\}\right) \propto \exp\left(-\sum_{k\geq 4} \frac{\left(m_k^{\mathrm{neq}}\right)^2}{2\mu\rho b_k}\right)$$

- more convenient: ortho-normal modes

$$\hat{m}_k = \sum_i \hat{e}_{ki} \frac{f_i}{\sqrt{w_i \mu\rho}}$$

Implementation of the fluctuations

- introduce stochastic term into the collision step

$$m_k^{*\mathrm{neq}} = \gamma_k m_k^{\mathrm{neq}} + \varphi_k r_k$$

r_k random number from normal distribution

- ensure detailed balance (like in Monte-Carlo)

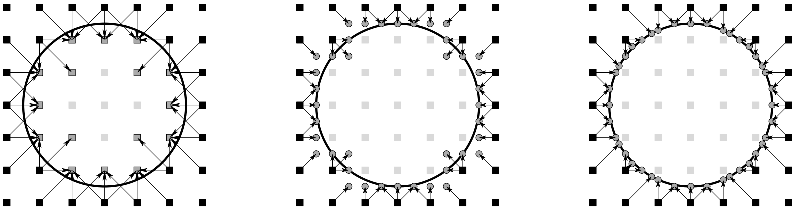
$$\frac{p(m \to m^*)}{p(m^* \to m)} = \frac{\exp(-m^{*2}/2)}{\exp(-m^2/2)} \qquad \Rightarrow \qquad \varphi_k = \sqrt{\mu \rho b_k \left(1 - \gamma_k^2\right)}$$

- **$\varphi_k \neq 0$ for all non-conserved modes**

$\rightarrow$ **all modes have to be thermalized**

[A. J. C. Ladd, JFM 271, 285–309 (1994)]
[Adhikari et al., EPL 71, 473-479 (2005)]
[B. Dünweg, UDS, A. J. C. Ladd, PRE 76, 036704 (2007)]

Lattice representation of rigid objects



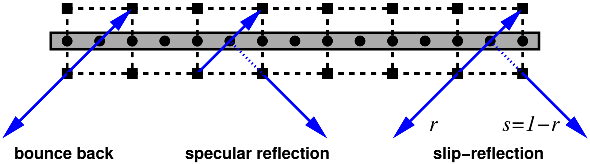
- determine the points where the surface of the rigid object intersects the lattice links

$\longrightarrow$ surface markers

"Accounting for these constraints may be trivial under idealized conditions [...] but generally speaking, it constitutes a very delicate (and sometimes nerve-probing!) task."

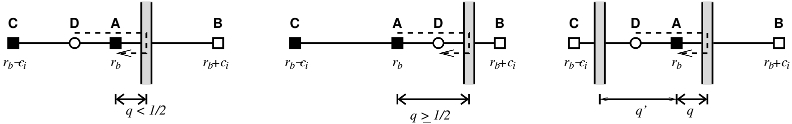
Sauro Succi

Boundary conditions



- these rules are simple to implement
- but they are only correct to first order
- the boundary location is always midway in between nodes

Interpolation boundary conditions

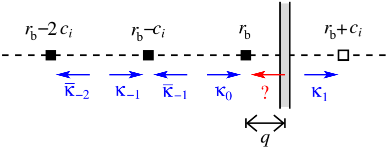


$$f_{i^-}(\mathbf{r}_B, t+\tau) = 2q f_i^*(\mathbf{r}_B, t) + (1-2q) f_i^*(\mathbf{r}_B - \tau \mathbf{c}_i, t), \qquad q < \frac{1}{2},$$

$$f_{i^-}(\mathbf{r}_B, t+\tau) = \frac{1}{2q} f_i^*(\mathbf{r}_B, t) + \frac{2q-1}{2q} f_{i^-}^*(\mathbf{r}_B, t), \qquad q \geq \frac{1}{2}.$$

[Bouzidi et al., Phys. Fluids 13, 3452 (2001)]

Multi-reflection boundary conditions



$$f_{i^-}(\mathbf{r}_B, t+\tau) = f_i^*(\mathbf{r}_B, t) - \frac{1-2q-2q^2}{(1+q)^2} f_{i^-}^*(\mathbf{r}_B, t) + \frac{1-2q-2q^2}{(1+q)^2} f_i^*(\mathbf{r}-\tau\mathbf{c}_i, t)$$
$$- \frac{q^2}{(1+q)^2} f_{i^-}^*(\mathbf{r}-\tau\mathbf{c}_i, t) + \frac{q^2}{(1+q)^2} f_i^*(\mathbf{r}-2\tau\mathbf{c}_i, t).$$

- match Taylor expansion at the boundary with Chapman-Enskog result

$\longrightarrow$ yields a condition for the relaxation rate of the kinetic modes

$$\lambda_g(\lambda_s) = -8\frac{2+\lambda}{8+\lambda}$$

[Ginzburg and d'Humières, Phys. Rev. E 68, 066614 (2003).]

Equilibrium interpolation

$$f_{i^-}^{\text{eq}}(\mathbf{r}_B, t+\tau) = 2q f_i^{\text{eq}}(\mathbf{r}_B, t) + (1-2q) f_i^{\text{eq}}(\mathbf{r}_B - \tau\mathbf{c}_i, t) \qquad q < \frac{1}{2}$$

$$f_{i^-}^{\text{eq}}(\mathbf{r}_B, t+\tau) = \frac{1-q}{q} f_i^{\text{eq}}(\mathbf{r}, t) + \frac{2q-1}{q} f_i^{\text{eq}}(\mathbf{r}_B + q\tau\mathbf{c}_i) \qquad q \geq \frac{1}{2}$$

$$f_{i^-}^{\text{neq}}(\mathbf{r}_B, t+\tau) = f_i^{\text{neq}}(\mathbf{r}_B, t)$$

[Chun and Ladd, Phys. Rev. E 75, 066705 (2007)]

- interpolation for equilibrium
- bounce-back for non-equilibrium
- non-equilibrium enters Chapman-Enskog one order later than equilibrium

$\longrightarrow$ still second order accurate!

Lattice Boltzmann in ESPResSo

```
setmd box_l $Lx $Ly $Lz
setmd periodic ...

cellsystem domain_decomposition -no_verlet_list

lbfluid density $lb_dens grid $lb_grid tau $lb_tau
lbfluid viscosity $lb_visc
lbfluid friction $lb_zeta
thermostat lb $temp

lb_boundary wall $px $py $pz normal $nx $ny $nz

integrate $nsteps

puts [analyze fluid mass]
puts [analyze fluid momentum]
```

Summary of lattice Boltzmann

- lattice kinetic approach to hydrodynamics
- easy to implement and to parallelize
- solid theoretical underpinning
- consistent thermal fluctuations
- beyond Navier-Stokes: possible but can get complicated
- challenges: non-ideal fluids, multi-phase fluids, thermal flows

Want more?

Dünweg and Ladd: "Lattice Boltzmann simulations of soft matter systems"
Adv. Polymer Sci. **221**, 89–166 (2009)

Aidun and Clausen: "Lattice-Boltzmann Method for Complex Flows"
Annu. Rev. Fluid Mech. **42**, 439–472 (2010)

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Thank you for your attention!

Eliminating fast variables: Chapman-Enskog

- introduce length and time scales

coarse-grained length: $\mathbf{r}_1 = \varepsilon \mathbf{r} \quad \rightarrow \quad \frac{\partial}{\partial \mathbf{r}} = \varepsilon \frac{\partial}{\partial \mathbf{r}_1}$

convective time scale: $t_1 = \varepsilon t$

diffusive time scale: $t_2 = \varepsilon^2 t \quad \rightarrow \quad \frac{\partial}{\partial t} = \varepsilon \frac{\partial}{\partial t_1} + \varepsilon^2 \frac{\partial}{\partial t_2}$

- use ε as a perturbation parameter and expand f

$$
\begin{aligned}
f &= f^{(0)} + \varepsilon f^{(1)} + \varepsilon^2 f^{(2)} + \ldots \\
\mathscr{C}[f] &= \mathscr{C}[f^{(0)}] + \varepsilon \int d\mathbf{r} d\mathbf{v} \frac{\delta C[f]}{\delta f} f^{(1)}(\mathbf{r}, \mathbf{v}) + \ldots
\end{aligned}
$$

$\rightarrow$ solve for each order in ε

Chapman-Enskog expansion

- ε^0: yields the collisional invariants, and the equilibrium distribution $f^{(0)} = f^{\mathrm{eq}}$

- ε^1: yields the Euler equations, and the first order correction $f^{(1)}$

$$\left(\frac{\partial}{\partial t_1} + \mathbf{v}\frac{\partial}{\partial \mathbf{r}_1}\right) f^{(0)} = \int d\mathbf{r}' d\mathbf{v}' \frac{\delta C[f^{(0)}]}{\delta f^{(0)}} f^{(1)}(\mathbf{r}', \mathbf{v}') \qquad (*)$$

- ε^2: adds viscous terms to the Euler equation

$\longrightarrow$ Navier-Stokes!

- the “only” difficulty is: no explicit solution of (*) is known...
(except for Maxwell molecules)

Chapman-Enskog expansion for LB

- original LBE

$$f_i(\mathbf{r}+\tau\mathbf{c}_i, t+\tau) - f_i(\mathbf{r}, t) = \Delta_i$$

- recall: coarse-grained length $\mathbf{r}_1$, convective time scale t_1, diffusive time scale t_2

$$f_i(\mathbf{r}_1+\varepsilon\tau\mathbf{c}_i, t_1+\varepsilon\tau, t_2+\varepsilon^2\tau) - f_i(\mathbf{r}_1, t_1, t_2) = \Delta_i$$

- Taylor expansion:

$$\varepsilon\tau\left(\frac{\partial}{\partial t_1}+\mathbf{c}_i\cdot\frac{\partial}{\partial \mathbf{r}_1}\right)f_i + \varepsilon^2\tau\left[\frac{\partial}{\partial t_2}+\frac{\tau}{2}\left(\frac{\partial}{\partial t_1}+\mathbf{c}_i\cdot\frac{\partial}{\partial \mathbf{r}_1}\right)\right]f_i = \Delta_i$$

Chapman-Enskog expansion for LB

- expand f_i and Δ_i in powers of ε

$$
\begin{aligned}
f_i &= f_i^{(0)} + \varepsilon f_i^{(1)} + \varepsilon^2 f_i^{(2)} + \dots \\
\Delta_i &= \Delta_i^{(0)} + \varepsilon \Delta_i^{(1)} + \dots
\end{aligned}
$$

- hierarchy of equations at different powers of ε

$$\mathscr{O}(\varepsilon^0): \quad \Delta_i^{(0)} = 0$$

$$\mathscr{O}(\varepsilon^1): \quad \left(\frac{\partial}{\partial t_1} + \mathbf{c}_i \cdot \frac{\partial}{\partial \mathbf{r}_1} \right) f_i^{(0)} = \frac{1}{\tau} \Delta_i^{(1)}$$

$$\mathscr{O}(\varepsilon^2): \quad \left[\frac{\partial}{\partial t_2} + \frac{\tau}{2} \left(\frac{\partial}{\partial t_1} + \mathbf{c}_i \cdot \frac{\partial}{\partial \mathbf{r}_1} \right)^2 \right] f_i^{(0)} + \left(\frac{\partial}{\partial t_1} + \mathbf{c}_i \cdot \frac{\partial}{\partial \mathbf{r}_1} \right) f_i^{(1)} = \frac{1}{\tau} \Delta_i^{(2)}$$

Zeroth order ε^0

- no expansion for conserved quantities!

$$\begin{aligned} f_i^{(0)} &= f_i^{\text{eq}} \\ \rho^{(0)} &= \rho = \sum_i f_i^{\text{eq}} \\ \mathbf{j}^{(0)} &= \mathbf{j} = \sum_i f_i^{\text{eq}} \mathbf{c}_i \end{aligned}$$

- linear part of collision operator

$$\Delta_i = \varepsilon \Delta_i^{(1)} = \varepsilon \sum_j \left. \frac{\partial \Delta_i}{\partial f_j} \right|_{f^{(0)}} f_j^{(1)} = \sum_j \mathscr{L}_{ij} f_j^{(1)}$$

Equations for the mass density

$$\frac{\partial}{\partial t_1}\rho + \frac{\partial}{\partial \mathbf{r}_1} \cdot \mathbf{j} = 0$$

$$\frac{\partial}{\partial t_2}\rho = 0$$

$\longrightarrow$ continuity equation holds!

Equations for the momentum density

$$\frac{\partial}{\partial t_1}\mathbf{j} + \frac{\partial}{\partial \mathbf{r}_1}\cdot \Pi^{(0)} = 0$$

$$\frac{\partial}{\partial t_2}\mathbf{j} + \frac{1}{2}\frac{\partial}{\partial \mathbf{r}_1}\cdot \left(\Pi^{*(1)} + \Pi^{(1)}\right) = 0$$

- Euler stress

$$\boxed{\Pi^{(0)} = \rho c_s^2 1 + \rho \mathbf{u}\mathbf{u} = \Pi^{\mathrm{eq}}}$$

- Newtonian viscous stress

$$\boxed{\tfrac{\varepsilon}{2}\left(\Pi^{*(1)} + \Pi^{(1)}\right) = -\Pi^{\mathrm{visc}}}$$

$\longrightarrow$ incompressible Navier-Stokes equation holds!

Diggin' deeper...

- the third moment $\Phi^{(0)} = \sum_i f_i^{(0)} \mathbf{c}_i \mathbf{c}_i \mathbf{c}_i$ enters through its equilibrium part!

$$\frac{\partial}{\partial t_1} \Pi^{(0)} + \frac{\partial}{\partial \mathbf{r}_1} \cdot \Phi^{(0)} = \frac{1}{\tau} \sum_i \Delta_i^{(1)} \mathbf{c}_i \mathbf{c}_i = \frac{1}{\tau} \left(\Pi^{*(1)} - \Pi^{(1)} \right)$$

- explicit form

$$\Phi^{(0)}_{\alpha\beta\gamma} = \rho c_s^2 \left(u_\alpha \delta_{\beta\gamma} + u_\beta \delta_{\alpha\gamma} + u_\gamma \delta_{\alpha\beta} \right)$$

- from continuity and Euler equation

$$\frac{\partial}{\partial t_1} \Pi^{(0)} = \frac{\partial}{\partial t_1} \left(\rho c_s^2 \mathbb{1} + \rho \mathbf{u}\mathbf{u} \right) = \ldots$$

- neglecting terms of $\mathscr{O}(u^3)$

$$\Pi^{*(1)} - \Pi^{(1)} = \rho c_s^2 \tau \left(\nabla \mathbf{u} + (\nabla \mathbf{u})^t \right)$$

Suitable LB models

- equilibrium values of the moments up to Φ^{eq}

$$
\begin{aligned}
\rho^{\text{eq}} &= \rho \\
\mathbf{j}^{\text{eq}} &= \mathbf{j} \\
\Pi^{\text{eq}} &= \rho c_s^2 1 + \rho \mathbf{u}\mathbf{u} \\
\Phi^{\text{eq}}_{\alpha\beta\gamma} &= \rho c_s^2 \left(u_\alpha \delta_{\beta\gamma} + u_\beta \delta_{\alpha\gamma} + u_\gamma \delta_{\alpha\beta} \right)
\end{aligned}
$$

- collision operator

$$\sum_i \Delta_i = 0 \qquad \sum_i \Delta_i \mathbf{c}_i = 0$$

$$\overline{\Pi}^{*\text{neq}} = \gamma_s \overline{\Pi}^{\text{neq}} \qquad \text{tr}(\Pi^{*\text{neq}}) = \gamma_b \text{tr}(\Pi^{\text{neq}})$$

Why all parts need to be thermalized?

- Equations of motion are stochastic differential equations
- Fokker-Planck formalism

$\rightarrow$ Kramers-Moyal expansion

$$\begin{aligned}
\text{Particle, conservative:} \quad \mathscr{L}_1 &= -\sum_i \left(\frac{\partial}{\partial \mathbf{r}_i} \cdot \frac{\mathbf{p}_i}{m_i} + \frac{\partial}{\partial \mathbf{p}_i} \cdot \mathbf{F}_i \right) \\
\text{Particle, Langevin:} \quad \mathscr{L}_2 &= \sum_i \frac{\Gamma_i}{m_i} \frac{\partial}{\partial \mathbf{p}_i} \mathbf{p}_i \\
\text{Particle, stochastic:} \quad \mathscr{L}_3 &= k_B T \sum_i \Gamma_i \frac{\partial^2}{\partial \mathbf{p}_i^2}
\end{aligned}$$

- Fluctuation-Dissipation relation

$$\left(\sum_i \mathscr{L}_i \right) \exp(-\beta \mathscr{H}) = 0$$

Why all parts need to be thermalized?

$$\text{Particle, conservative:} \quad \mathscr{L}_1 = -\sum_i \left(\frac{\partial}{\partial \mathbf{r}_i} \cdot \frac{\mathbf{p}_i}{m_i} + \frac{\partial}{\partial \mathbf{p}_i} \cdot \mathbf{F}_i \right)$$

$$\text{Fluid, conservative:} \quad \mathscr{L}_4 = \int d\mathbf{r} \left(\frac{\delta}{\delta\rho} \partial_\alpha j_\alpha + \frac{\delta}{\delta j_\alpha} \partial_\beta \Pi^{\text{eq}}_{\alpha\beta} \right)$$

$$\text{Fluid, viscous:} \quad \mathscr{L}_5 = \eta_{\alpha\beta\gamma\delta} \int d\mathbf{r} \frac{\delta}{\delta j_\alpha} \partial_\beta \partial_\gamma u_\delta$$

$$\text{Fluid, stochastic:} \quad \mathscr{L}_6 = k_B T \eta_{\alpha\beta\gamma\delta} \int d\mathbf{r} \frac{\delta}{\delta j_\alpha} \partial_\beta \partial_\gamma \frac{\delta}{\delta j_\delta}$$

$$\text{Particle, coupling:} \quad \mathscr{L}_7 = -\sum_i \zeta_i \frac{\partial}{\partial p_{i\alpha}} u_{i\alpha}$$

$$\text{Fluid, coupling:} \quad \mathscr{L}_8 = -\sum_i \zeta_i \int d\mathbf{r} \Delta(\mathbf{r},\mathbf{r}_i) \frac{\delta}{\delta j_\alpha(\mathbf{r})} \left(\frac{p_{i\alpha}}{m_i} - u_{i\alpha} \right)$$

$$\text{Fluid, stochastic:} \quad \mathscr{L}_9 = k_B T \sum_i \zeta_i \int d\mathbf{r} \Delta(\mathbf{r},\mathbf{r}_i) \frac{\delta}{\delta j_\alpha(\mathbf{r})} \int d\mathbf{r}' \Delta(\mathbf{r}',\mathbf{r}_i) \frac{\delta}{\delta j_\alpha(\mathbf{r}')}$$

$$\text{Particle, stochastic:} \quad \mathscr{L}_{10} = -2 k_B T \sum_i \zeta_i \frac{\partial}{\partial p_{i\alpha}} \int d\mathbf{r} \Delta(\mathbf{r},\mathbf{r}_i) \frac{\delta}{\delta j_\alpha(\mathbf{r})}$$