Atomistic, mesoscopic and continuum hydrodynamics

coupling liquid models with different resolution

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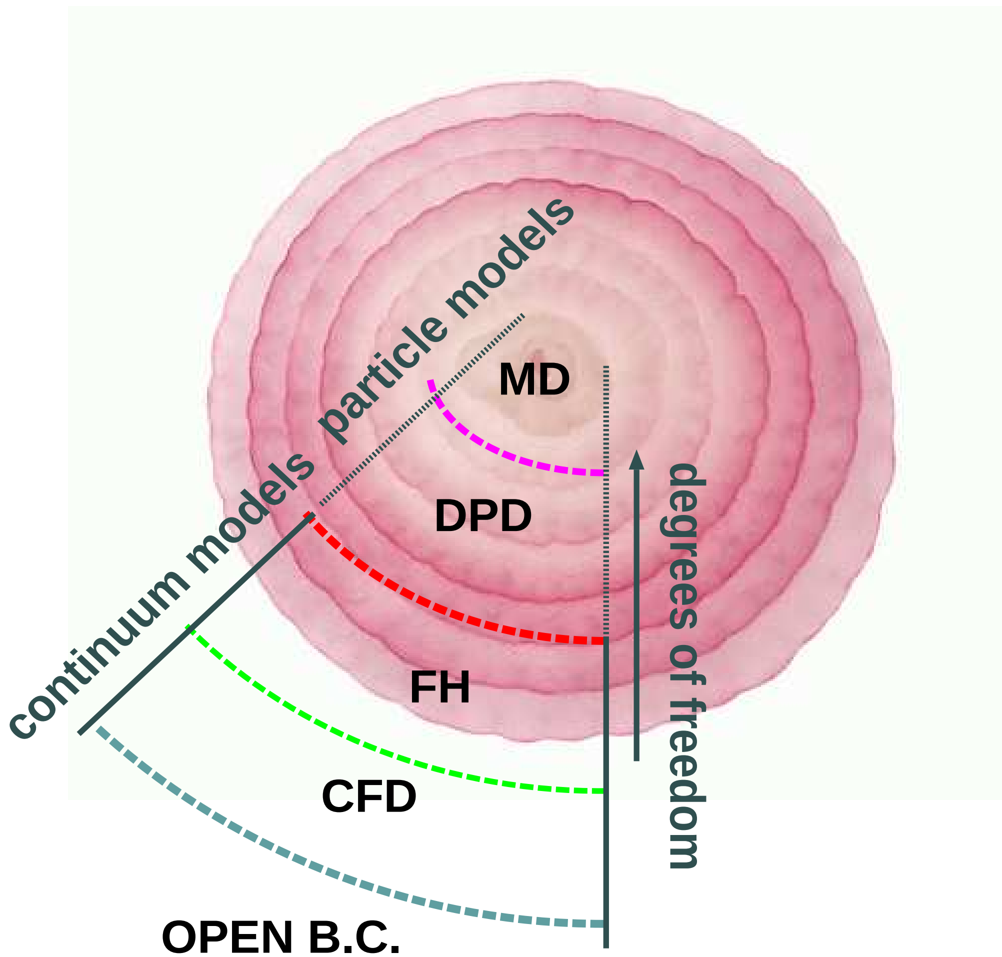
Domain decomposition

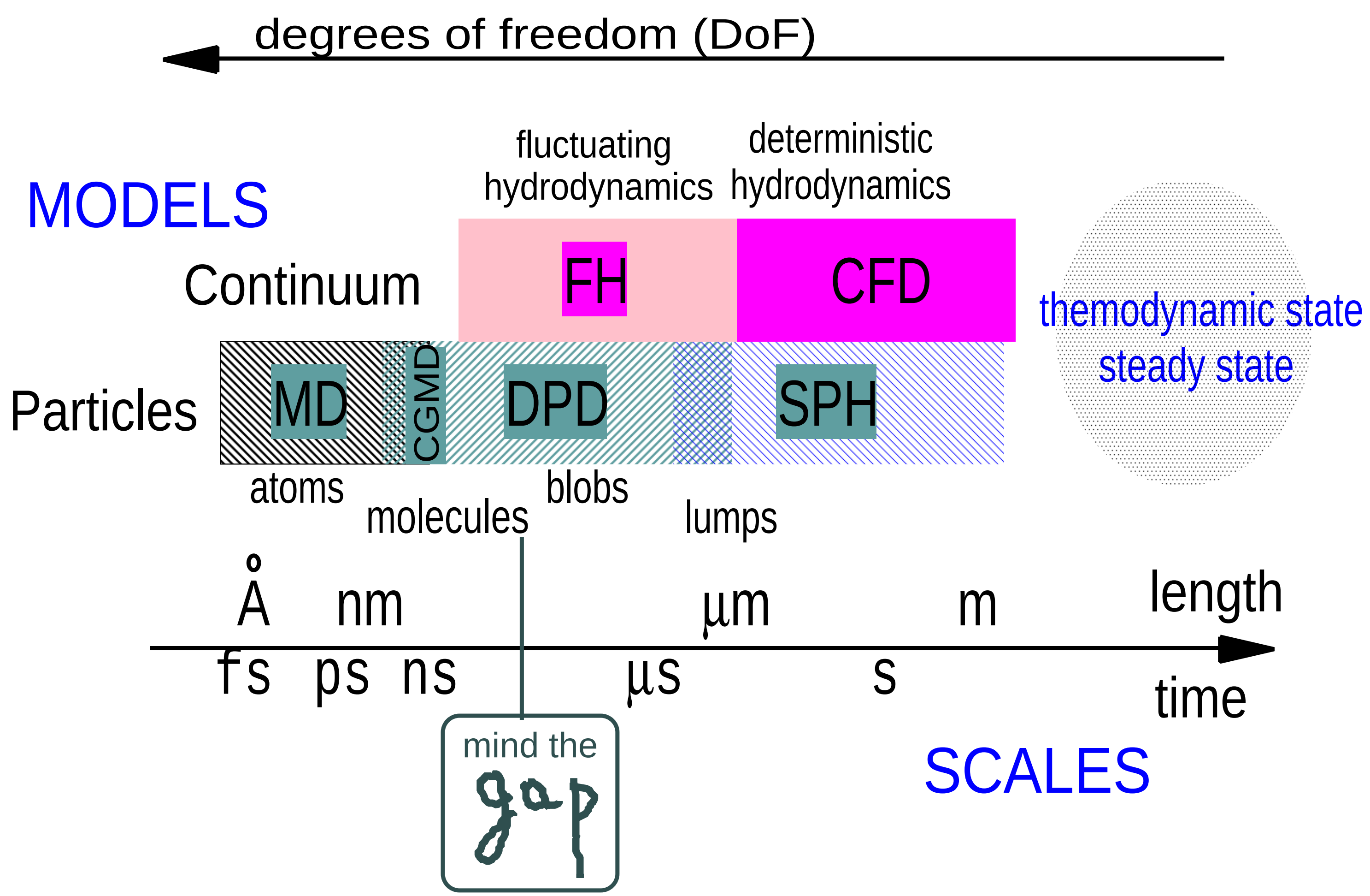
Interfacing models with different degrees of freedom



Open boundary conditions:
OUTSIDE WORLD
steady state,
thermodynamic reservoir

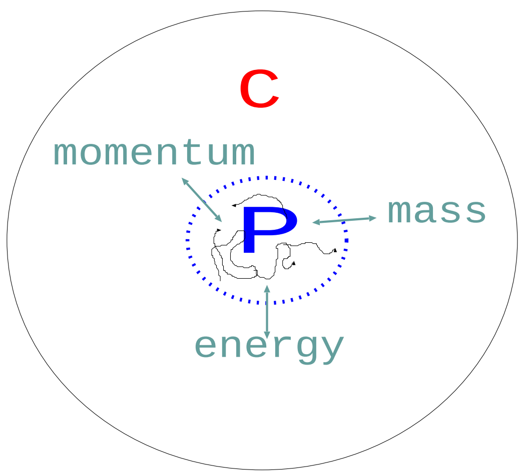
Scales and models

with hydrodynamics



Multiscale modelling: Motivation. Applications.

- Multiscale models: predicted as a scientific milestone in near future by the 2020 Science Group. [*Nature* **440** (7083): 383 (2006)]
- Complex fluids near interfaces: microfluidics, slip of liquid flow past surfaces.
- Fluid-fluid or soft interfaces (e.g., Rayleigh-Taylor instability, membrane's dynamics)
- Macromolecules-sound interaction (proteins) [*Science*, 309:1096, 2005.]
- Crystal growth from liquid phase.
- Wetting phenomena: microscopic treatment of the wetting front. Lubrication
- Confined systems: driven to chemical equilibrium, osmosis driven flows through membranes, thin films, water between membranes, clays,
- etc...

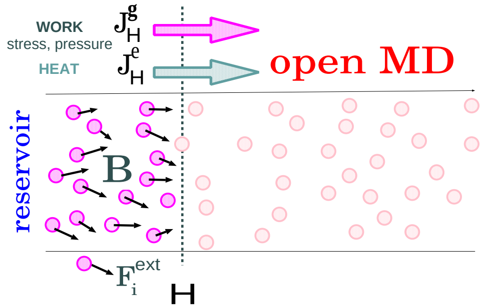


Coworkers

- *MD-CG-continuum.*
  - **Kurt Kremer**, Max-Plank Institute for Polymer Research (Mainz, Germany).
  - **Matej Praprotnik**, Max-Plank Institute for Polymer Research.
- *MD-continuum hydrodynamics*
  - **Gianni De Fabritiis**, U. Pompeu Fabra (Barcelona)
  - **Peter Coveney**, UCL (London)
- *Open boundaries for Fluctuating hydrodynamics*
  - **Anne Dejoan**, CIEMAT (Madrid)
- *Coarse-graining with proper dynamics.*
  - **Pep Español**, UNED (Madrid).

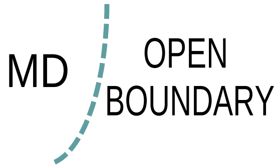
Outline of the talk

A Imposing fluxes in open MD

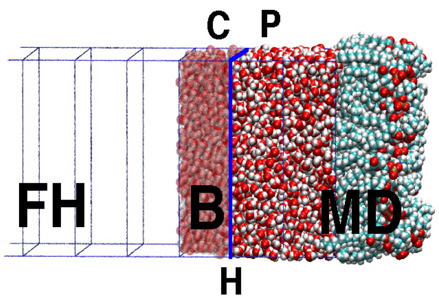


Flux boundary conditions for particle simulations

E. Flekkoy, RDB, P. Coveney, PRE (2005)



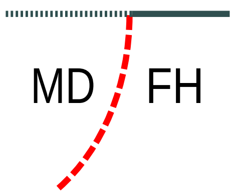
B Particle-continuum coupling: HybridMD



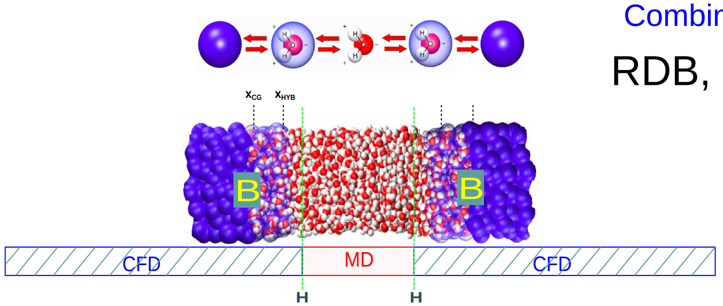
Molecular dynamics - fluctuating hydrodynamics

G. De Fabritiis, RDB, P. Coveney, PRL (2006)

RDB, G. De Fabritiis, PRE (2007)

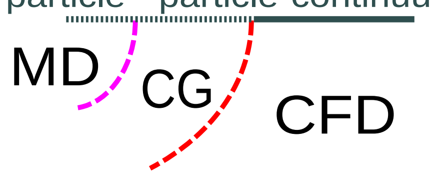


C Triple scale model: AdResS-HybridMD



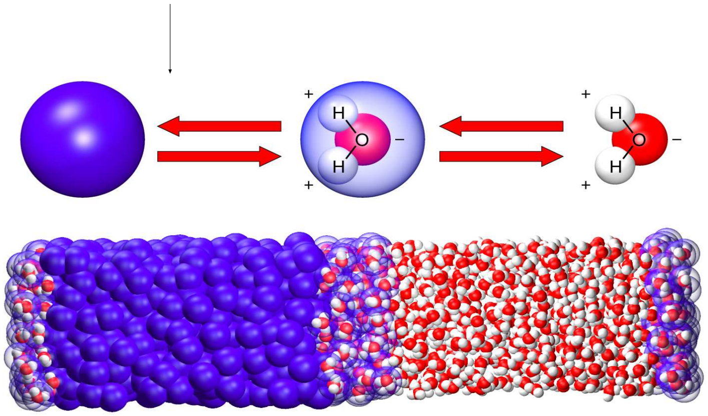
Combining Adaptive Resolution and Hybrid MD

RDB, M. Praprotnik, K. Kremer JCP (2008)



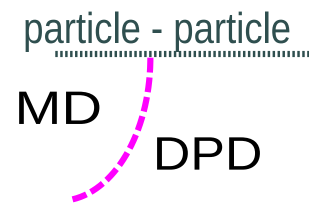
Outline of the talk (cont.)

D **Adaptive coarse Graining: AdResS** **(previous talk)**

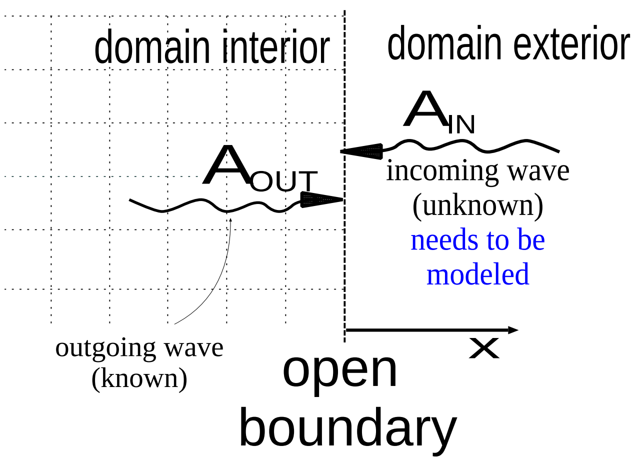


Changing the degrees of freedom, "on the fly"

M. Praprotnik, L. Delle Site, K. Kremer, JCP (2005)

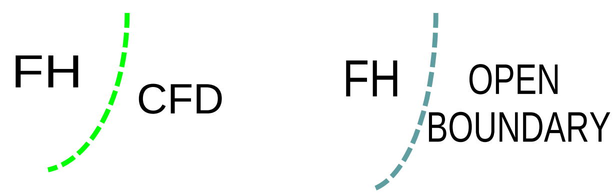


E **Open Fluctuating Hydrodynamics** **(probably not today)**



Non-reflecting boundary conditions for Fluctuating Hydrodynamics

RDB, A. Dejoan, PRE , **78**, 046708 (2008)



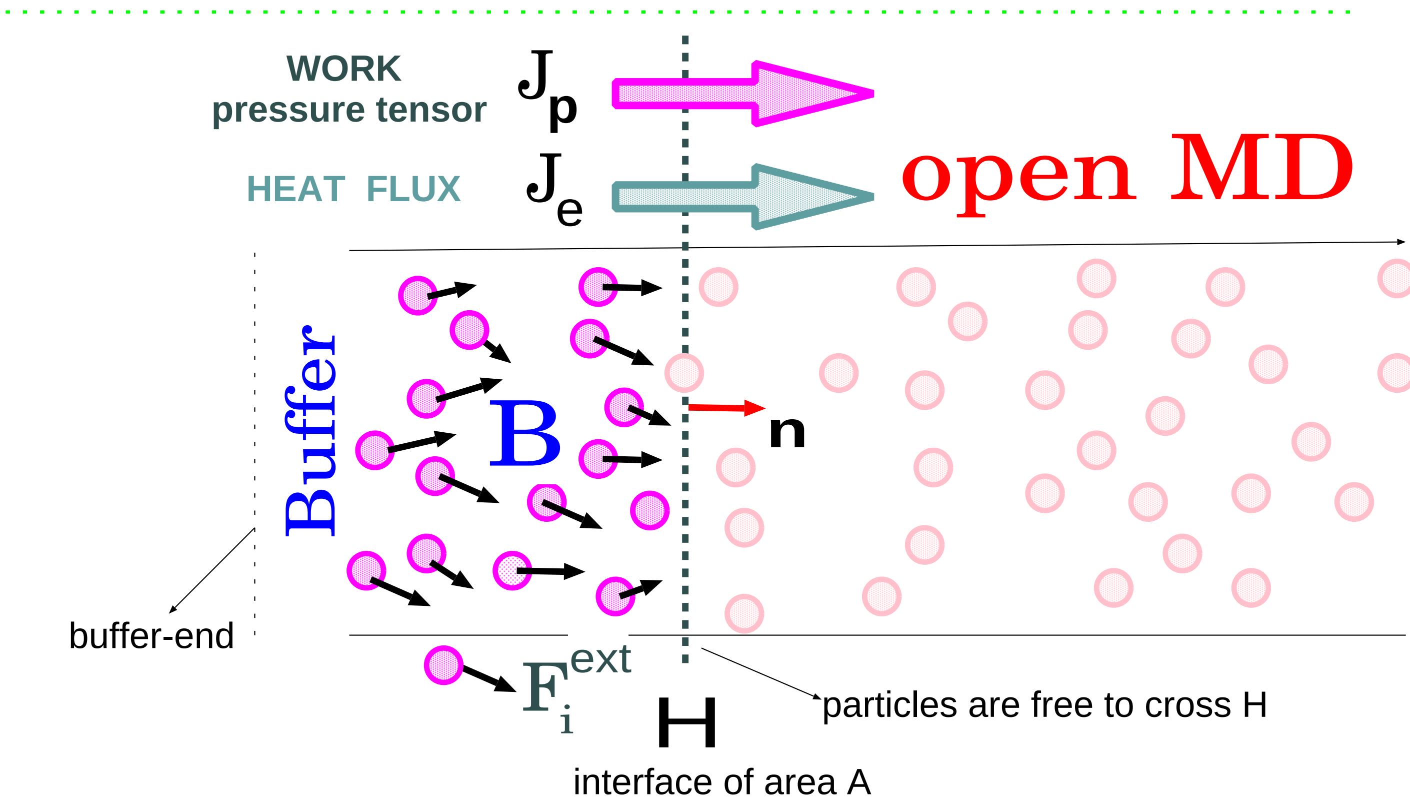
MD-CFD: Hybrid schemes depending upon the exchanged information

- **Coupling through variables:**
  - **Schwartz scheme**: steady state, closed system (only shear), no fluctuations.
  - **Constraint particle dynamics** (velocity imposition): unsteady, closed (only shear), no fluctuations.

- **Coupling through fluxes** (of momentum and energy)
  - **Unsteady** flows
  - **Open** molecular dynamics: **grand canonical ensemble**, generalized ensembles for MD.
  - Shear, **sound and heat** transfers (avoid finite size effect)
  - **Fluctuations** included (MD-Fluctuating hydrodynamics)

Open molecular dynamics

Flux boundary conditions for molecular dynamics



$$\mathbf{F}_i^{ext} = \frac{g_i A}{\sum_{i \in B} g_i} \mathbf{J_p} \cdot \mathbf{n} \simeq \frac{A}{N_B} \left(P\, \mathbf{n} + \mathbf{T} \cdot \mathbf{n} \right)$$

P pressure, **T** shear stress tensor.

Flux boundary conditions for MD

Flekkoy, RDB, Coveney, PRE **72**, 026703 (2005)

Energy flux J_e and momentum flux $\mathbf{J_p}$ imposed into MD across H

Momentum over Δt $\quad \mathbf{J_p} A\Delta t = \sum_{i\in B} \mathbf{F}_i^{ext}\Delta t + \sum_{i'} \Delta(m\mathbf{v}_{i'})$

Energy over Δt $\quad \underbrace{J_e A\Delta t}_{\text{Total input}} = \underbrace{\sum_{i\in B} \mathbf{F}_i^{ext}\cdot \mathbf{v}_i\Delta t}_{\text{External force}} + \underbrace{\sum_{i'} \Delta\epsilon_{i'}}_{\text{Particle insertion/removal}}$

External forces: $\mathbf{F}_i^{ext} = \langle \mathbf{F}_i^{ext}\rangle + \tilde{\mathbf{F}}_i^{ext}$ (particle $i \in B$)

Momentum: introduced by the mean external force $\langle \boldsymbol{F}_i \rangle$

$$\langle \mathbf{F}^{ext}\rangle = \frac{A}{N_B}\tilde{\mathbf{j}}_p \quad \text{where } \tilde{\mathbf{j}}_p \equiv \mathbf{J_p} - \frac{\sum_{i'}\Delta(m\mathbf{v}_{i'})}{A\,dt}\,.$$

Energy: introduced by the fluctuating force $\tilde{\mathbf{F}}_i^{ext}$ via dissipative work.

$$\tilde{\mathbf{F}}_i^{ext} = \frac{A\mathbf{v}_i'}{\sum_{i=1}^{N_B}\mathbf{v}_i'^2}\left[\tilde{j}_e - \tilde{\mathbf{j}}_p\cdot\langle\mathbf{v}\rangle\right] \qquad \text{with } \tilde{j}_e \equiv J_e - \frac{\sum_{i'}\Delta\epsilon_{i'}}{Adt}.$$

Open MD enables several ensembles

Flekkoy, RDB, Coveney, PRE, **72**, 026703 (2005)

Grand canonical	μ_BVT	Dynamics of confined systems
Isobaric ensemble	$\mathbf{J_p} = P\hat{\mathbf{n}}.$	
Constant enthalpy	$\mathbf{J}_e = M\langle\mathbf{v}\rangle \cdot F = -p\Delta V$ $\Delta N = 0$ $\Delta E + p\Delta V = \Delta H = 0$	Joule-Thompson, MD-calorimeter
Constant heat flux, Q	$\mathbf{J}_e = Q$	(**melting dynamics**, growth of solid phase -ice-, heat exchange at complex surfaces...)

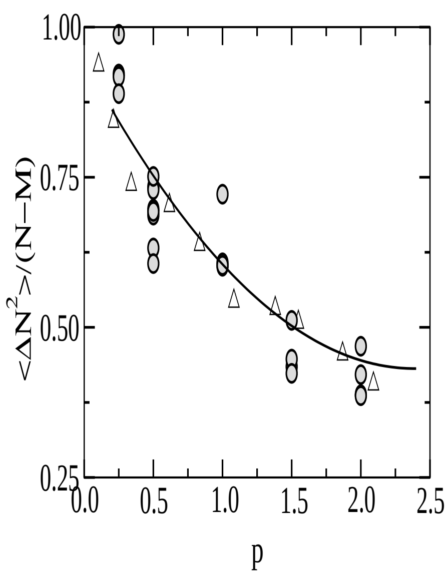
Mass fluctuations: grand canonical ensemble

$$\texttt{Var}[\rho] = k_B T \rho / (V c_T^2) \text{ with } c_T^2 = (\partial p / \partial \rho)_T$$

Flux particle BC´s are thermodynamically consistent

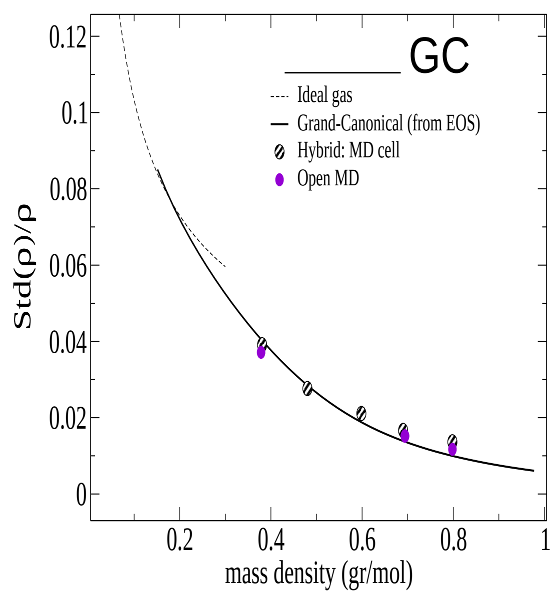
with the Grand Canonical ensemble

Soft-particles
DPD-like



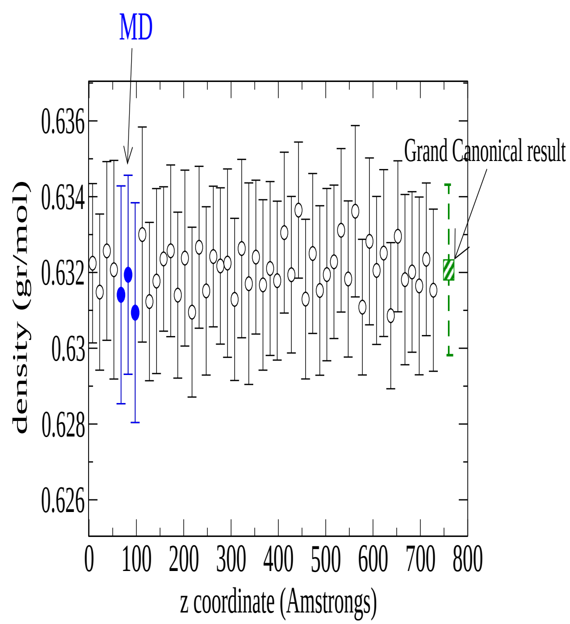
pressure

Liquid argon



density

Water (T=300 K)



cell-coordinate

The particle buffer

- **How to distribute the external force to the particles.**

$$\mathbf{F}_i^{ext} = \frac{g(x_i) A \mathbf{J}_p}{\sum_i g(x_i)}$$

(NB: to allow energy exchange one need $g(x_i) = 1$)

- **Control the average buffer mass to a fixed value** $\langle N_B \rangle$
  Use a simple relaxation algorithm:

$$\frac{\Delta N_B}{\Delta t} = \frac{1}{\tau_B} \left(\langle N_B \rangle - N_B \right)$$

with $\tau_B \simeq [10 - 100] fs$ (faster than any hydrodynamic time).

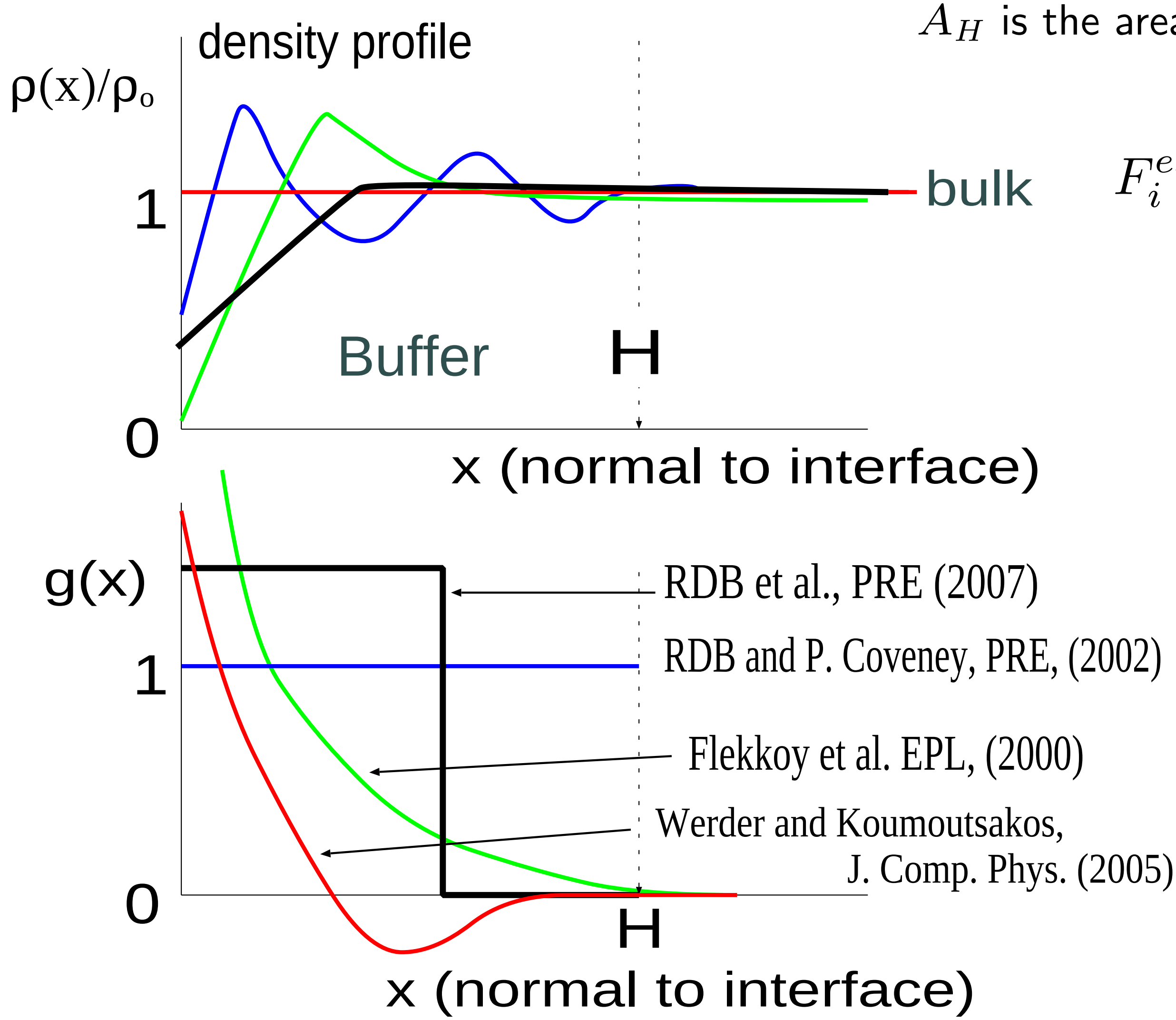
- **Open system: Particle insertion**
  - `Delete particle:` $\Delta N_B < 0$ or if leaving the buffer-end.
  - `Insert particle:` $\Delta N_B > 0$ USHER algorithm [J. Chem. Phys, **119**, 978 (2003)]

Force distribution at the buffer

Momentum flux across H: $\mathbf{J}_H \cdot \mathbf{e}_H$

A_H is the area of the interfase H

$$F_i^{ext} = \frac{g(x_i)}{\sum_{i \in B} g(x_i)} A_H \mathbf{J}_H \cdot \mathbf{e}_H$$



USHER energy controlled molecule insertion

J. Chem. Phys **119**, 978 (2003); J. Chem. Phys. **121**, 12139 (2004) (water)

- **Objective**: Insert a new molecule at target potential energy E_T.

- **Method**: Newton-Raphson-like search in the potential energy landscape.
  Succesful insertion $|\Delta E/E_T| < 0.01$ where $\Delta E = E_T - E_i^{(n)}$

Translation of the centre of mass along force direction $\boldsymbol{F}$

$$\mathbf{r}_{cm}^{n+1} = \mathbf{r}_{cm}^{n} + \frac{\mathbf{F}_{cm}^{n}}{F_{cm}^{n}} \delta r$$

Rotation around the torque axis: (water)

$$\mathbf{r}_{cm,i}^{n+1} = \mathcal{R}_{\delta\theta}^{(n)} \mathbf{r}_{cm,i}^{n}$$

$\delta r = \min(\Delta E/F, \Delta R_{\max})$; $\Delta R_{\max} \simeq$ half distance of first peak of radial distribution

$\delta\theta = \min(\Delta E/\tau, \Delta\Theta_{\max})$ the maximum rotation allowed is $\Delta\Theta_{\max} \sim 45^o$

Thermodynamically controllable process: Local ENERGY, TEMPERATURE and PRESSURE and are kept at the proper equation of state values.

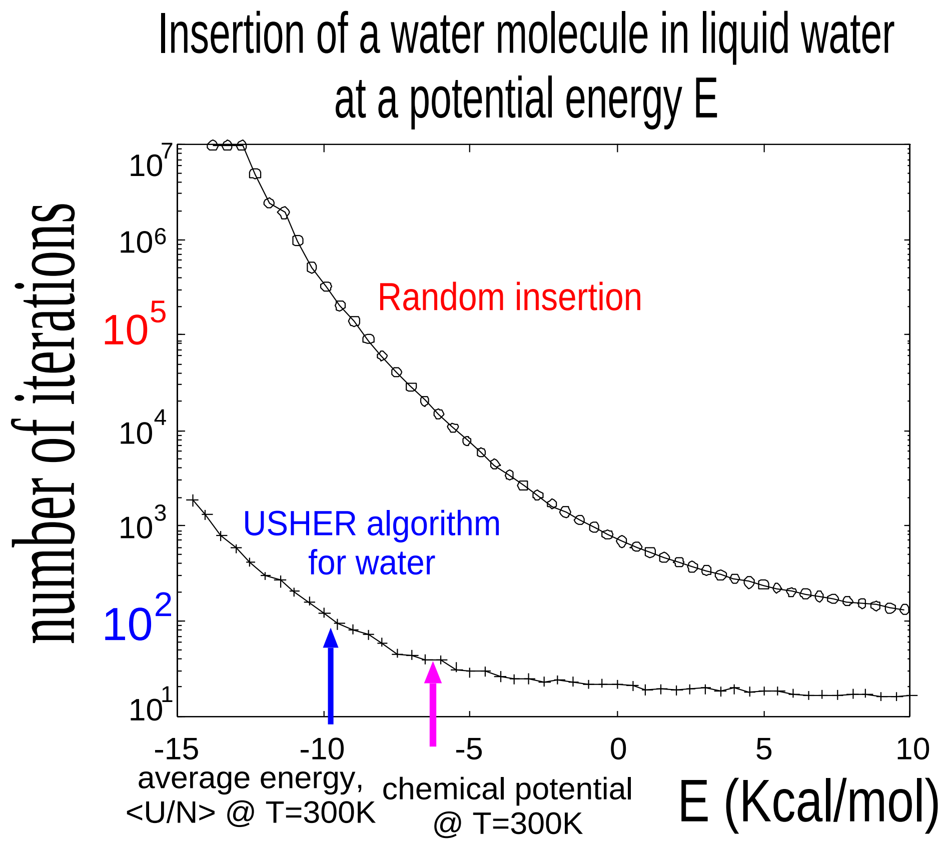
Negligible insertion cost:		
	LJ particles ($\rho < 0.85$)	$< 1\%$ total CPU
	Water into water	$\sim 3\%$ total CPU

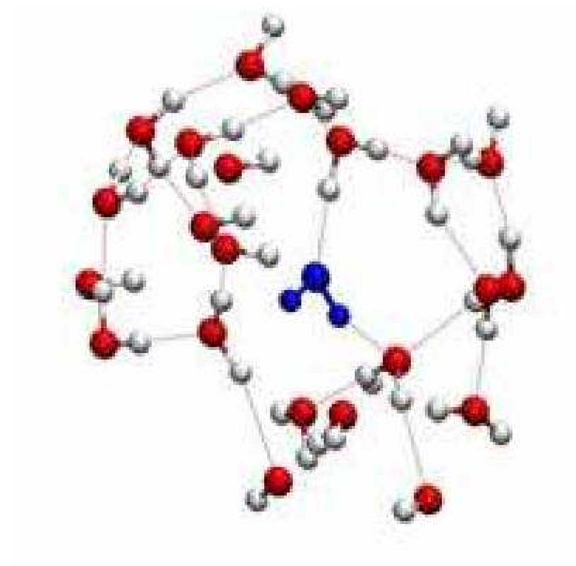
Insertions done at the mean energy/molecule contribution $E_T = 2U_{eos}$

USHER: fast and controlled particle insertion

J. Chem. Phys **119**, 978 (2003); J. Chem. Phys. **121**, 12139 (2004)
(water)

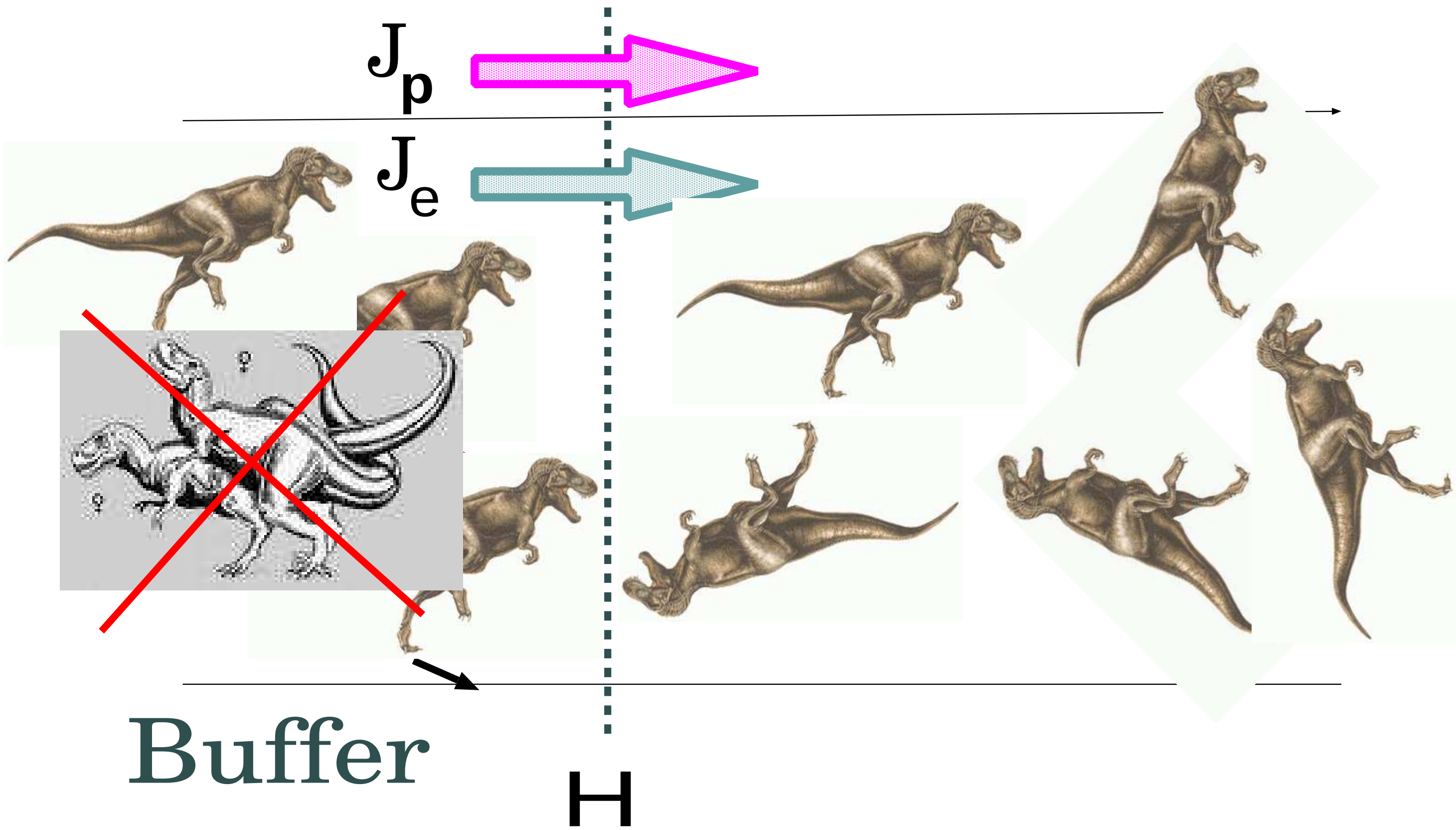
Applications: Constant chemical potential simulations, unfolding of proteins via water insertion (Goodfellow), water insertion in confined systems (e.g. proteins).





USHER has limitations

open MD for complex molecules



big particles cannot easily be inserted

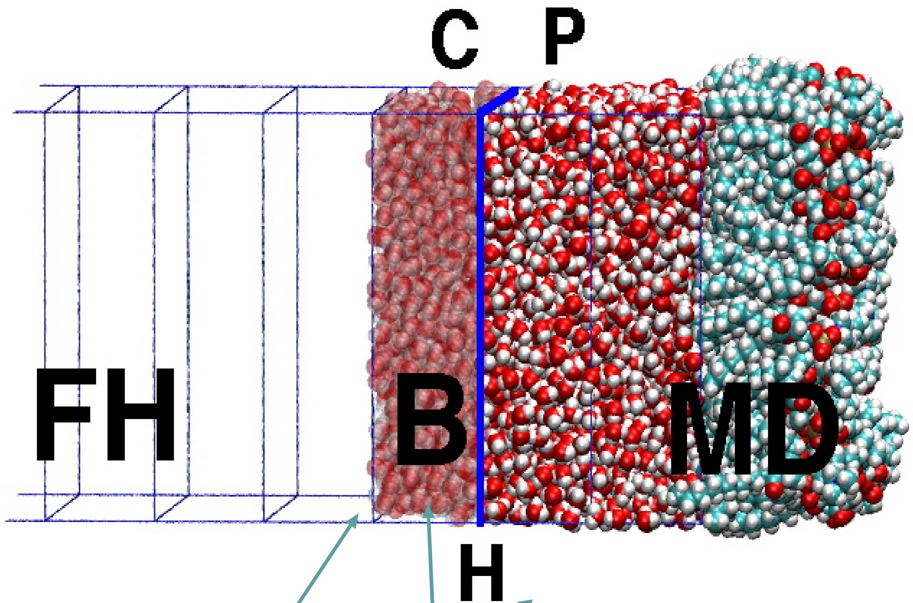
particle - continuum

MD FH

MD-FH: Domain decomposition

Coupling molecular dynamics (MD)
and fluctuating hydrodynamics (FH)

General issues concerning particle-continuum coupling



Information exchanged
variables / fluxes

Time dependence
Steady / Unsteady

Fluctuations
Deterministic CFD or
Fluctuating Hydrodynamics (**FH**)

End of particles box
open / closed

Particle buffer B
Maxwell daemons or
External forces

Continuity at interface H
- imposed to P / imposed to C
- continuity in fluctuations

Continuum fluid dynamics

- **Conservation law** conserved quantity per unit volume Φ

$$\partial\Phi/\partial t = -\mathbf{\nabla}\cdot\mathbf{J}^{\phi}$$

mass	$\Phi = \rho$	$\mathbf{J}^{\rho} = \rho\mathbf{u}$
momentum	$\Phi = \boldsymbol{g} \equiv \rho\mathbf{u}(\mathbf{r},\mathbf{t})$	$\mathbf{J}^{\boldsymbol{g}} = \rho\mathbf{u}\mathbf{u} + \boldsymbol{P}$
energy	ρe	$\mathbf{J}^{e} = \rho\mathbf{u}e + \boldsymbol{P} : \mathbf{u} + \boldsymbol{Q}$

- **Closure relations**

Equation of state $p = p(\rho)$

`Constitutive relations`

Pressure tensor $\boldsymbol{P} = p\mathbf{1} + \Pi + \tilde{\Pi}$

Viscous tensor $\Pi = -\eta\left(\nabla\boldsymbol{u} + \nabla\boldsymbol{u}^{T}\right) + (2\eta/3 - \xi)\,\nabla\cdot\boldsymbol{u}$

Conduction heat flux $\boldsymbol{Q} = -\kappa\nabla T + \tilde{Q}$

`Fluctuating heat and stress a la Landau`

Stress fluctuations $\langle\tilde{\Pi}(\boldsymbol{r}_1,t)\tilde{\Pi}(\boldsymbol{r}_2,0)\rangle = 2k_BTC_{\alpha\beta\gamma\delta}\delta(\boldsymbol{r}_2-\boldsymbol{r}_1)\delta(t)$

$C_{\alpha\beta\gamma\delta} = \left[\eta(\delta_{\alpha\delta}\delta_{\beta\gamma} + \delta_{\alpha\gamma}\delta_{\beta\delta} + (\zeta - \frac{2}{3}\eta)\delta_{\alpha\beta}\delta_{\delta\gamma}\right]$

Heat flux fluctuations $\tilde{Q}$

The finite volume scheme

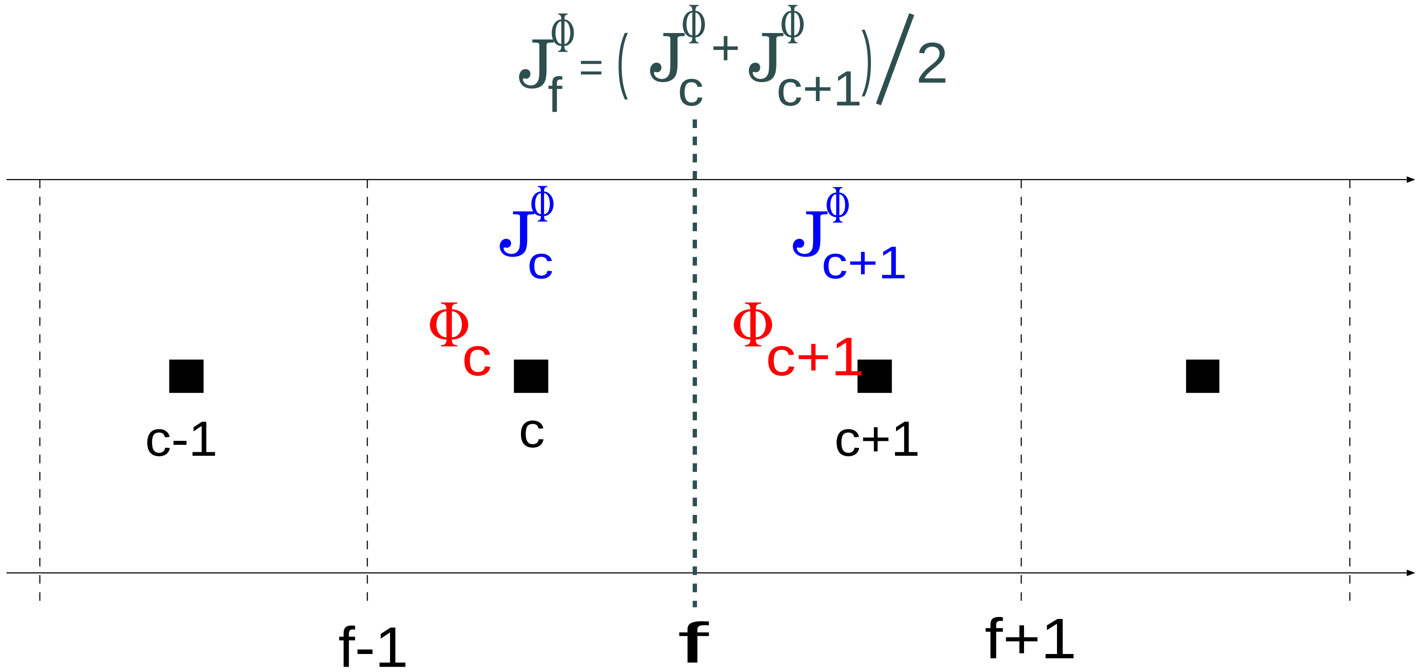
Finite volume schemes for fluctuating hydrodynamics

- FH for argon and water: G. De Fabritiis et al PRE, **75** 026307 (2007)
- Open BC for FH: RDB and A. Dejoan, PRE ,**78** 046708 (2008)
- Staggered grid for FH: RDB and A. Dejoan, (preprint)

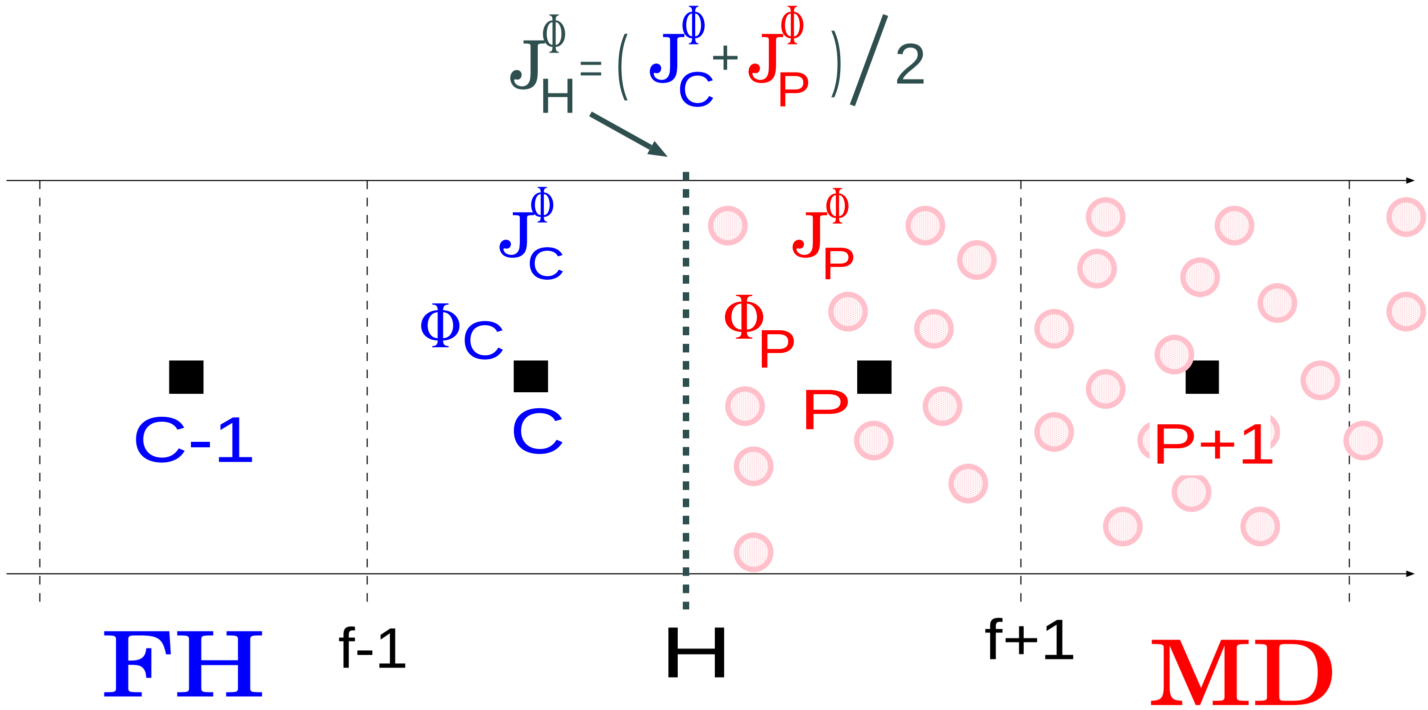
$$
\begin{aligned}
\int_{V_c} \partial\Phi/\partial t &= -\oint_{S_\alpha} \mathbf{J}^{\phi} \cdot \boldsymbol{ds} \\
V_c \frac{\Delta\Phi_c}{\Delta t} &= -\sum_{f=\text{faces}} A_f \mathbf{J}_f^{\phi} \cdot \boldsymbol{e}_f \quad \text{(explicit Euler scheme)}
\end{aligned}
$$

mass	$\Phi = \rho$	$\mathbf{J}^{\rho} = \rho\mathbf{u}$
momentum	$\Phi = \boldsymbol{g} \equiv \rho\mathbf{u}(\mathbf{r}, \mathbf{t})$	$\mathbf{J}^{\boldsymbol{g}} = \rho\mathbf{u}\mathbf{u} + \boldsymbol{P}$
energy	ρe	$\mathbf{J}^{e} = \rho\mathbf{u}e + \boldsymbol{P} : \mathbf{u} + \boldsymbol{Q}$

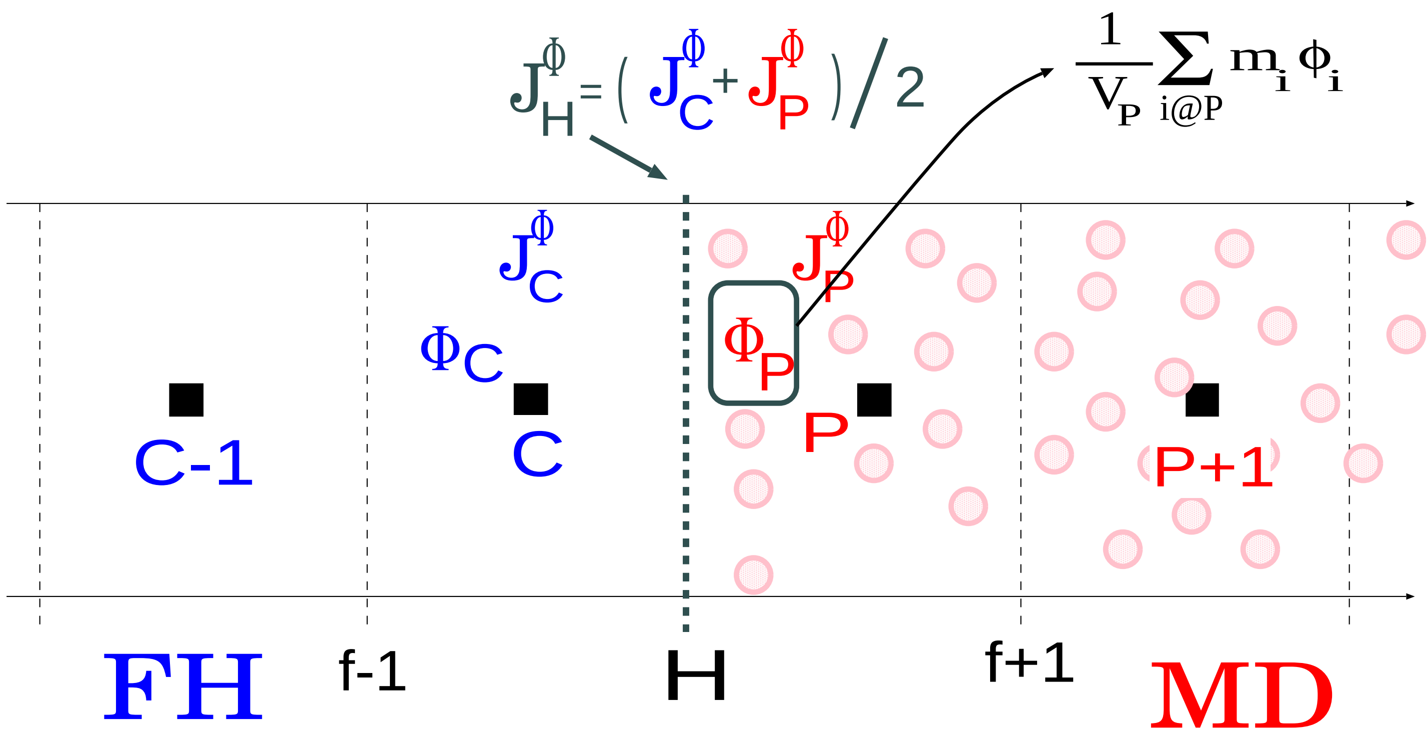
Finite volume scheme



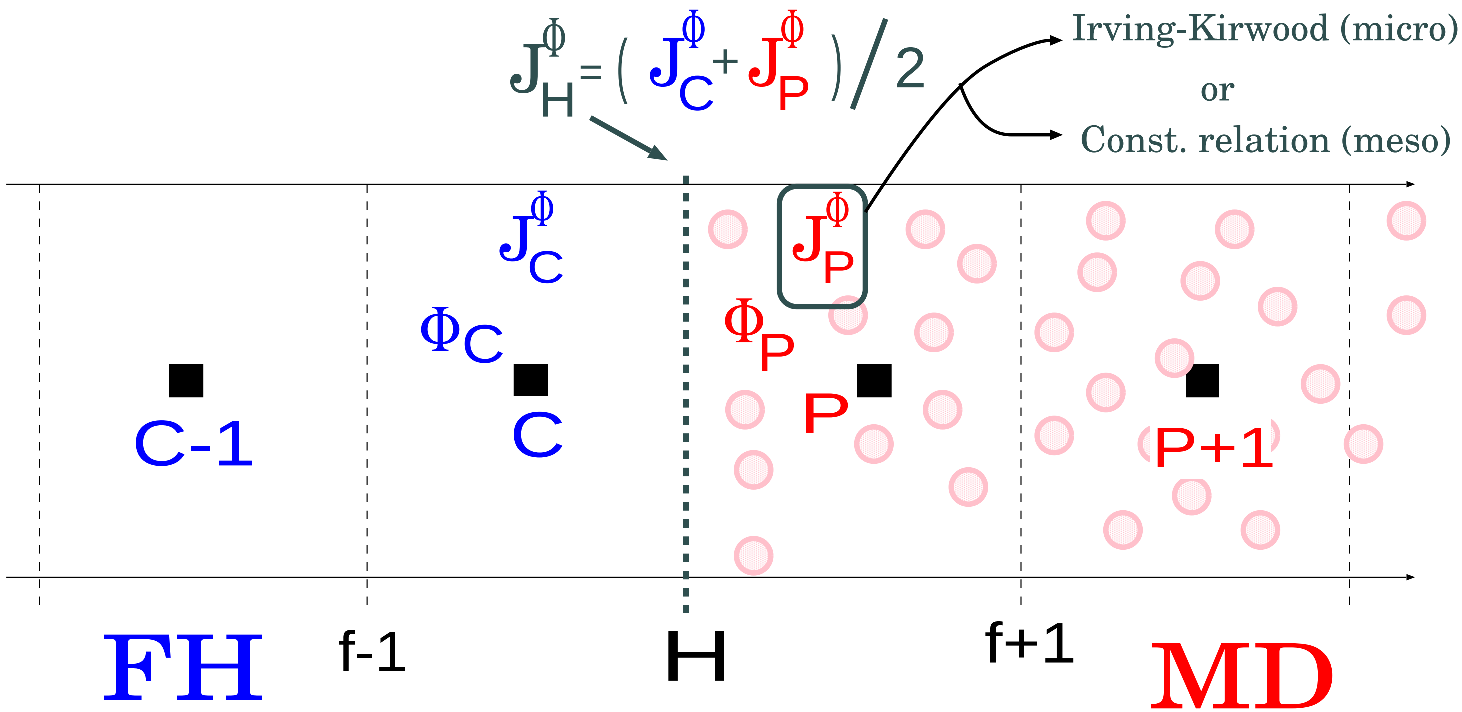
MD-FH: **hybridMD scheme**



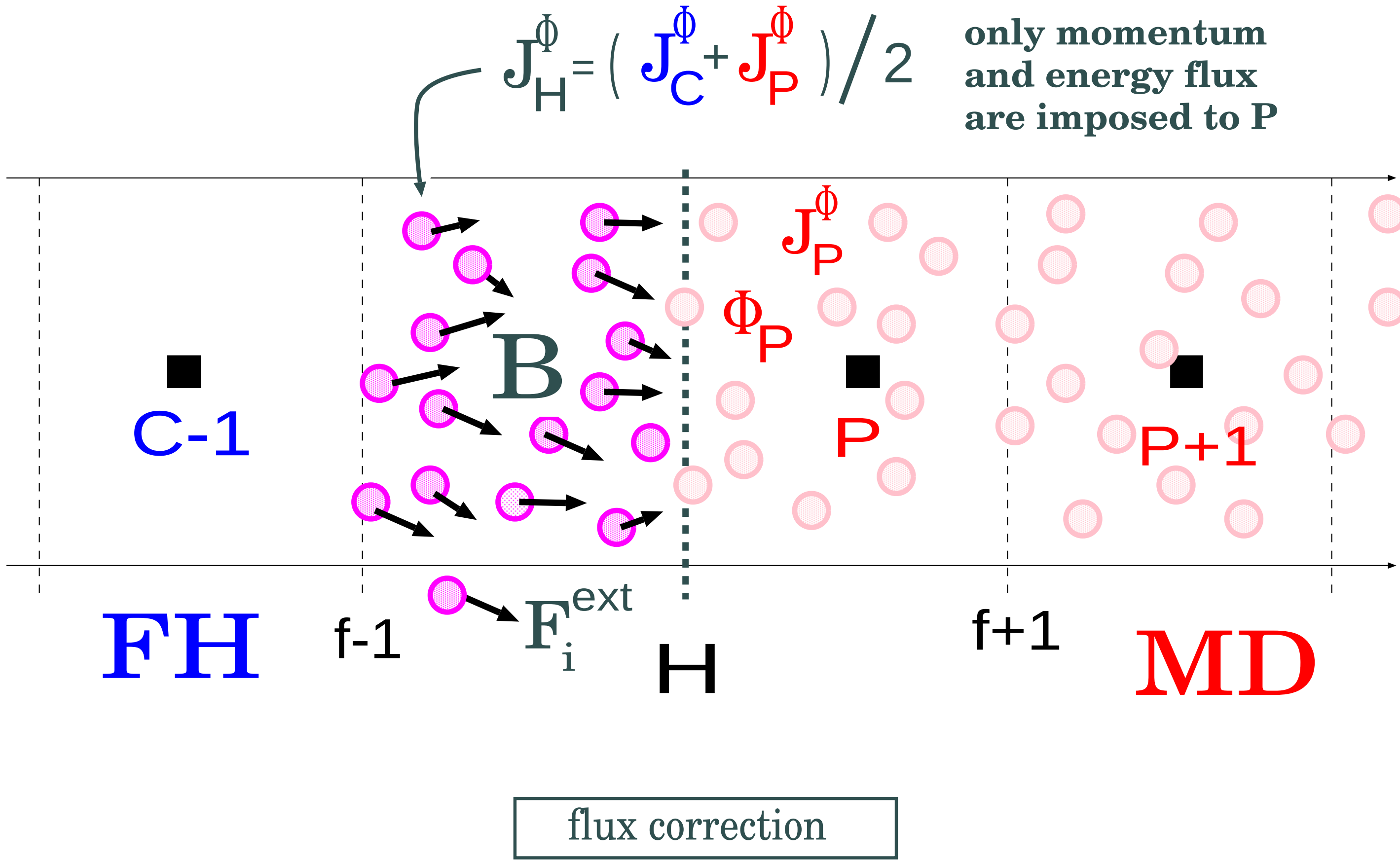
MD-FH: **Local P variables**



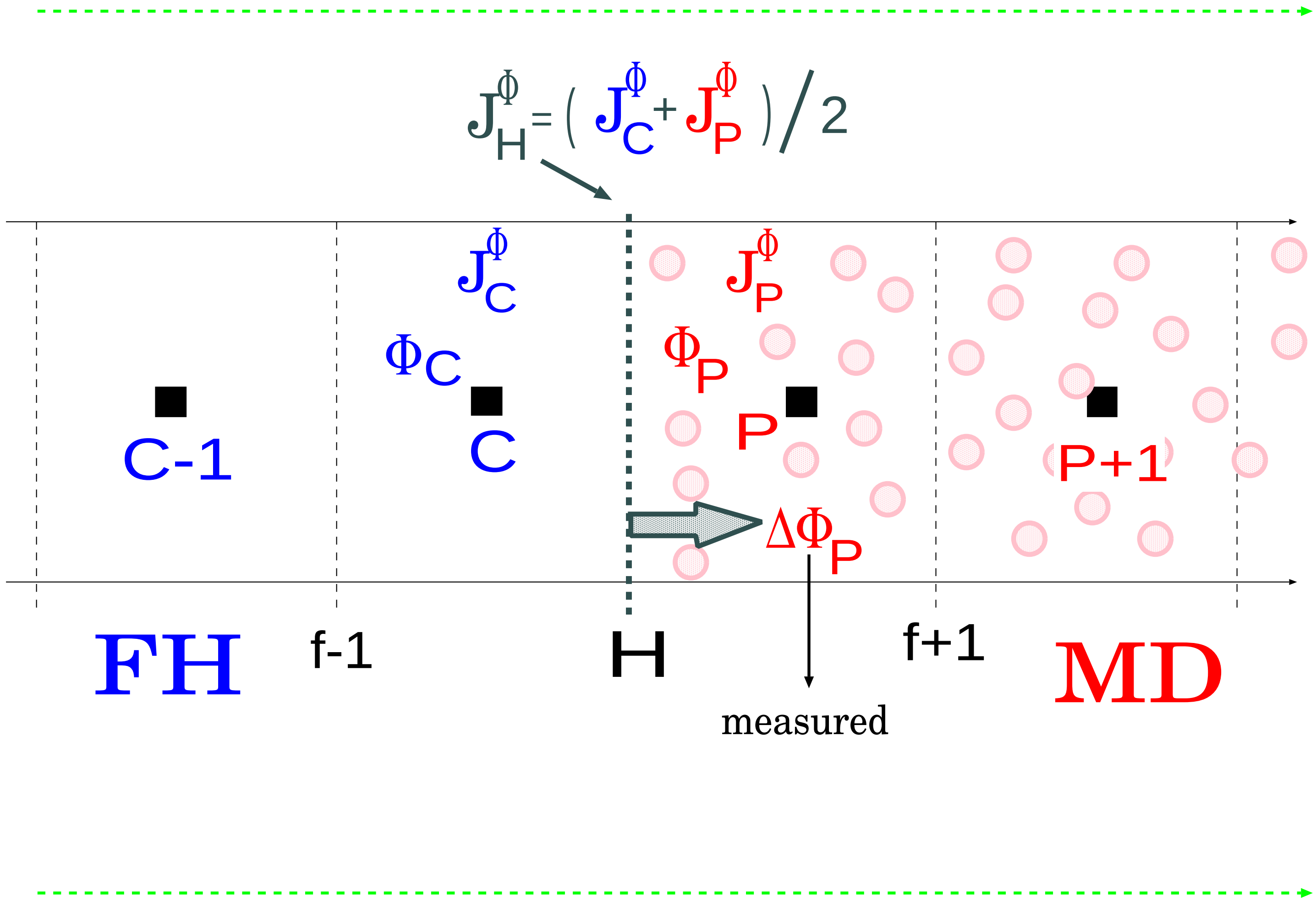
MD-FH: **Local P fluxes**



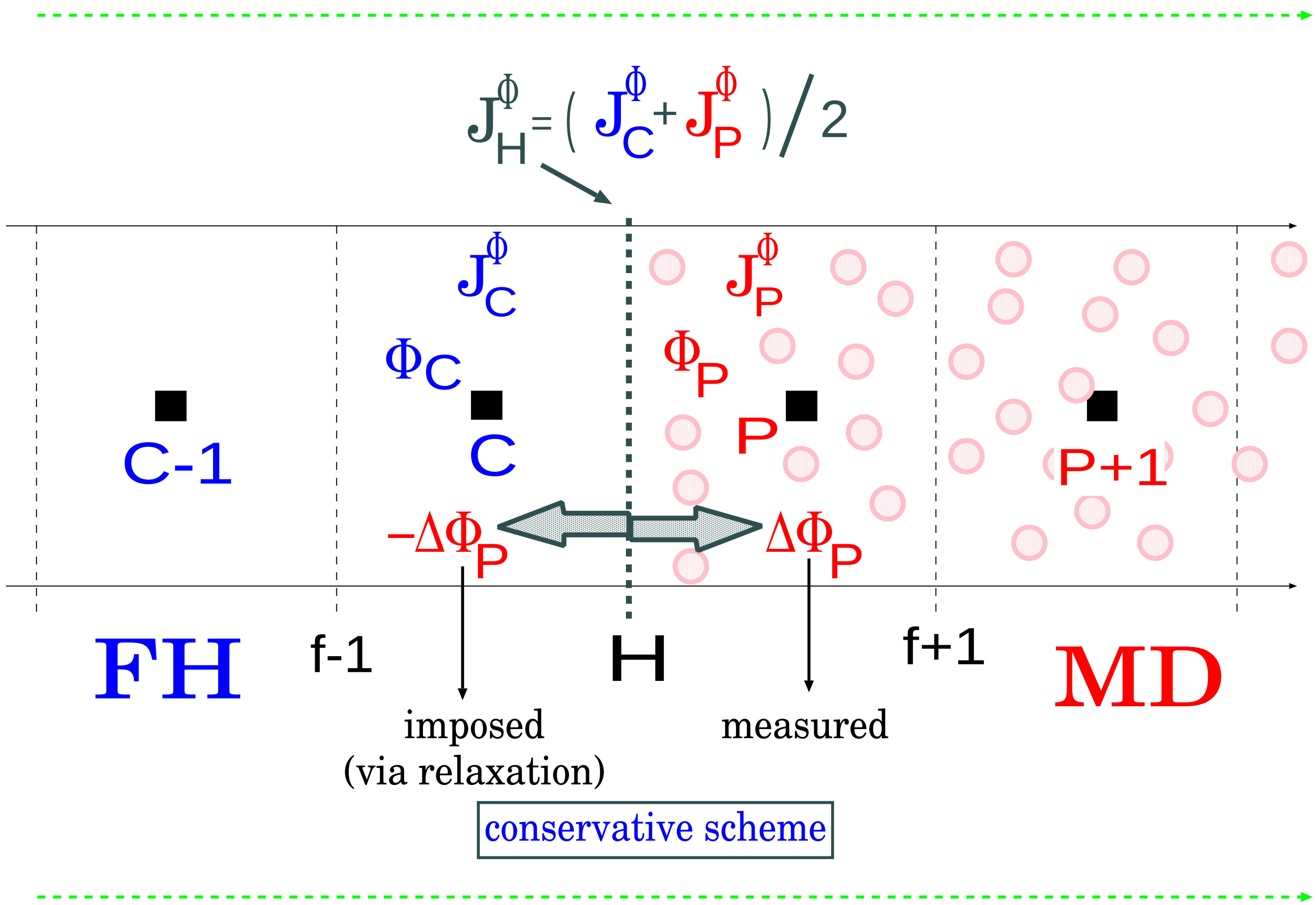
MD-FH: **Imposing fluxes into MD**



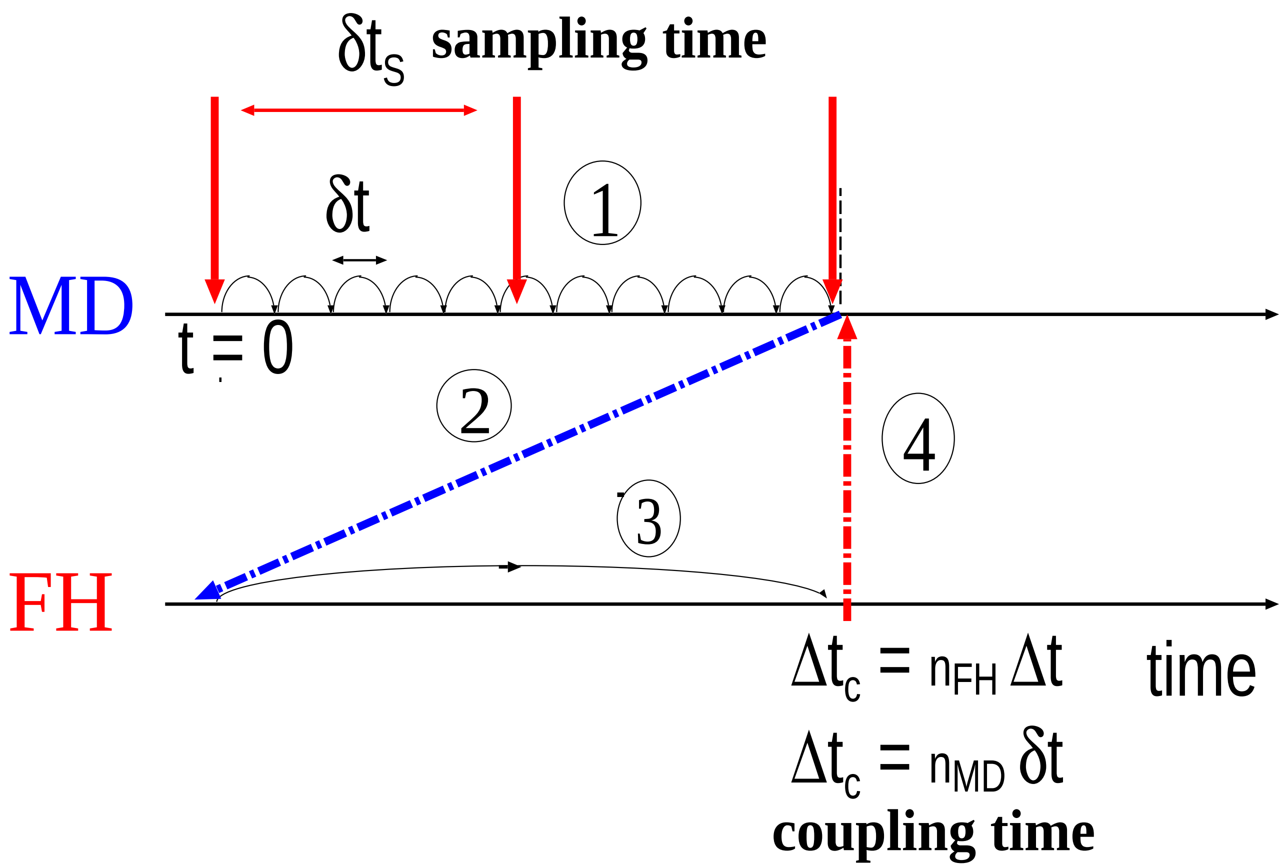
MD-FH: flux balance



MD-FH: **flux balance: conservative scheme**



MD-FH: Time coupling in flux based scheme



MD-FH: Coupling time and stress fluctuations

Green-Kubo relations

- Molecular dynamics: decorrelation time $\tau_c \sim 100$fs (simple liquids)

$$\langle J_{MD}^2 \rangle = \frac{\eta k_B T}{V \tau_c} \text{ with, } \tau_c \equiv \frac{\int_0^\infty \langle J(t) J(0) \rangle dt}{\langle J(0)^2 \rangle}$$

- Fluctuating hydrodynamics: decorrelation time $\Delta t_{FH}/2$,

$$\langle J_{FH}^2 \rangle = \frac{2 \eta k_B T}{V \Delta t_{FH}}$$

Thus, to balance the stress fluctuations, $\boxed{\langle J_{MD}^2 \rangle = \langle J_{FH}^2 \rangle}$:

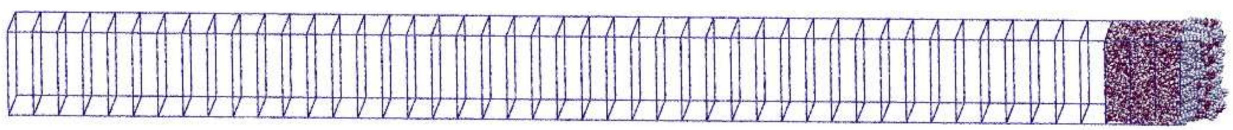
$$\Delta t_{FH} = 2\tau_c = \delta t_S \;\; \text{Sampling time} = \text{twice MD decorrelation time}$$

Coupling time, in general, $\boxed{\Delta t_c = n_{FH} \Delta t_{FH} = N_s \delta t_s}$

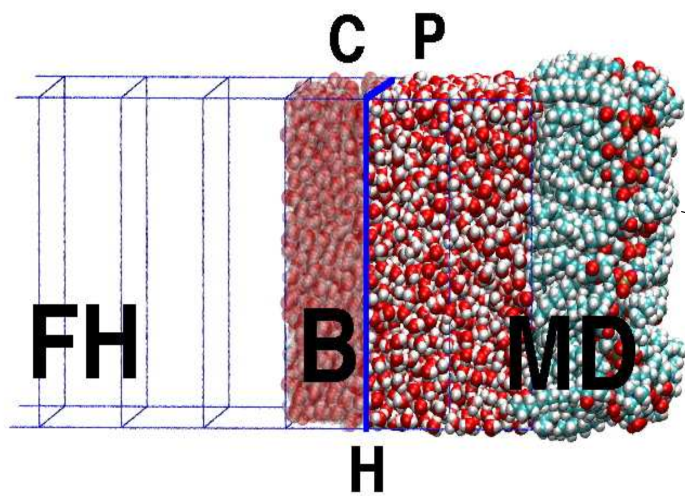
MD-FH Setup for tests

Water against a lipid layer at $T = 300K$
[G.Fabritiis,RDB, Coveney PRL, **97** (2006)].

Multiscale modelling
Embedding molecular dynamics within fluctuating hydrodynamics

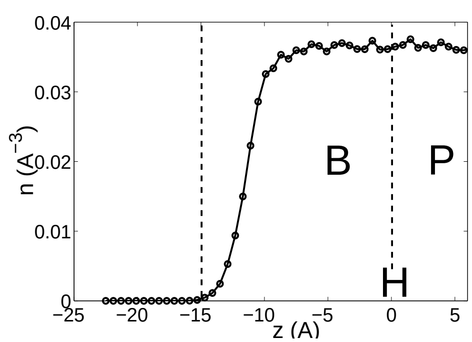


Hybrid MD-FH setup



DMPC (lipid layer)

water density profile



PRL, 97, 134501 (2006)

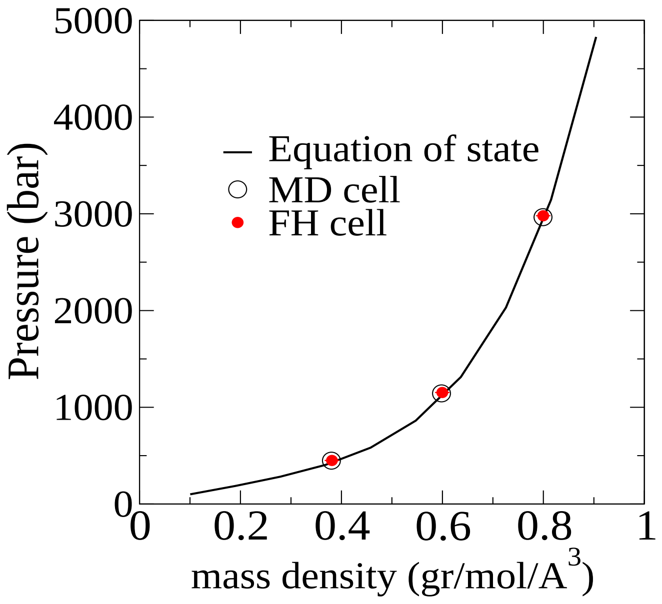
PRE, 76, 036709 (2007)

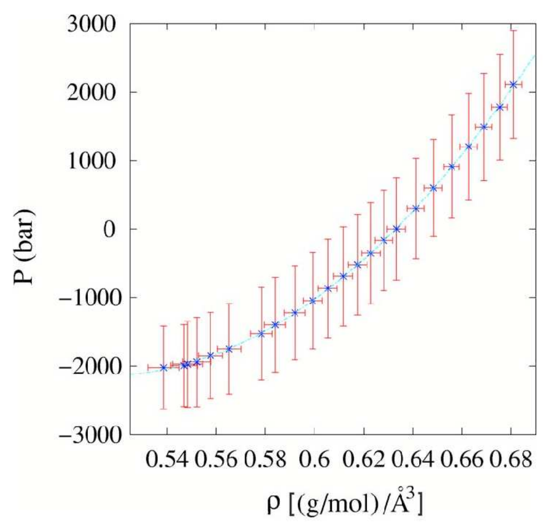
MD-FH Equilibrium

Equation of state $p = p(\rho)$ for argon and water TIP3P, $T = 300K$ [G.Fabritiis et al. PRE, **76** (2007)].

OPEN MD can be used to measure $p = p(\rho)$

argon (LJ) water (TIP3P)

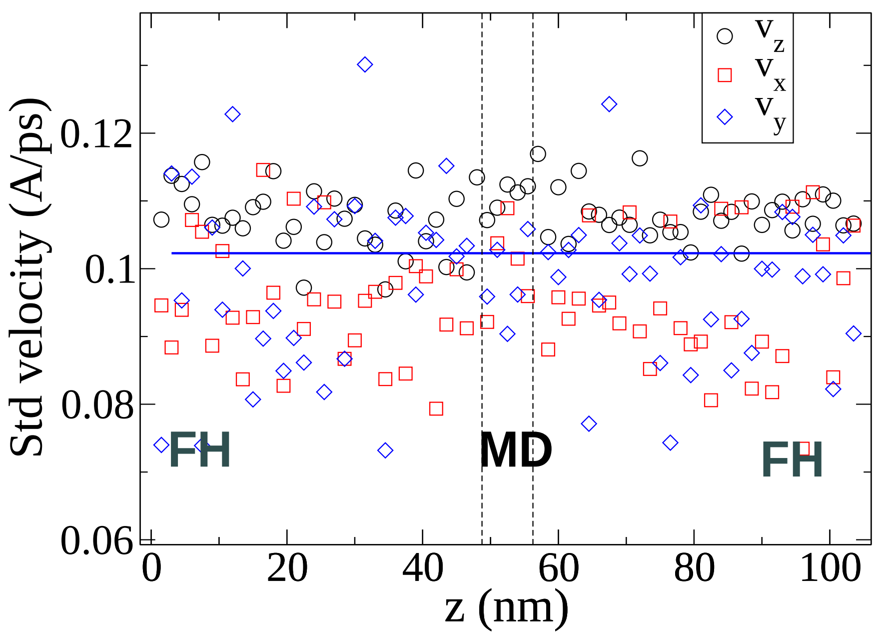




MD-FH Velocity and stress fluctuations

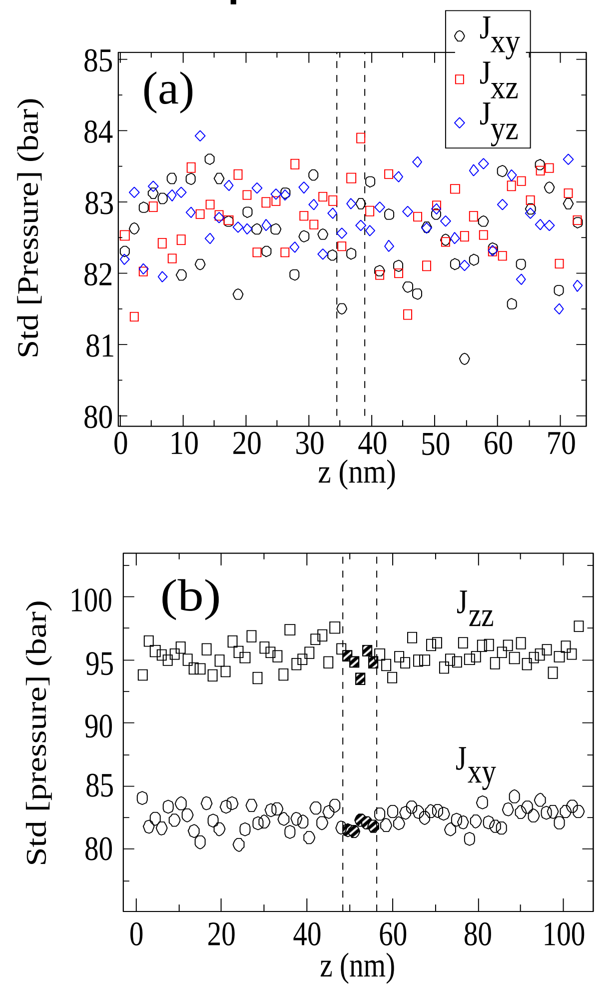
Standard deviation of velocity (kinetic temperature)
liquid argon @ $T = 300K$ [RDB and G.Fabritiis et al. PRE, **76** (2007)].

STD velocity



position

STD Stress tensor components



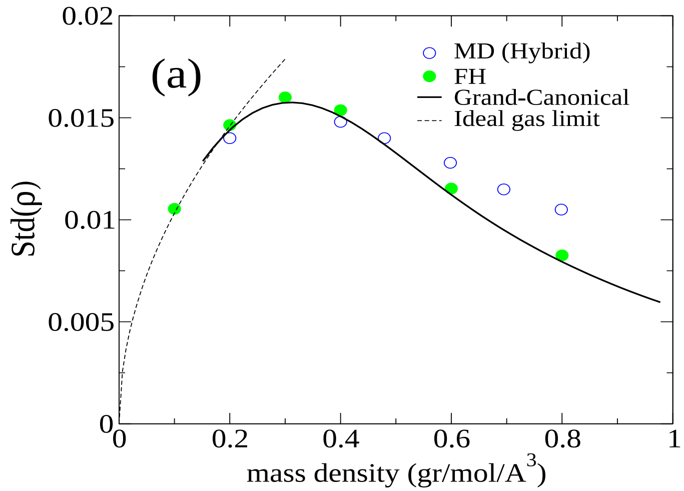
position

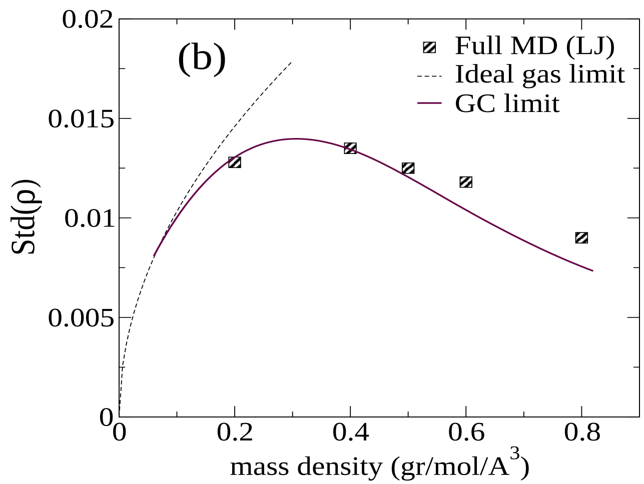
MD-FH Density fluctuations

Standard deviation of density

argon at several densities, $T = 300K$

RDB and G.Fabritiis et al. PRE, **76** (2007)





HybridMD

FH

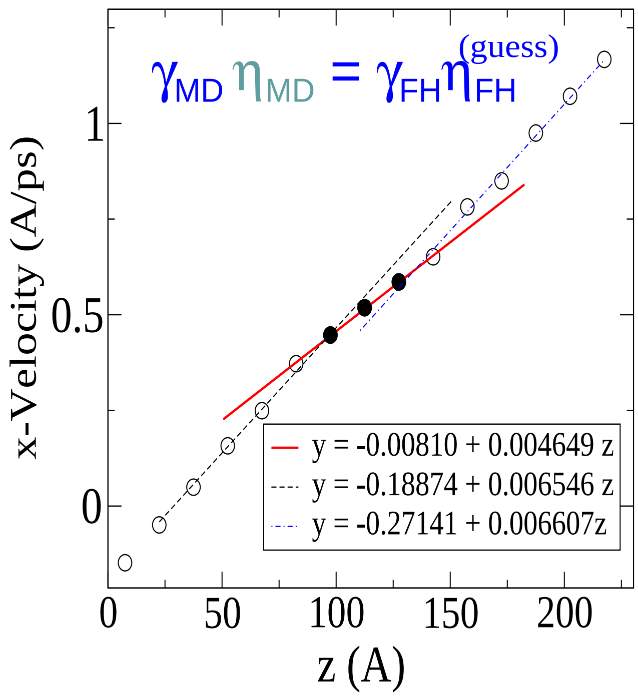
Grand canonical

full MD

MD-FH **Shear flow**

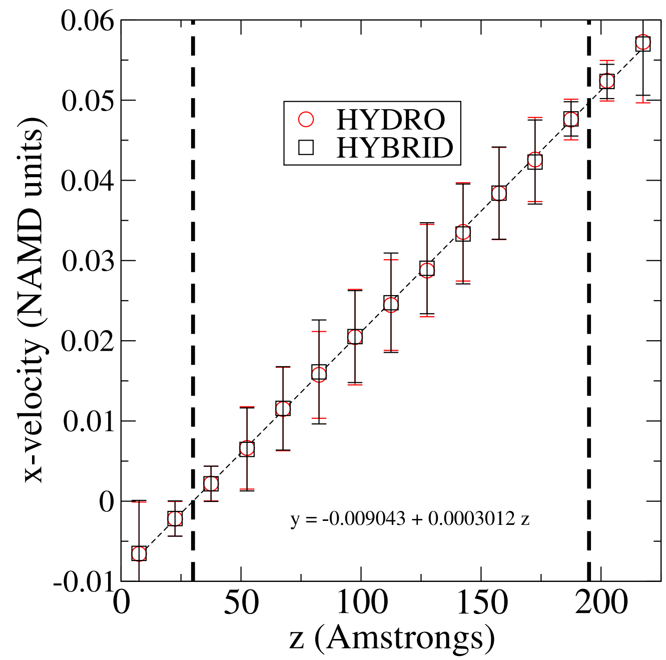
viscosity calibration

hybridMD as a rheometer



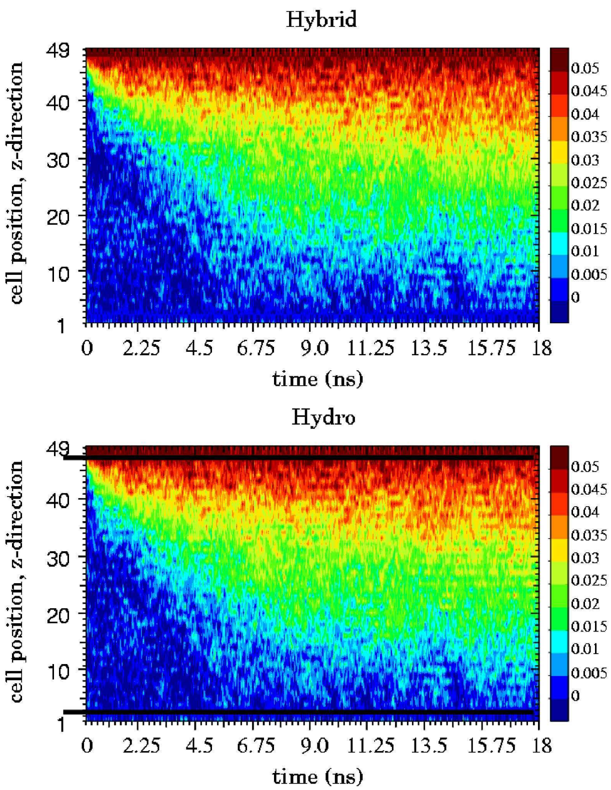
Couette flow

steady solution

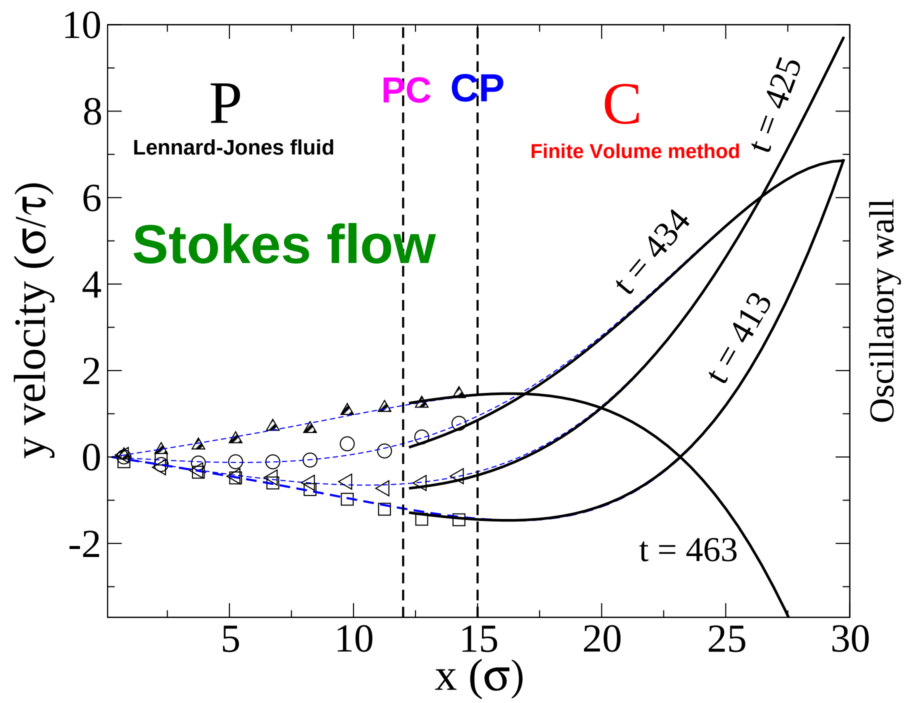


MD-FH **Unsteady shear**

Start-up Couette

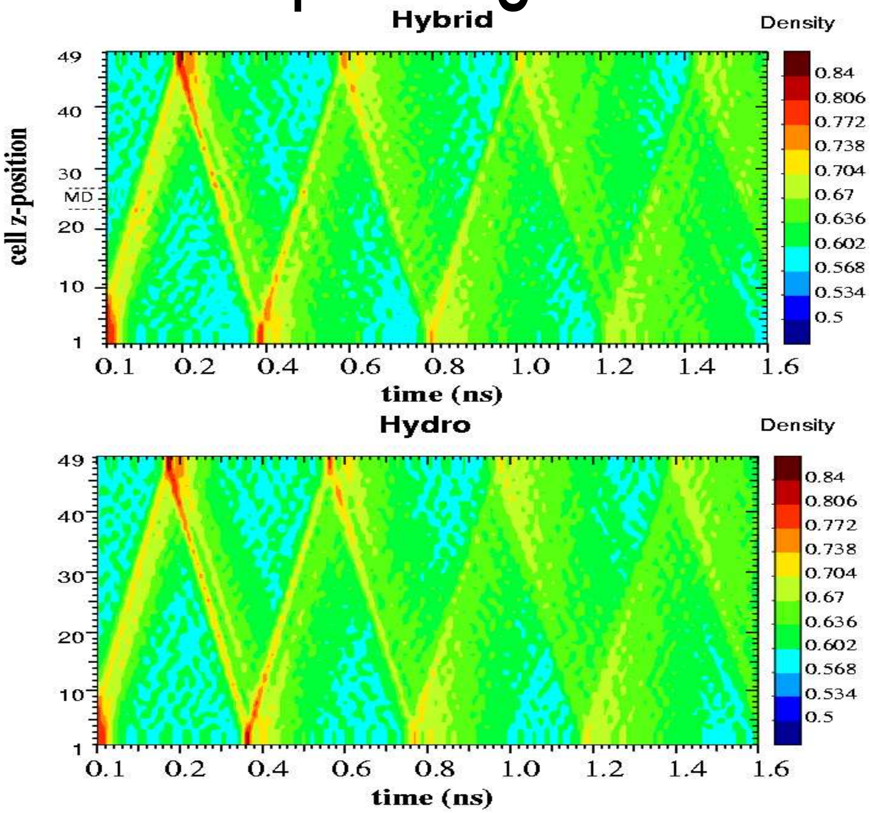


Oscillatory shear

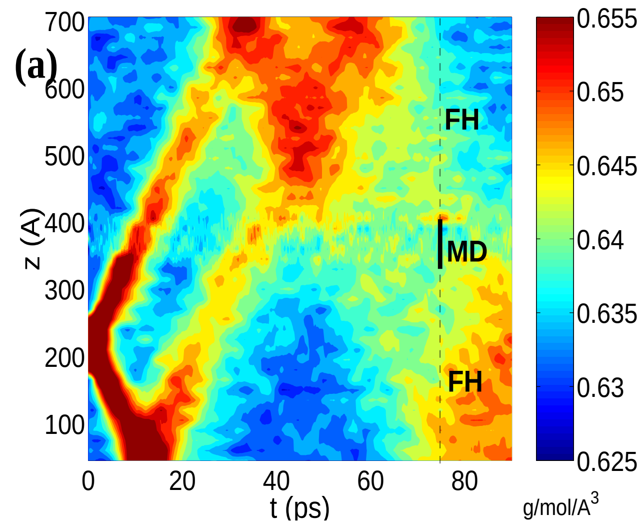


MD-FH **Sound waves**

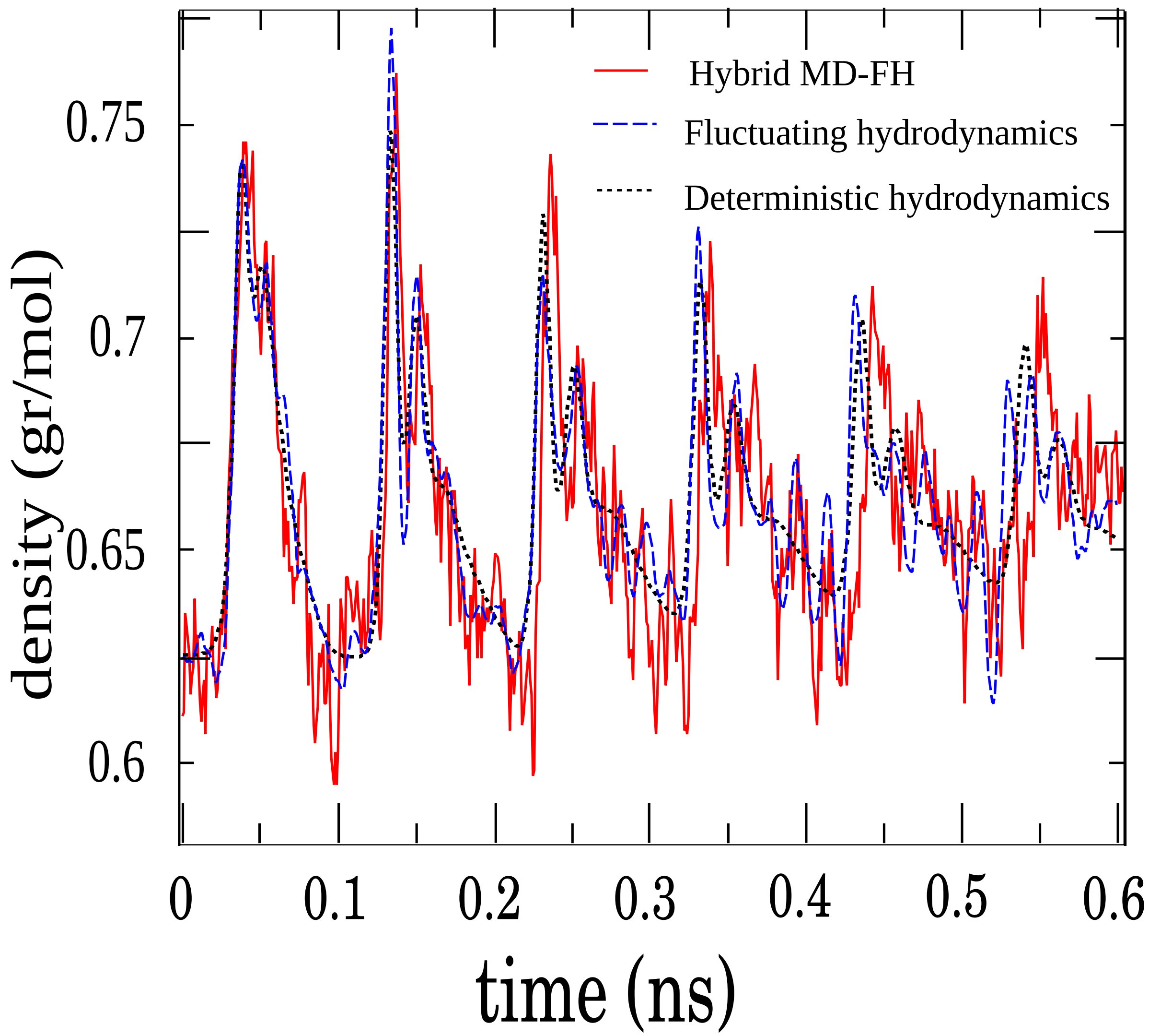
liquid argon



water

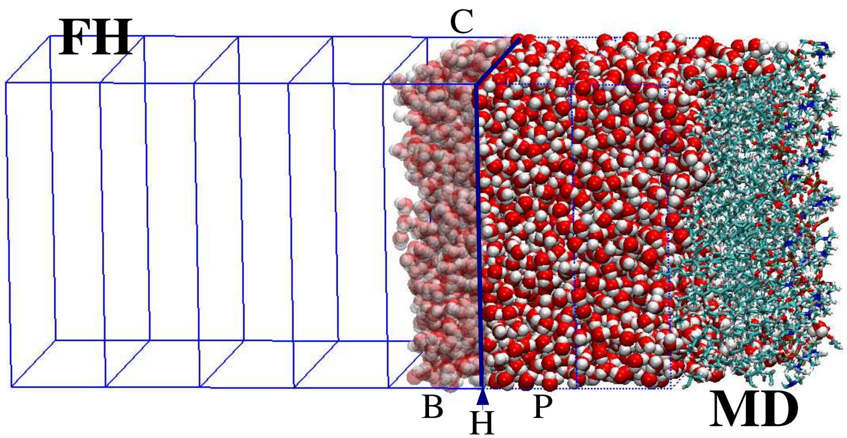


MD-FH **Sound waves: time resolution** ~ 0.02 **ns**

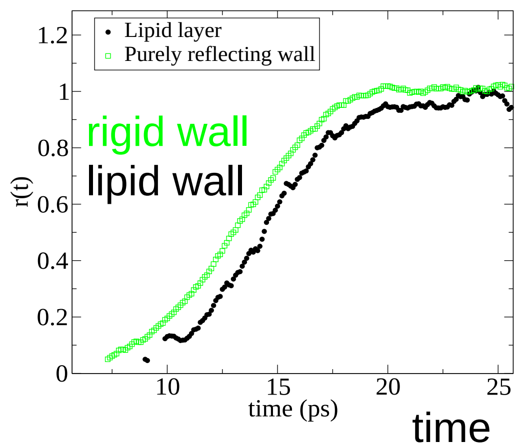


MD-FH **Sound - (soft) mattter interaction**

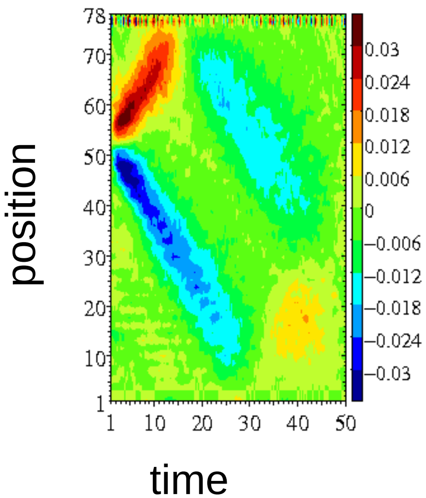
RDB et al, J. Mech. Engineering Sci. (2008)



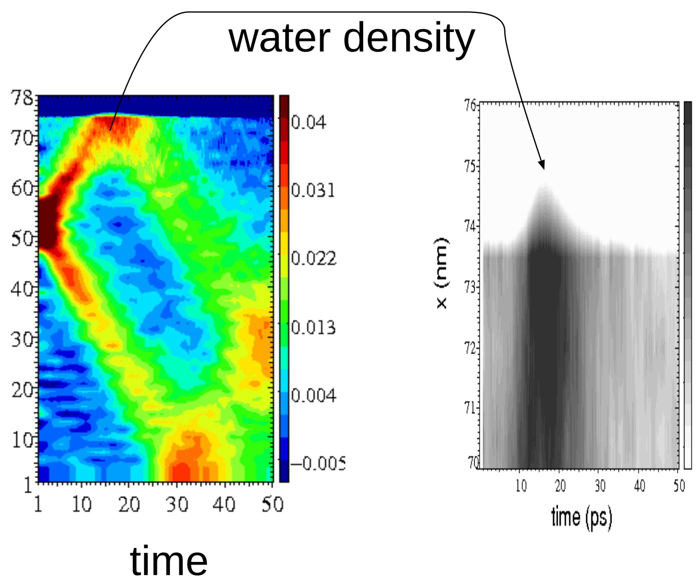
Reflection coefficient



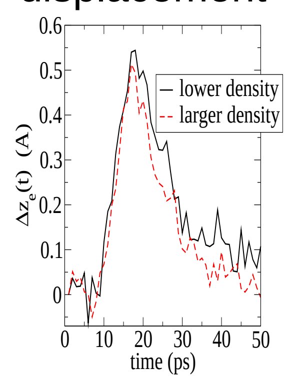
velocity

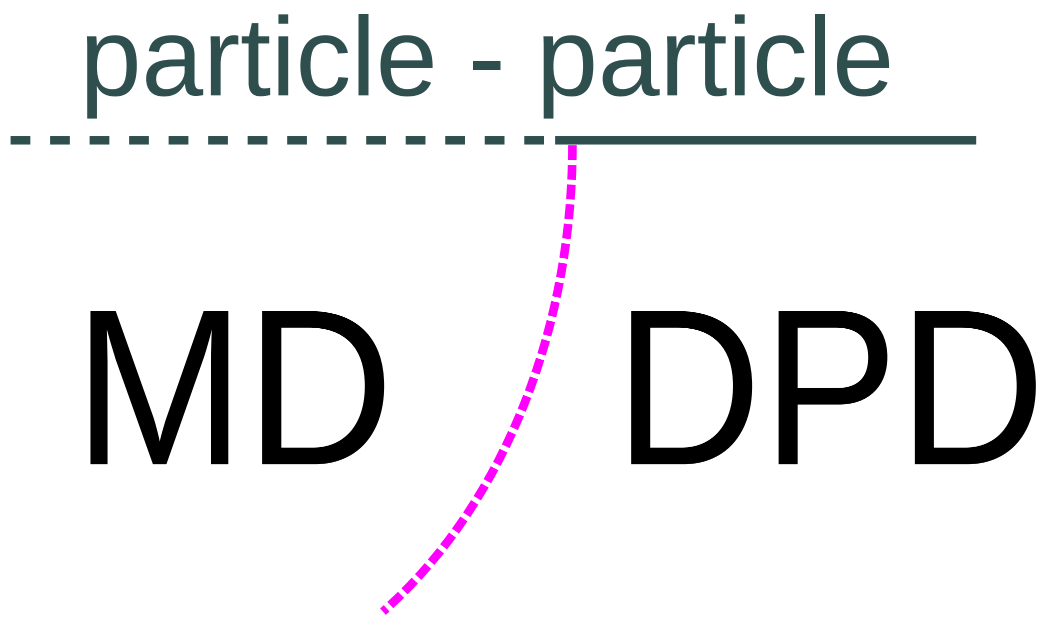


water density



lipids' tail displacement

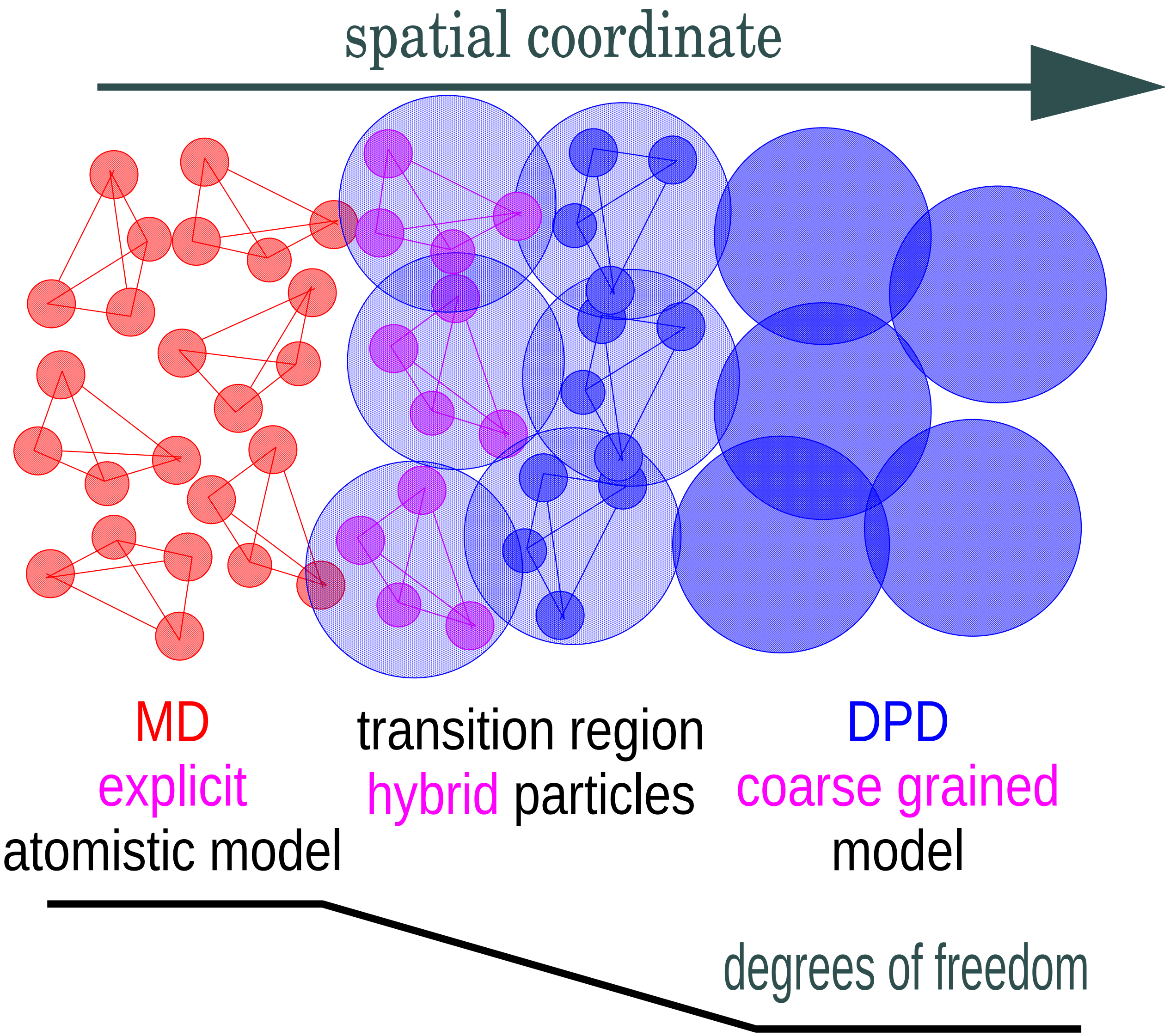



particle - particle
MD
DPD

Coupling MD to DPD

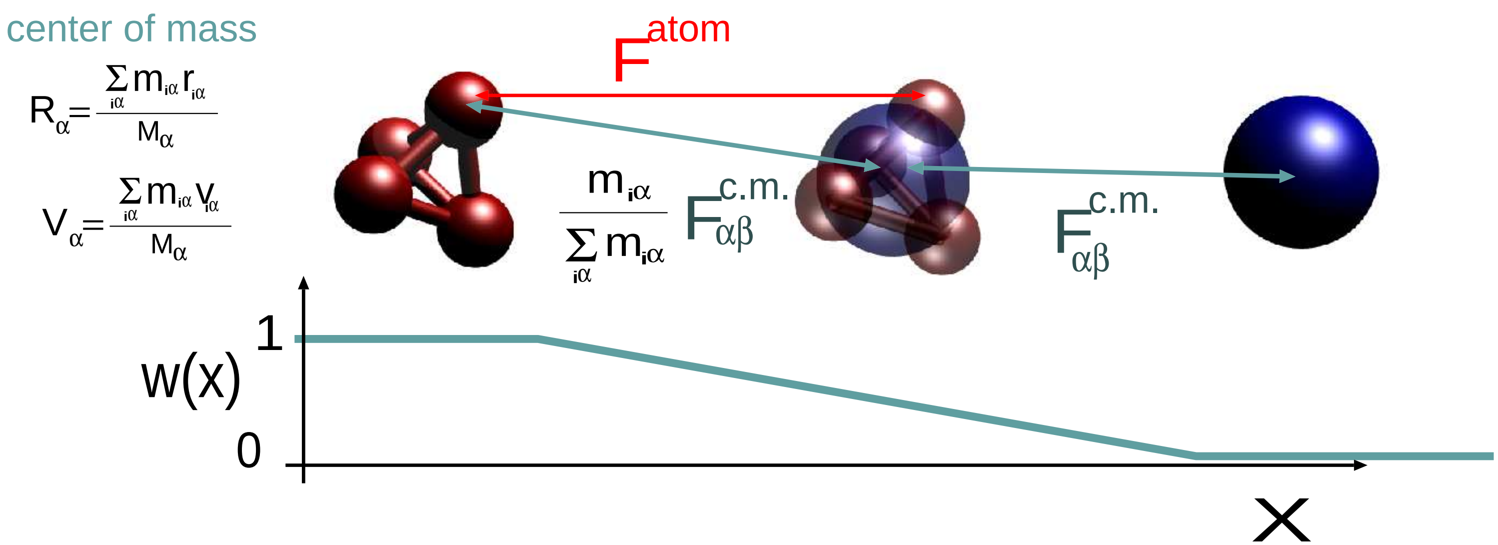
Adaptive Resolution Scheme (AdResS)

M. Praprotnik, L. DelleSite and K.Kremer, J. Chem.Phys **123** 224106 (2005), Ann. Rev. Phys. Chem. **59** 545 (2008)



Coupling MD to "DPD"

Adaptive Resolution Scheme



$$\mathbf{F}_{\alpha\beta} = w(x_\alpha)w(x_\beta)\sum_{i\alpha j\beta}\mathbf{F}^{\text{atom}}_{i\alpha j\beta} + [1 - w(x_\alpha)w(x_\beta)]\mathbf{F}^{\text{c.m.}}_{\alpha\beta}$$

$$\mathbf{F}^{\text{atom}}_{i\alpha j\beta} = -\frac{\partial U^{\text{atom}}}{\partial \mathbf{r}_{i\alpha j\beta}} \quad \textbf{Atomistic}$$

$$\mathbf{F}^{\text{c.m.}}_{\alpha\beta} = -\frac{\partial U^{\text{c.m.}}}{\partial \mathbf{R}_{\alpha\beta}} \quad \textbf{Coarse - Grained} \qquad (1)$$

Coupling MD to DPD

Effective potential for c.m. interaction

- The effective pair potential $U^{c.m.}$ is determined so as to match the center of mass radial distribution function of the *explicit* atomistic model, $g^{ex}{}_{cm}(r)$.

- This can be done using the `iterative Bolzmann inversion` [J. Comput. Chem. **24**1624 (2003)], which starts from the Potential of Mean Force as initial guess $(k = 0)$.

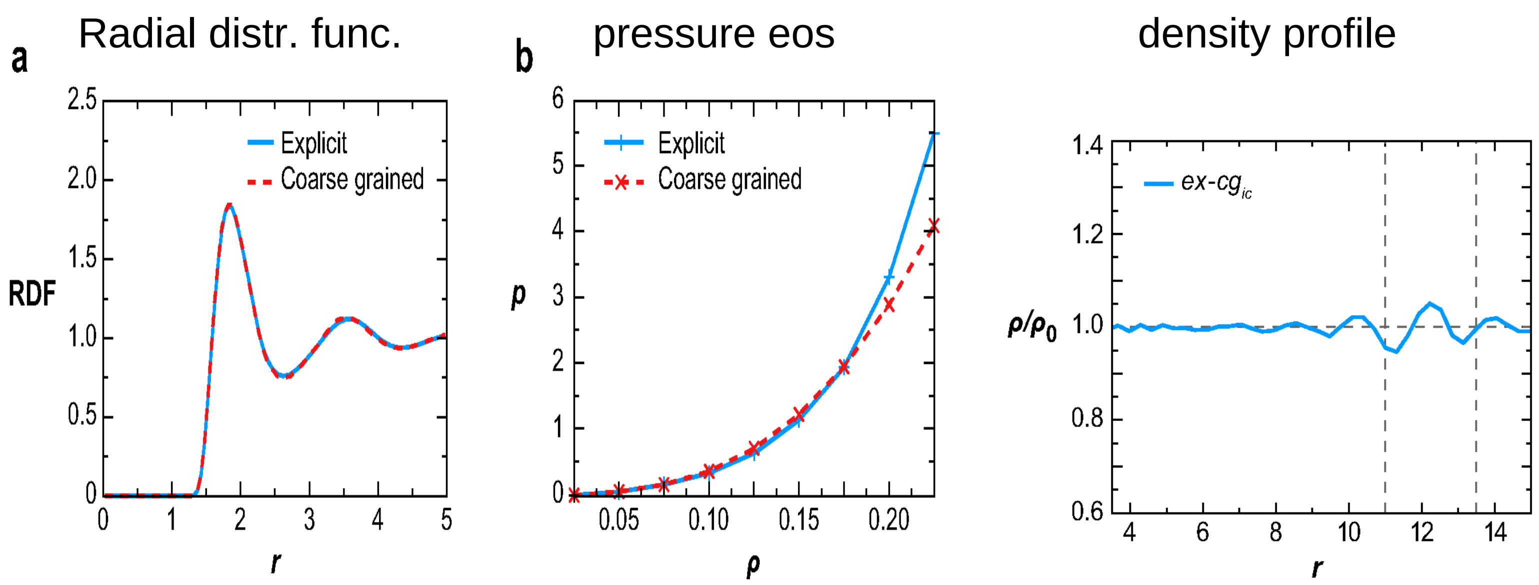
$$U_{k+1}^{cm}(r) = U_k^{cm}(r) + T \log \frac{g_k^{cg}(r)}{g^{ex}{}_{cm}(r)} \qquad (2)$$

- Small correction $\Delta U^{cm} = U_0(1 - r/r_c)$ to equilibribrate pressures.

Coupling MD and DPD

Matching liquid structure and pressure

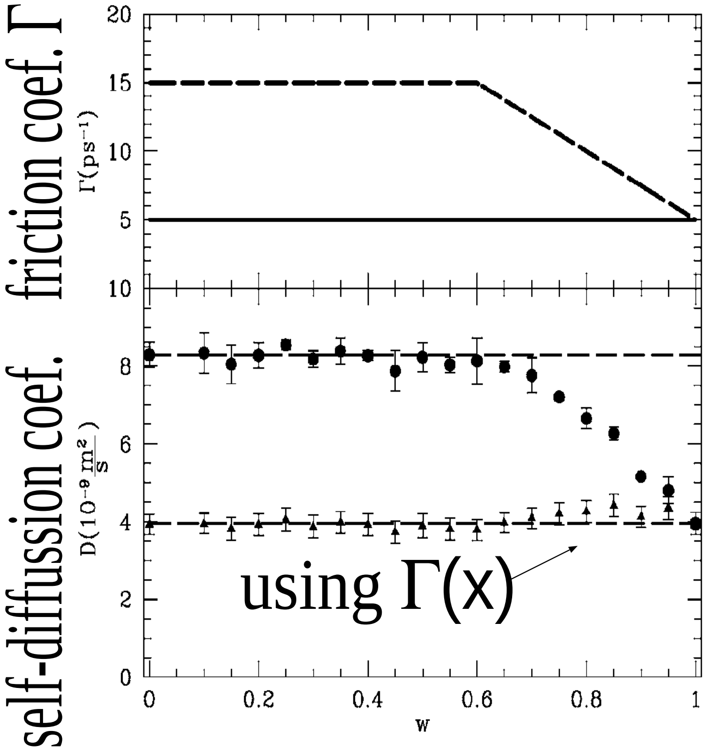
Tetraedral fluid
kT = 1; ρ=0.175



Coupling MD and DPD

Dynamics: self-diffusion across interfase

Position dependent Langevin thermostat



$$m_i \frac{dv_i}{dt} = F_i - m_i \Gamma(x_i) v_i + W_i(x_i, t)$$

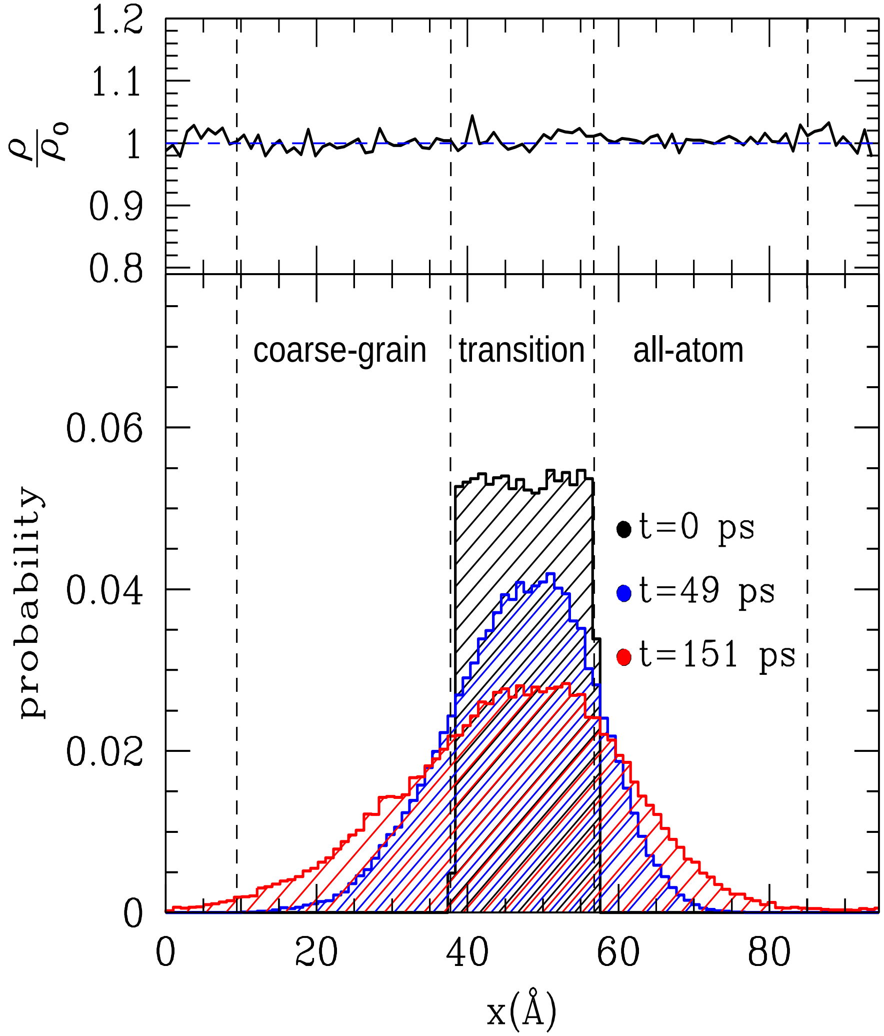
$$\langle W_i(x,0) \rangle = 0$$

$$\langle W_i(x,\tau) W_j(x,0) \rangle = 2\Gamma(x) kT \delta(\tau) \delta_{ij}$$

The thermostat at the "DPD" region is also needed to equilibrate the removed /added degrees of freedom (i.e. to add / remove the latent heat of transition).

Coupling MD and DPD

Dynamics: self-diffusion across interfase



Coupling MD and DPD

AdResS

`pros`

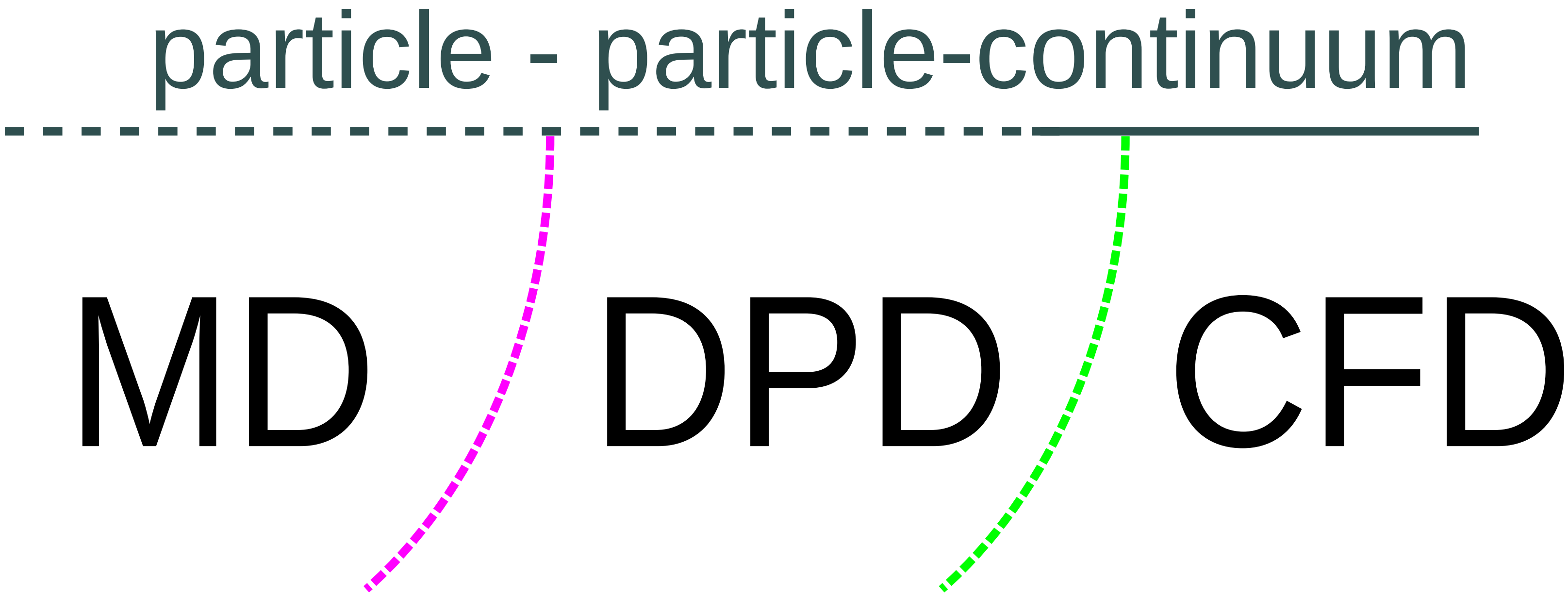
- Reduction of degrees of freedom for the liquid outside the region of interest.
- Conserves momentum (3rd Newton Law by construction)
- Recovers the fluid structure and pressure in the coarse-grained domain
- Self-diffusion of atomistic and coarse-grained domains can be *somehow* matched (a first-principles theory is lacking in the literature).

Coupling MD and DPD

AdResS

`cons`

- It does not conserves energy $\Longrightarrow$ heat transfer is not described.
- Substantial work to pre-evaluate the effective potential U^{cm} using `iterative Bolzmann inversion` for:
  - Both `cg` and `hyb` models.
  - Each thermodynamic state considered
- Dynamically restricted to homogenous, or near equilibrium states
- Pressure fits required for `cg` and `hyb` models
- Viscosity mismatch between coarse-grained and atomistic models $\Longrightarrow$ incorrect shear transfer.


particle - particle-continuum
MD
DPD
CFD

MD-DPD-CFD

Triple scale coupling

RDB, K. Kremer, M. Praprotnik, J. Chem. Phys, **128** 114110, (2008)

General motivation

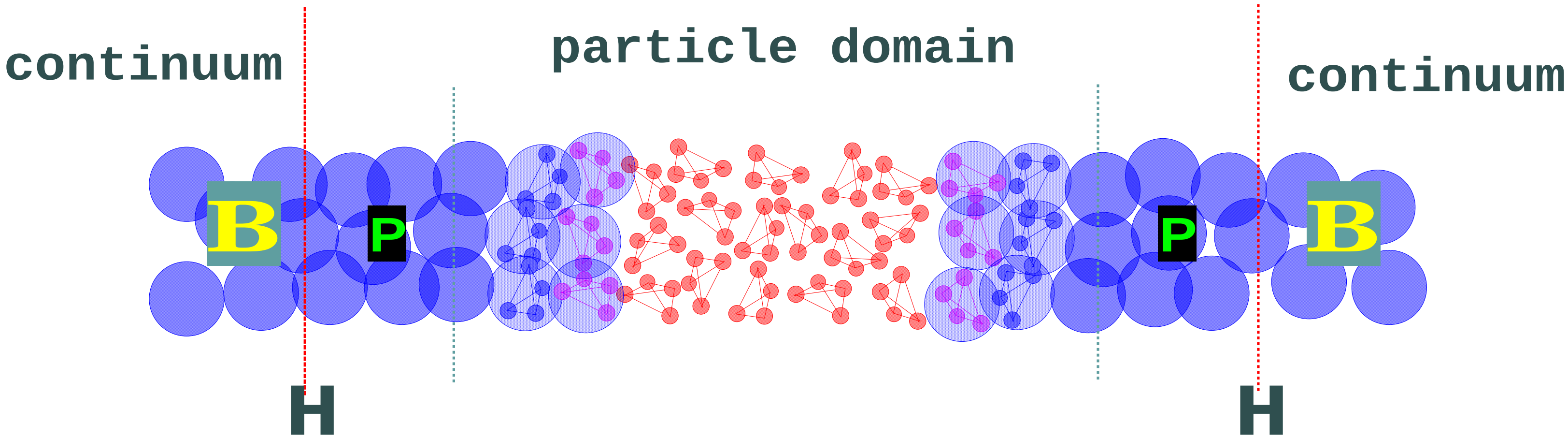
Complex molecules

- Technical issues
  - Generalize the (MD-DPD) AdResS scheme to include **hydrodynamics**
  - Solve the **insertion** of larger molecules in hybridMD
- Applications
  - Phenomena involving flow-matter interaction at multiple length scales complex fluids near surfaces, lubrication, macromolecules in flow,...
  - Grand canonical molecular dynamics involving complex molecules confined systems

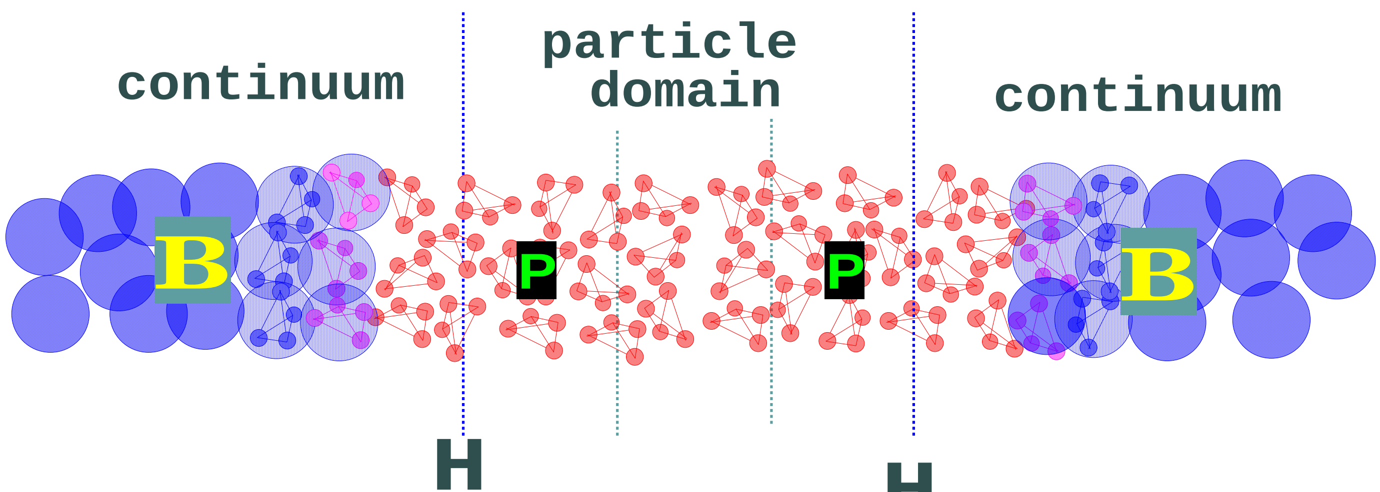
MD-DPD-CFD Two possible setups

RDB, K. Kremer, M. Praprotnik, J. Chem. Phys, **128** 114110, (2008)

Homogeneous (CG) buffer



Heterogeneous model buffer



MD-DPD-CFD: Two possible setups

RDB, K. Kremer, M. Praprotnik, J. Chem. Phys, **128** 114110, (2008)

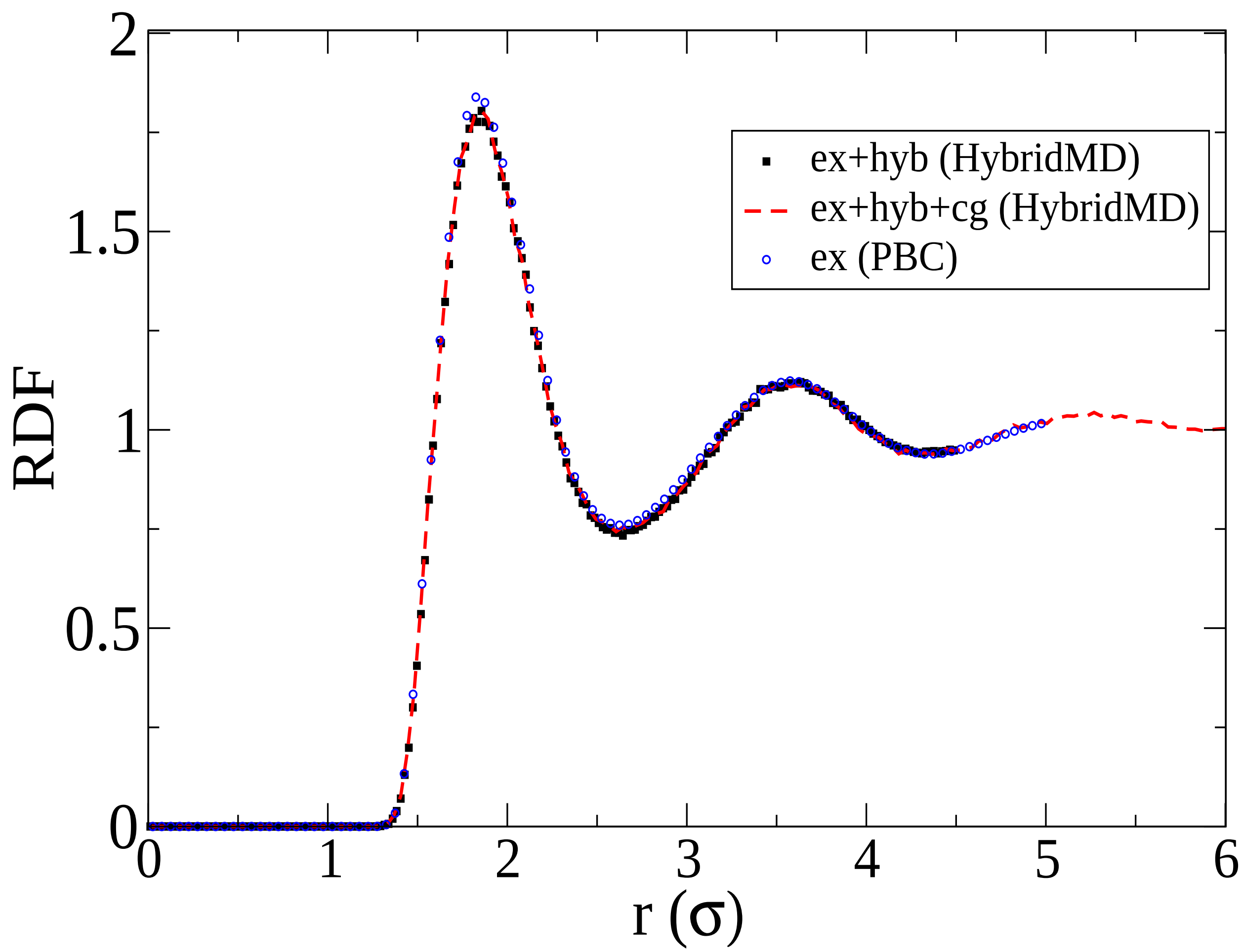
- **Homogeneous buffer**
  - `con`: Requires fine tunning of `CG` model
    - Viscosity **or** molecular diffusion coefficient
      Transversal DPD C. Junghans, et al., Soft Matter 4, 156 (2008)
    - Equation of state
  - `pro`: Requires smaller buffer size
  - `pro`: Permits to introduce CG molecular information into the MD (explicit) region (structure, diffusion rates, etc...)

- **Heterogeneous buffer**
  - `con`: Larger buffer size
  - `pro`: Fully atomistic MD region: proper viscosity, EOS, fluctuations.
  - `pro`: Does not requires fine tunning of `CG` model and `HYB` models
  - `pro`: Enables **energy exchange**, as the MD region is fully explicit.

MD-DPD-CFD: Equilibrium

liquid structure around the hybrid interface

Radial distribution function
high density tetraedral liquid



MD-DPD-CFD: **Equilibrium: grand canonical**

Mass fluctuations

- Scaled standard deviation of mass $\sigma_N^2/V = \rho k_B T \left(\frac{\partial p}{\partial \rho}\right)_T^{-2}$

ρ	simulation	Grand canonical
0.1	0.2	0.17
0.175	0.1	0.07

- Standard deviation number of particles in one cell, $V = 15 \times 15 \times 3\sigma^3$ similar values within error bars

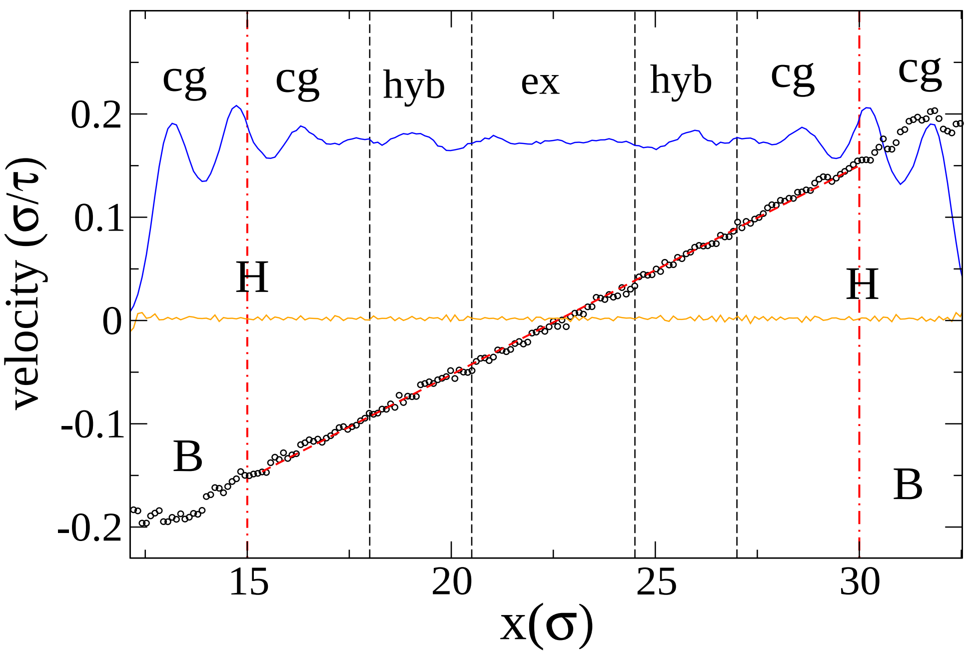
Coarse Grained	hyb	atomistic
13.9	14.2	14.5

MD-DPD-CFD: **Shear flow**

Homogeneous buffer

high density tetraedral liquid under shear

density and velocity profiles

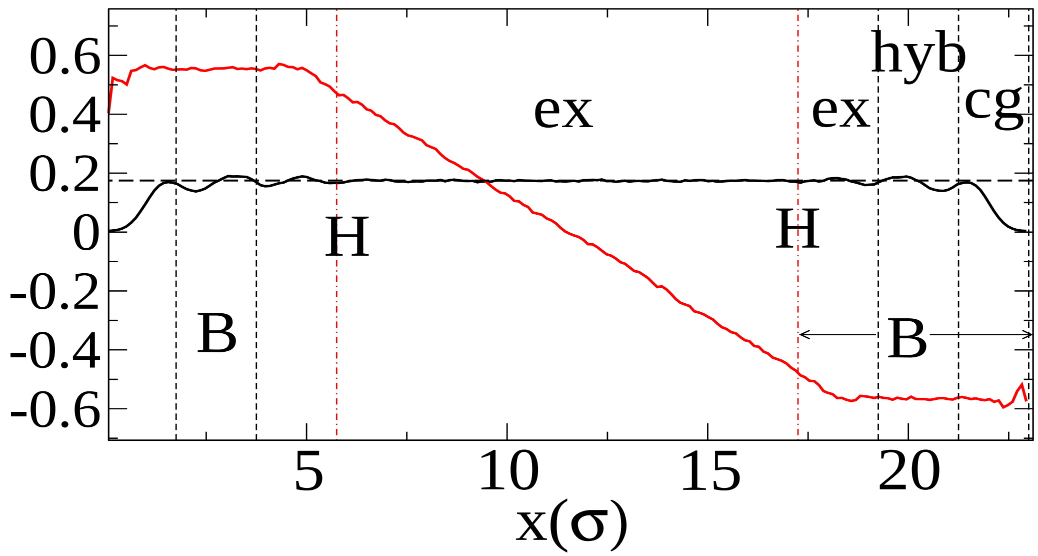


MD-DPD-CFD: **Shear flow**

Heterogeneous buffer

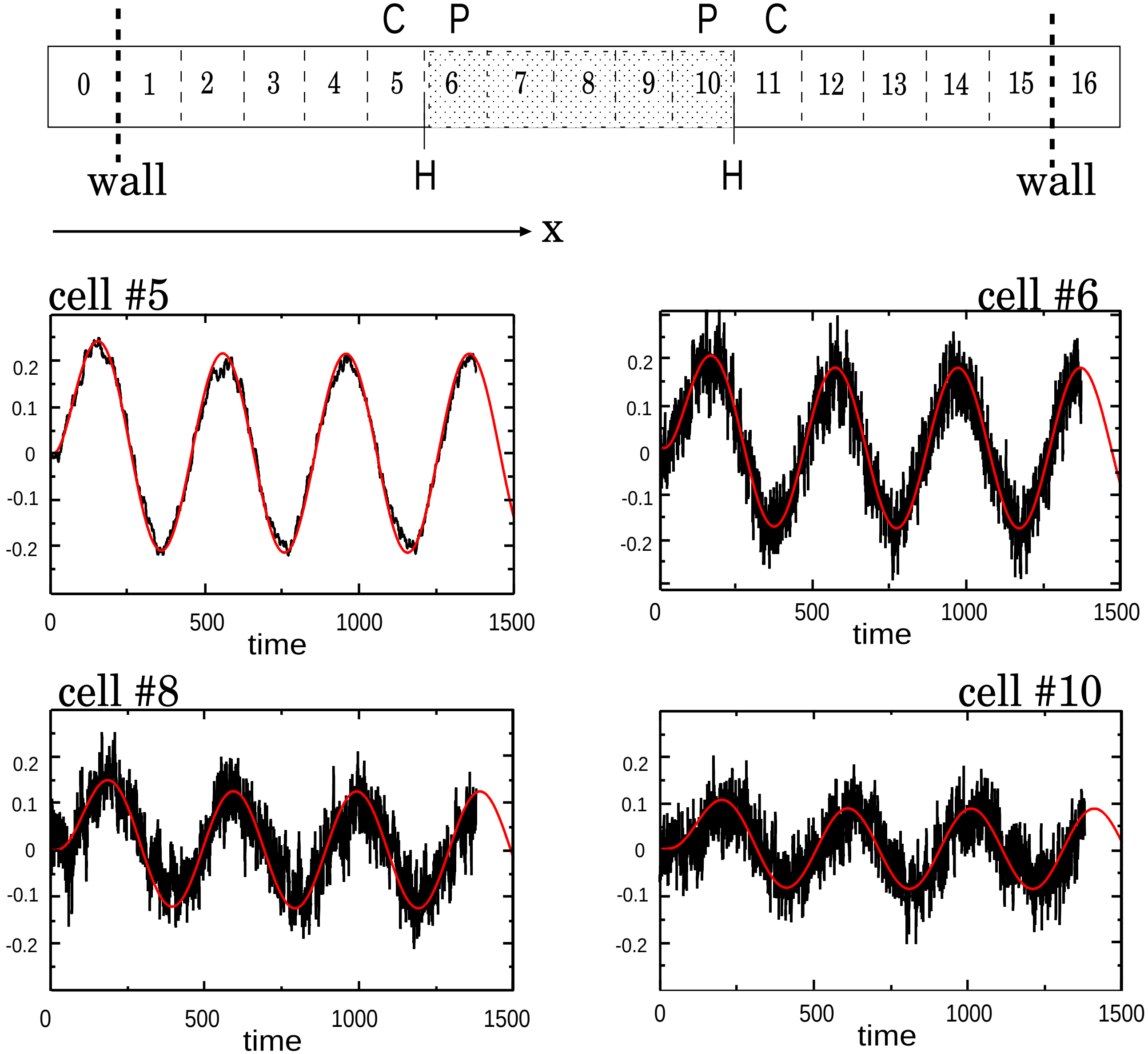
high density tetraedral liquid under shear

density and velocity profiles



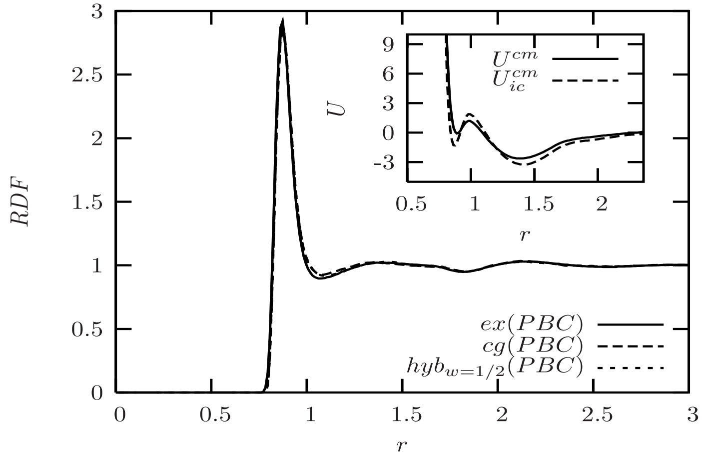
MD-DPD-CFD: Unsteady flows

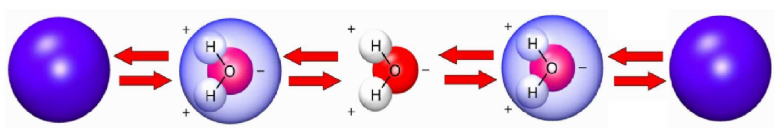
Stokes flow (oscillatory shear)

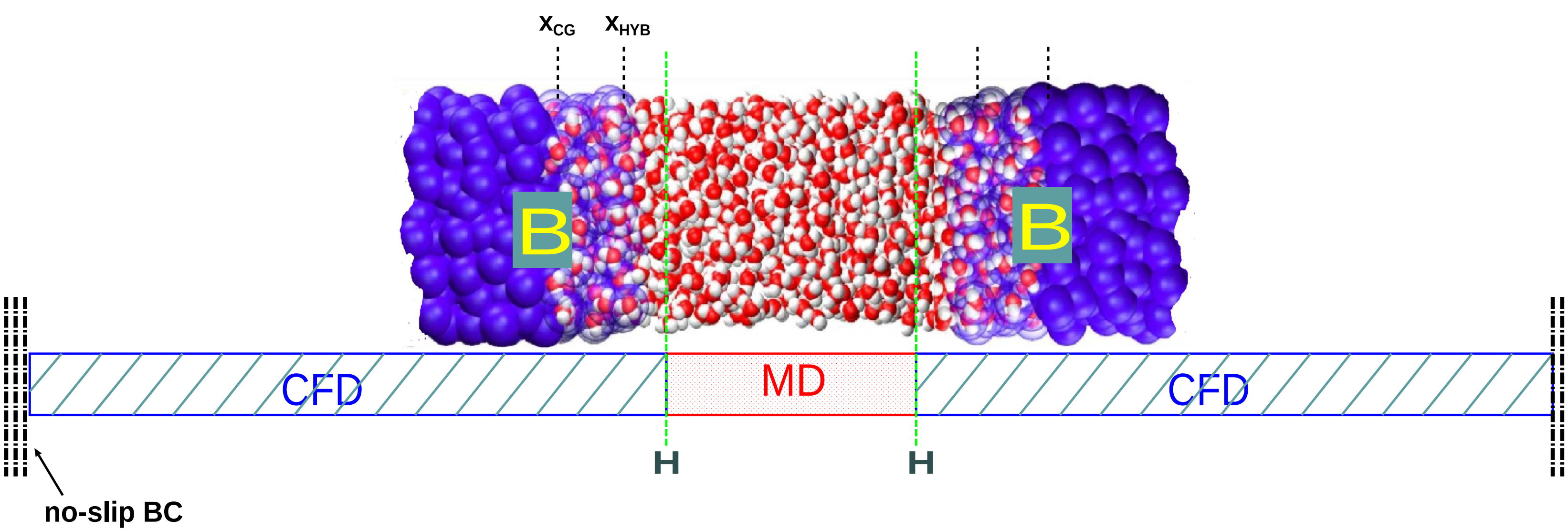


Triple scale for water using an heterogeneous buffer

RDB, Praprotnik, Kremer, (to be submitted)







The heterogeneous buffer does not require accurate fits for the CG and HYB models

Viscosities (oxigen-LJ units)

CG $\eta=20$

EX $\eta=45$

Flexible TIP3P water model

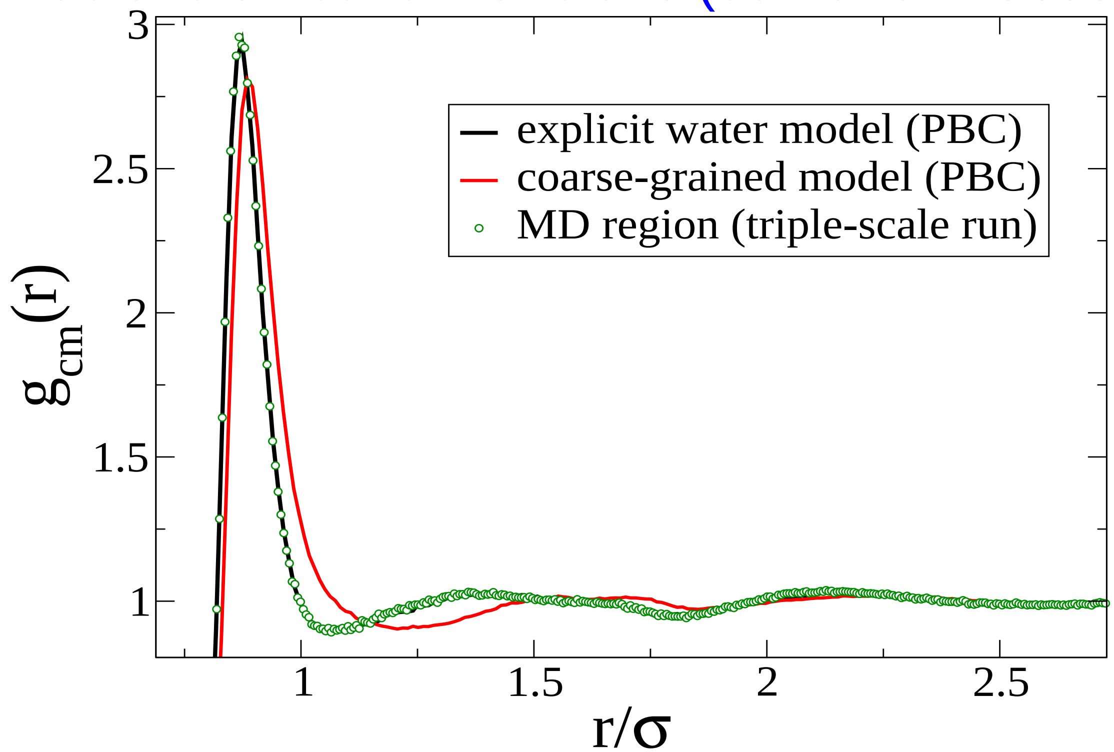
Mass fluctuation

Volume = 10.5x6.18x11.2σ^{-3}

Var[ρ] = 0.0108, Thermodynamics

Var[ρ]=0.011(2), 3-S simulations

Radial distribution funcions (center of masses)

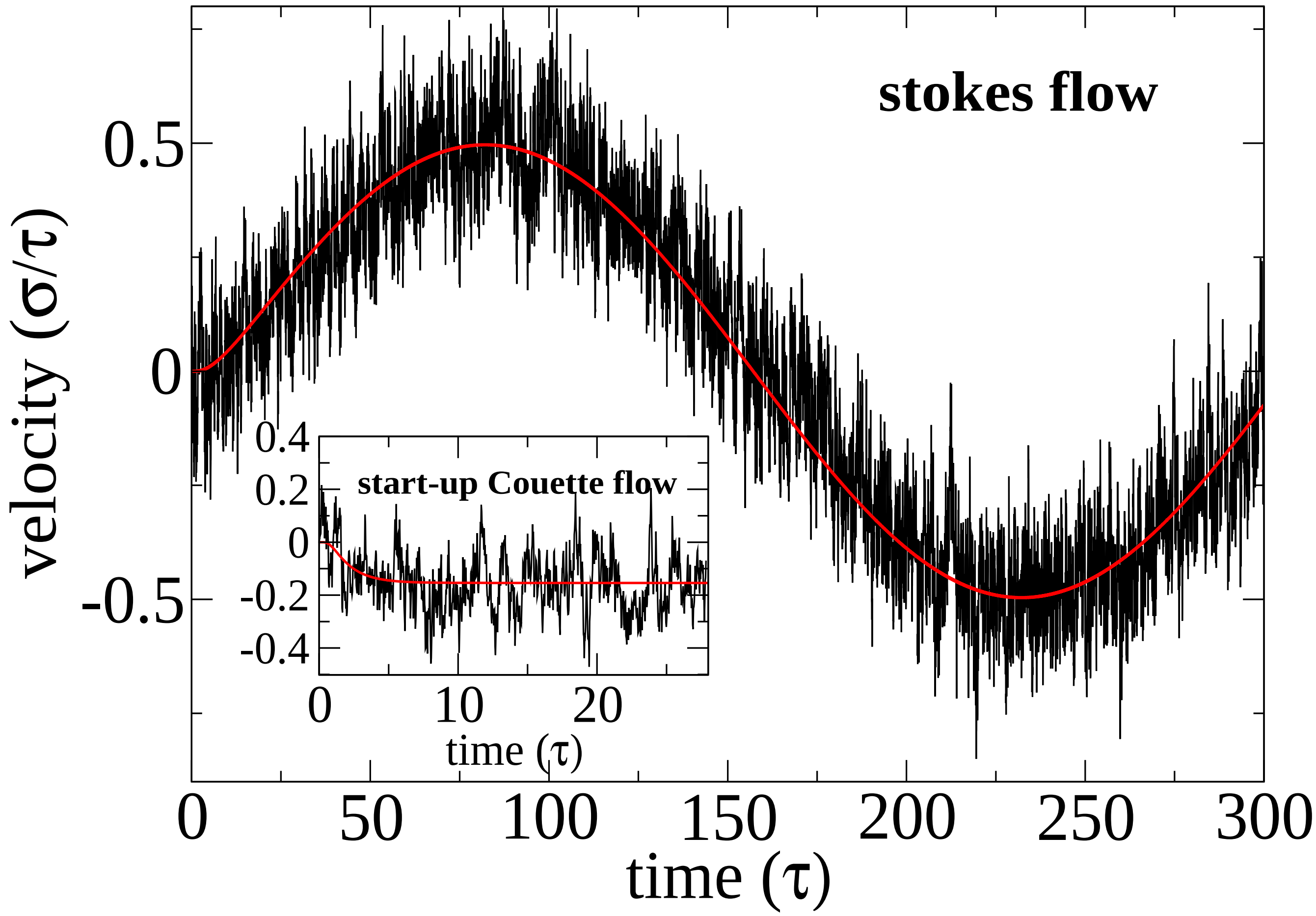


Triple scale for water

Viscosities (oxigen-LJ units)

CG $\eta=20$ EX $\eta=45$

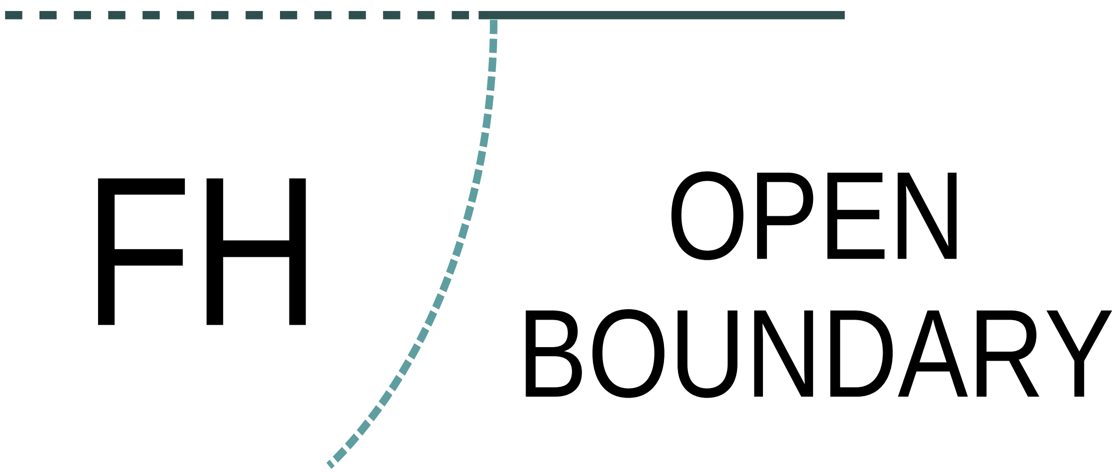
Flexible TIP3P water model



Concluding remarks

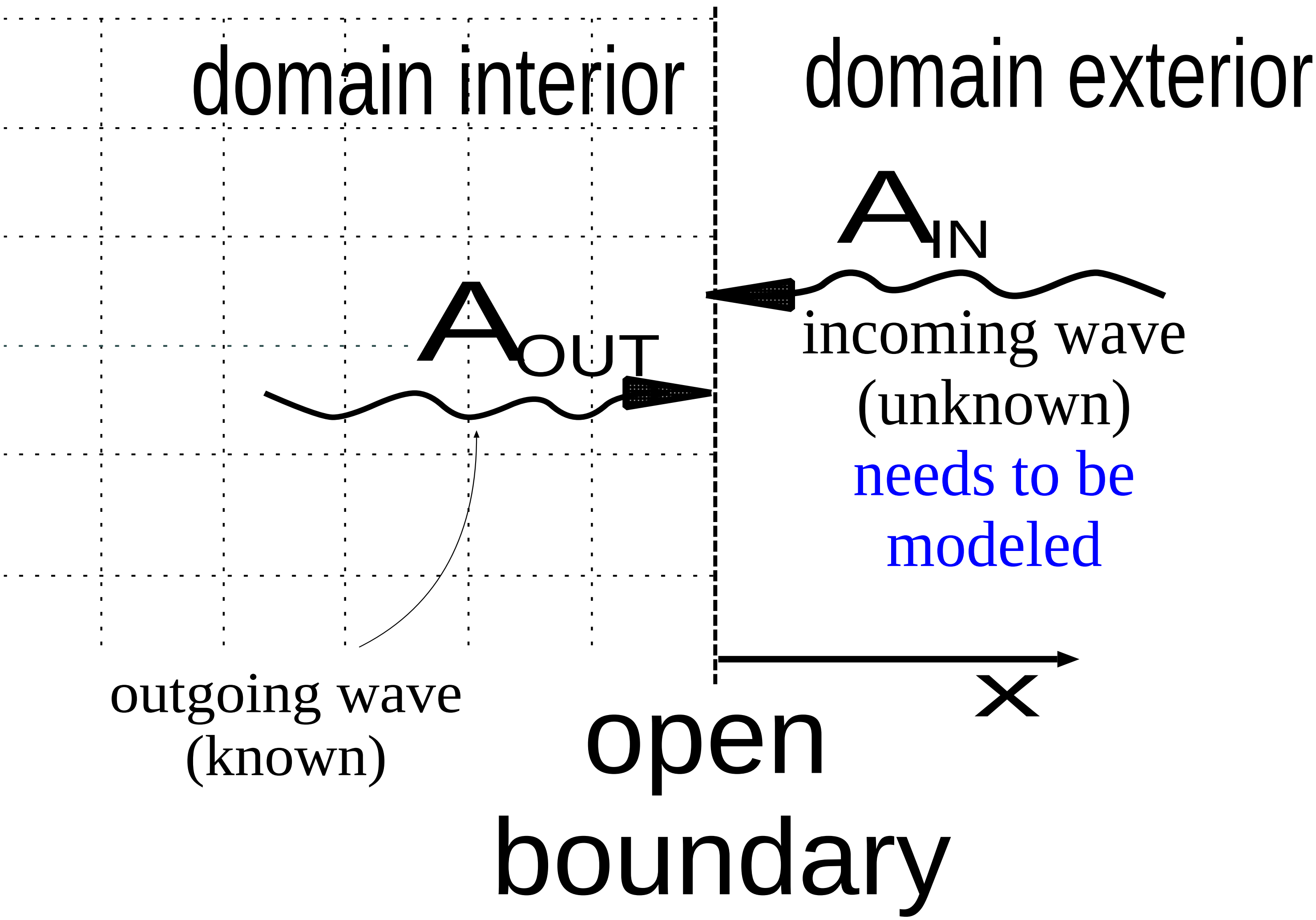
- Multiscale modeling based on domain decomposition
  - HybridMD: MD-Fluctuating hydrodynamics.
    - Sound, heat and energy transfer
    - Open molecular dynamics (grand canonical μVT and other ensembles)
  - Adaptive coarse-graining: MD-CG
    - Proper coarse-grained structure and pressure
    - Diffusive (mass) transport across hybrid interface can be matched
  - Triple scale model: MD-CG-continuum
    - Coarse-grained (DPD like) intermediate model
    - Proper hydrodynamics on shear and isothermal sound transport (not heat)
    - Solves insertion of complex molecules in hybrid schemes
    - Heterogeneous buffer: more flexible and robust.
  - Open boundaries for fluctuating hydrodynamics:
    - Evacuation of sound waves.
    - Can be generalized to energy and vorticity.

Non-reflecting boundary conditions for fluctuating hydrodynamics



RDB, Anne Dejoan, Phys Rev E. (in press)

Non-reflecting boundary conditions for CFD: set-up



Implementation of non-reflecting boundary conditions.

$$\text{density} \quad : \quad \frac{\partial \rho}{\partial x} = 0$$

$$\text{velocity} \quad : \quad \frac{\partial u}{\partial t} + \frac{1}{2\rho_e c}(L_{OUT} - L_{IN}) = 0$$

Closure models for the incoming waves

$L_{OUT} = \lambda_{OUT}\left(\frac{\partial P}{\partial x} + \rho c \frac{\partial u}{\partial x}\right)$	Evaluated at the interior domain
$L_{IN} = 0$	`cons:` ill posed, overall pressure drift
$L_{IN} = K(p - p_{eq}) \quad K = \frac{\sigma c}{L}$	`cons:` reflection of low freqs.
$L_{IN} = K(\rho c A_{IN}) = \frac{K}{2}(\delta p - \rho_e c \delta u)$	`pros:` *Wave masking*. Enables fluctuation-dissipation balance.

NRBC for FH: Fluctuation-dissipation balance for incoming waves

- Stochastic eq. for incoming wave amplitude:

$$\frac{dA_{IN}(x_b)}{dt} + KA_{IN}(x_b) = F(t)$$

- Fluctuating stress: $F(t) \equiv \frac{1}{\Delta x \rho_e}\left[\tilde{\Pi}_{xx}(x_b + \frac{\Delta x}{2}) - \tilde{\Pi}_{xx}(x_b - \frac{\Delta x}{2})\right]$

$$\langle F(t)F(0)\rangle = 2\Phi\delta(t) = \frac{4k_B T \eta_L}{\Delta x^2 \rho_e^2 V_c}\delta(t)$$

- Stochastic boundary **dynamics**: $\langle A_{IN}(t)A_{IN}(0)\rangle = \frac{\Phi}{K}\exp(-Kt)$.

  $\langle A_{IN}\rangle = 0$ and $\boxed{\langle A_{IN}^2\rangle = \frac{\Phi}{K}}$.

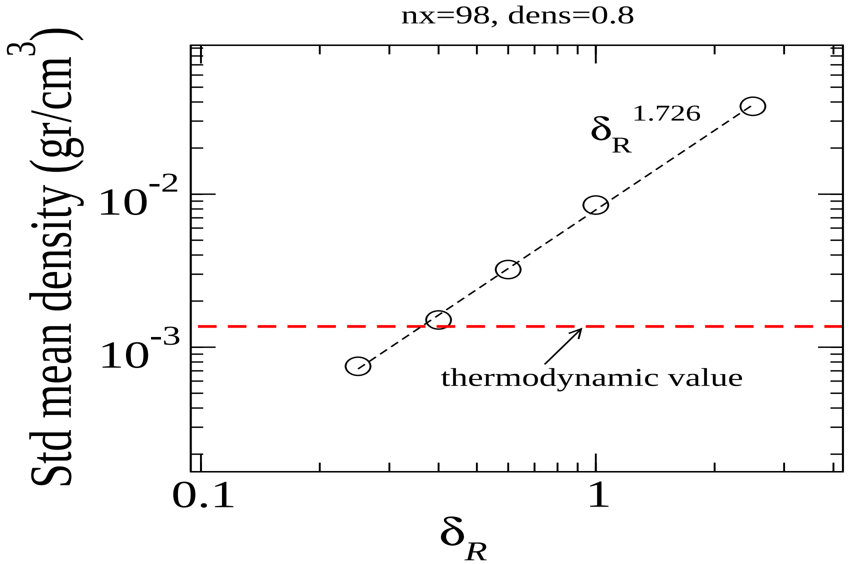
- Sound amplitude variance, **thermodynamics,** $A_{IN} = (1/2)(c\delta\rho/\rho_e - \delta u)$.

  $\boxed{\langle A_{IN}^2\rangle = \frac{1}{2}\frac{k_B T}{\rho_e V_c}}$

- **Relaxation rate**: $\boxed{K = \frac{\nu_L}{(\delta_R \Delta x)^2}}$ with $\delta_R^{(theor)} = 0.5$

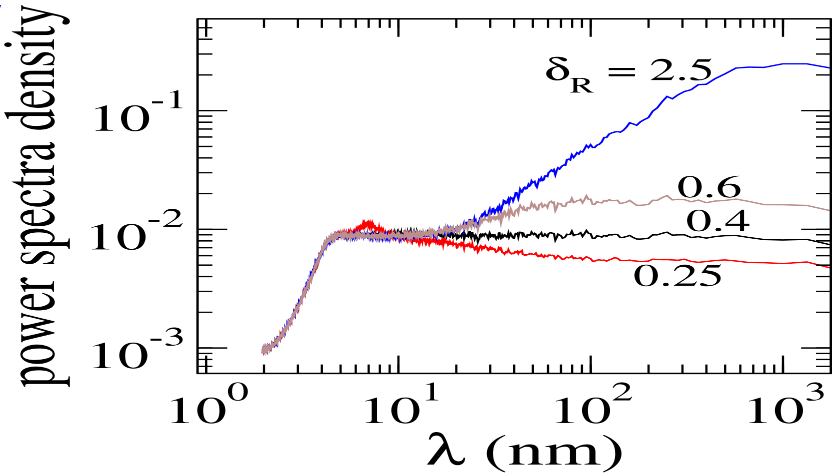
Mean density fluctuation at equilibrium: grand canonical ensemble,

$$\langle(\delta\bar{\rho})^2\rangle = \frac{k_B T}{c^2 V}$$

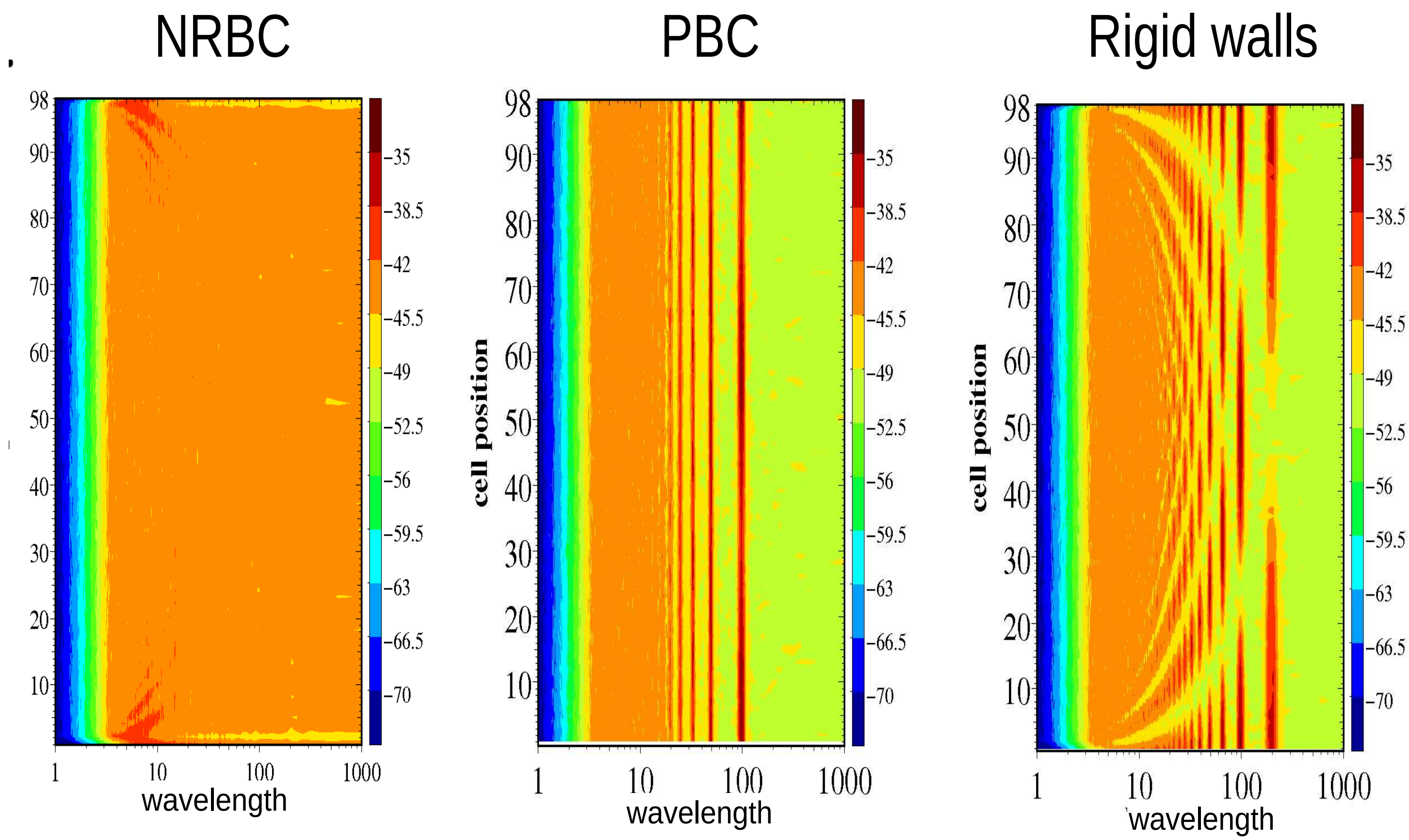


$\delta_R^{(num)} = 0.4$

Sound power spectral density within the system interior

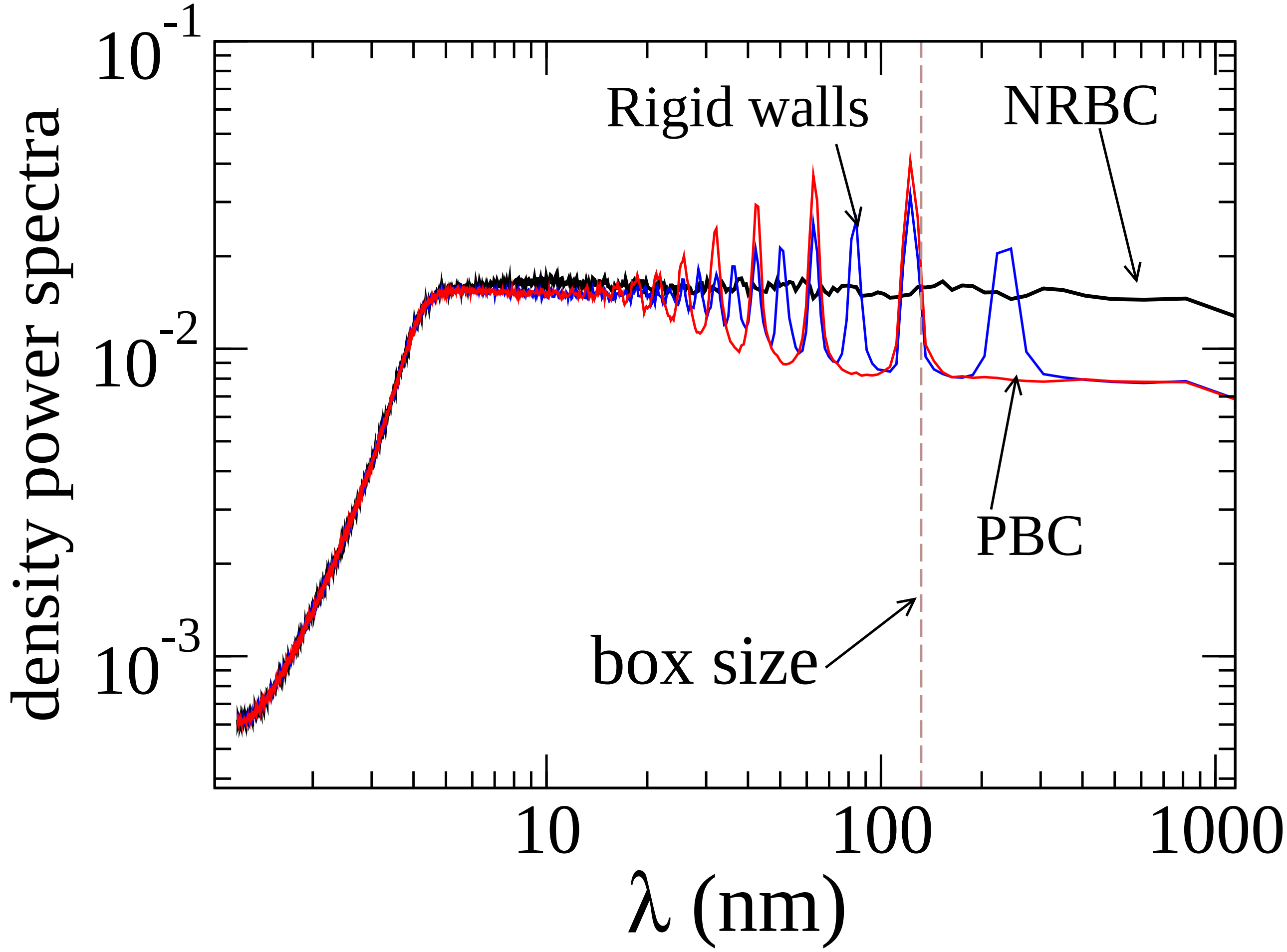


Comparsison between Non-reflecting boundaries (NRBC), periodic (PBC) and rigid walls

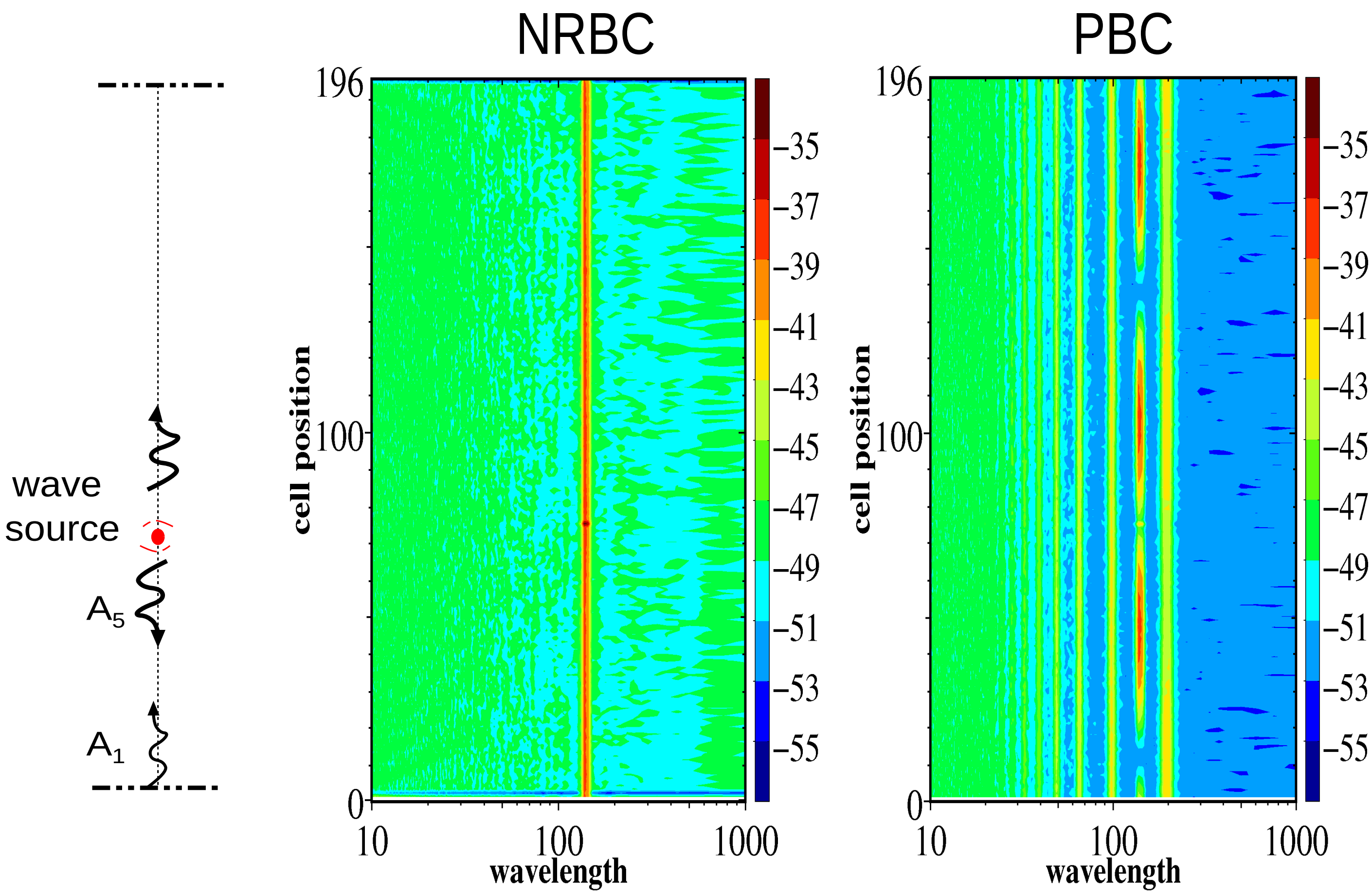


Comparison with PBC and Rigid walls:

PSD of waves within the system



Forced waves: evacuation of sound



Reflection coefficient

$$r \simeq 10^{-3}(f\Delta x)^{1.5}$$