Analytical and Numerical Study of Coupled Atomistic-Continuum Methods for Fluids

Weiqing Ren
Courant Institute, NYU

Joint work with Weinan E
Multiscale modeling for two types of problems:

- Complex fluids - Constitutive modeling
- Microfluidics - Atomistic-based boundary condition modeling

(i) Heterogeneous multiscale method: Macro solver + missing data from MD

(ii) Domain-decomposition framework

Koplik et al. PRL ’88; Thompson & Robbins, PRL ’89; Qian et al., PRE ’03; Ren & E, PoF ’07

Koumoutsakos, JCP ’05
Multiscale method in the domain decomposition framework

The two descriptions are coupled through exchanging boundary conditions in the overlapping region after each time interval $T_c$.
Two fundamental issues:

• What information need to be exchanged between the two descriptions?
  (i) Fields (e.g. velocity):
  (ii) Fluxes of conserved quantities

• How to accurately impose boundary conditions on molecular dynamics?
Existing multiscale methods for dense fluids

• Velocity coupling:
  
  O’Connel and Thompson 1995
  Hadjiconstantinou and Patera 1997
  Li, Liao and Yip 1999
  Nie, Chen, E and Robbins 2004
  Werder, Walther and Koumoutsakos 2005

• Flux coupling:
  
  Flekkoy, Wagner and Feder 2000
  Delgado-Buscalioni and Coveney 2003

• Mixed scheme:
  
  Ren and E 2005
Present work:

• Stability and convergence rate of the different coupling schemes; Propagation of statistical errors in the numerical solution.

• Error introduced when imposing boundary conditions in MD.
Problem setup: Lennard-Jones fluid in a channel

\[
\begin{align*}
U & \rightarrow \quad & z=L \\
& \leftarrow -U & z=-1 \\
C\text{-region} & \\
& \quad z=b \\
& \quad \quad \quad \quad P\text{-region} \\
& \quad \quad \quad \quad \quad C\text{-region} \\
\end{align*}
\]

(i). Static \((U=0)\);  \hspace{1cm} (ii). Impulsively started shear flow

\[
\begin{align*}
\rho \partial_t u - \partial_z \tau &= 0 \\
\tau &= \mu \partial_z u \\
u(L, t) &= U
\end{align*}
\]

\[
\begin{align*}
m_i \ddot{r}_i &= F_i \\
F_i &= -\sum_{j\neq i} \nabla V(r_{ij})
\end{align*}
\]

\[
V(r) = 4\varepsilon \left( \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^6 \right)
\]
Four coupling schemes:

- Velocity - Velocity
- Momentum flux - Velocity
- Velocity - Flux
- Flux - Flux

The two models exchange BCs after every time interval $T_c$. 
The rest of the talk:

• Algorithmic details of the multiscale method for the benchmark problems;
• Numerical results;
• Assessment of the error introduced in the imposition of boundary condition in MD.
Solving the continuum model

\[
\rho \partial_t u - \partial_z \tau = 0 \quad \rightarrow \quad \rho \frac{u_{i+1/2}^{n+1} - u_{i-1/2}^{n+1}}{\Delta t} - \frac{\tau_{i+1}^n - \tau_i^n}{h} = 0
\]

\[
\tau = \mu \partial_z u \quad \rightarrow \quad \tau_i = \mu \frac{u_{i+1/2} - u_{i-1/2}}{h}
\]

\[u_{1/2} \text{ or } \tau_1 \text{ is supplied by MD.}\]
Correspondence of hydrodynamics and molecular dynamics

\[ \partial_t m^\omega + \nabla \cdot \tau^\omega(x, t) = 0 \]

\[ m^\omega(x, t) = \sum_i p_i(t) \delta(r_i(t) - x) \]

\[ \tau^\omega(x, t) = \sum_i \frac{1}{m_i} (p_i \otimes p_i) \delta(r_i - x) \]

\[ + \frac{1}{2} \sum_{j \neq i} (r_{ij} \otimes F_{ij}) \int_0^1 \delta(\lambda r_i + (1 - \lambda)r_j - x) \, d\lambda \]

( Irving-Kirkwood 1950)

\[ \langle m^\omega \rangle \Rightarrow \rho u \]

\[ \langle \tau^\omega \rangle \Rightarrow \tau = \rho u \otimes u \]

\[ - \mu(\nabla u + \nabla u^T) + pI \]  for Newtonian fluids

Using these formulae to calculate the continuum BCs from MD.
Boundary conditions for MD:
Reflection BC + Boundary force

\[ F_n(r_w) = \rho \int_{z\geq r_w} \frac{\partial V}{\partial z} g(r) \, dr \]

\[ g(r) = \text{radial distribution function} \]
Matching with continuum -
Imposing velocity BC on MD

\[ m_i \dot{v}_i = F_i + f(t), \quad \text{for } r_i \in \Omega \]

\[ f(t) : \quad N^{-1}_\Omega \sum_{r_i \in \Omega} v_i = u_c \]
Matching with continuum:
Imposing shear stress on MD

Particle with distance $r_w$ to the boundary experiences a shear force $\tau_c f_s(r_w)$.
Microscopic profile of shear stress

Left: Shear stress profile at various shear rates; Right: Shear stress profile rescaled by the shear rate.
Summary of the multiscale method

- **Continuum solver**: Finite difference in space + forward Euler’s method in time

- **Molecular dynamics**:
  1. Velocity Verlet, Langevin dynamics to control temperature;
  2. Reflection BC + Boundary force;
  3. Projection method to match the velocity;
  4. Distribute the shear stress based on an universal profile.
Numerical result for the static problem:

\[ T_c = \Delta t \]

(i). Errors are due to statistical errors in the measured boundary condition (velocity, or shear stress) from MD.

(ii). Errors are bounded in VV, FV, VF schemes, while accumulate in the FF scheme.
Numerical result for the static problem:

\[ T_c = 10\Delta t \]
Numerical result for the static problem:

\[ T_c = 1.67 \times 10^3 \]
Analysis of the problem for \( T_c = +\infty \)

Velocity - Velocity coupling scheme:

\[
\begin{align*}
    u_1(z) &= \frac{L - z}{L - a} \xi_1 \\
    u_2(z) &= \left( \frac{a(L - b)}{b(L - a)^2} \xi_1 + \frac{1}{L - a} \xi_2 \right) (L - z) \\
    u_n(z) &= \left( \sum_{i=1}^{n} k^{n-1}_{vv} \xi_i \right) \frac{L - z}{L - a} \\
    k_{vv} &= \frac{a(L - b)}{b(L - a)} \approx 1 - \frac{c}{b}
\end{align*}
\]

--- Amplification factor

\[
\langle \| u_n \|_2 \rangle \leq \left( \frac{1}{3(1 - k_{vv}^2)} \right)^{1/2} \sigma_v \quad \text{where} \quad \sigma_v^2 = \langle \xi_i^2 \rangle
\]
Analysis of the problem for \( T_c = +\infty \)

The numerical solution has the following form:

\[
\begin{align*}
    u_n(z) &= \left( \sum_{i=1}^{n} k^{n-i} \xi_i \right) g(z) \\
    k_{vv} &= \frac{a(L-b)}{b(L-a)} \approx 1 - c/b \\
    &\quad \left| k_{vv} \right| < 1 \\
    k_{fv} &= \frac{a}{a-L} \approx -a/L \\
    &\quad \left| k_{fv} \right| \ll 1 \\
    k_{vf} &= \frac{b-L}{b} \approx -L/b \\
    &\quad \left| k_{vf} \right| \gg 1 \\
    k_{ff} &= 1 \\
    g_{vv}(z) &= g_{fv}(z) = (L-z)/(L-a) \\
    g_{vf}(z) &= g_{ff}(z) = z-L
\end{align*}
\]
Analysis of the problem for finite $T_c$

Conclusions:

(i). The VV and FV schemes are stable;

(ii). Velocity-Flux is stable when $T_c < T^*$, and unstable when $T_c > T^*$

(iii). Flux-Flux scheme is weakly unstable.
A dynamic problem: Impulsively started shear flow

\[ U \quad z=L \]

\[ z=b \quad z=a \]

\[ P\text{-region} \]

\[ C\text{-region} \]

\[ C\text{-region} \]

\[ z=-1 \]

\[ \rho \partial_t u - \partial_z \tau = 0 \]

\[ \tau = \mu \partial_z u \]

\[ u(L, t) = U \]

\[ u(x, 0) = 0 \]
Numerical results: $T_c = \Delta t$

**Velocity-Velocity**

**Flux-Velocity**
Numerical results: $T_c = \Delta t$
Steady state calculation: Comparison of convergence rate

\[ k_{vv} = 1 - \frac{\text{overlapping region}}{\text{particle region}} = 0.7 \]

\[ T_c = 8000 \Delta t \]

\[ k_{fv} = \frac{\text{size of p-region}}{\text{system size}} = 0.08 \]
Assessment of the error from the imposition of velocity BC in MD

\[ e_\tau = \frac{|\tau(d) - \tilde{\tau}|}{|\tilde{\tau}|} \]

\( \tilde{\tau} = \text{exact shear stress from MD using Lees-Edwards BC} \)
Assessment of the error from the imposition of velocity BC in MD

\[ e_\tau = \text{Error of stress} = \frac{|\tau(d) - \tilde{\tau}|}{|\tilde{\tau}|} \]

\( \tilde{\tau} = \text{exact shear stress from MD using Lees-Edwards} \)
Assessment of the error for $d < r_c$

\[
\frac{\mu_d (U - u)}{l} = \frac{\mu_\infty u}{L}
\]

\[
u = \frac{\tilde{\mu}_d}{\tilde{\mu}_d + \tilde{l}} U
\]

\[
e_\tau = \left| \frac{u}{U} - 1 \right| = \frac{\tilde{l}}{\tilde{l} + \tilde{\mu}_d}
\]

\[
l = O(1\sigma) \quad L = \text{system size}
\]

\[
\tilde{\mu}_d = \frac{\mu_d}{\mu_\infty} \quad \tilde{l} = \frac{l}{L}
\]

$\mu_d = \mu_\infty$

:= viscosity in bulk when $d > r_c$

$\mu_d < \mu_\infty$ when $d < r_c$
Error of the stress: Analysis vs. Numerics

\[ \tilde{\mu}_d = \frac{\int_0^d f_s(z) \, dz}{\int_0^\infty f_s(z) \, dz} \]

Red curve: \( e_\tau = \frac{\tilde{l}}{\tilde{l} + \tilde{\mu}_d} \)

Blue curve: Numerics
Summary:


(2). Error introduced when imposing velocity boundary condition in MD.

Ongoing work:

Boundary conditions for non-equilibrium MD;

Incorporating fluctuations in the BC of MD;
Coupling fluctuating hydrodynamic with molecular dynamics.
Improved numerical scheme: Using ghost particles

- Less disturbance to fluid structure
- Mass reservoir for 2d velocity field
References:

• Analytical and Numerical study of coupled atomistic-continuum methods for fluids, *preprint*

• Boundary conditions for the moving contact line problem, *Physics of Fluids, 19, 022101 (2007)*

• Heterogeneous multiscale method for the modeling of complex fluids and microfluidics, *J. Comp. Phys. 204, 1 (2005)*