Coupled continuum hydrodynamics and molecular dynamics method for multiscale simulation

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Abstract We present a new hybrid methodology for carrying out multiscale simulations of flow problems lying between continuum hydrodynamics and molecular dynamics, where macro/micro lengthscale separation exists only in one direction. Our multiscale method consists of an iterative technique that couples mass and momentum flux between macro and micro domains, and is tested on a converging/diverging nanochannel case containing flow of a simple Lennard-Jones liquid. Comparisons agree well with a full MD simulation of the same test case.

Keywords: Micro Flows, Molecular Dynamics, Multiscale, Hybrid, Continuum Hydrodynamics

1 Introduction

The well established and traditional approach to solving fluid dynamic problems is to use continuum models, such as the Navier-Stokes (NS) equations (with no velocity slip at bounding surfaces). The NS equations work remarkably well for many cases, ranging from extremely large systems, such as flows around submarine hulls, right down to tiny systems, such as flows through capillaries. However, when the characteristic length-scale of the problem approaches an atomistic scale, the physics of the flow requires a more accurate molecular approach, such as molecular dynamics (MD). The major problem is that MD is too computationally intense to simulate reasonably large length and time-scales (e.g. micro-scale). Despite this limitation, simple Newtonian liquids at these scales show non-continuum phenomena occur only in proximity to surfaces [1–3]. In the bulk, the NS equations can be safely applied [4]. The problem in the last decade has therefore been how to deploy a multiscale physics methodology that bridges the gap between the micro and macro scales.

Hybrid methods can be loosely categorised into: (a) domain-decomposition techniques [5–8] and (b) heterogeneous multiscale methodologies (HMM) [9, 10], see Fig 1. Domain-decomposition techniques divide the computational domain into micro and macro sub-domains with an overlapping region at the two interfaces that serves for mutual coupling. The disadvantage of this approach is that it decouples length-scales, but not time-scales: the micro sub-domain still needs to be integrated over the same time-scale as the macro solver. Larger systems imply longer integration times to reach steady state, so the method is generally practical only for small systems in steady state. The HMM approach aims instead to decouple both length-scales and time-scales by distributing micro sub-domains on nodes of the macro domain (which is the whole domain), and running the micro and macro solvers on different clocks. The HMM approach is well suited to largely scale-separated systems.

In this paper we present a new multiscale approach similar to the HMM but with the differences shown in Fig 2, in this case applied to a converging/diverging nanochannel case.

(a) Domain-decomposition method
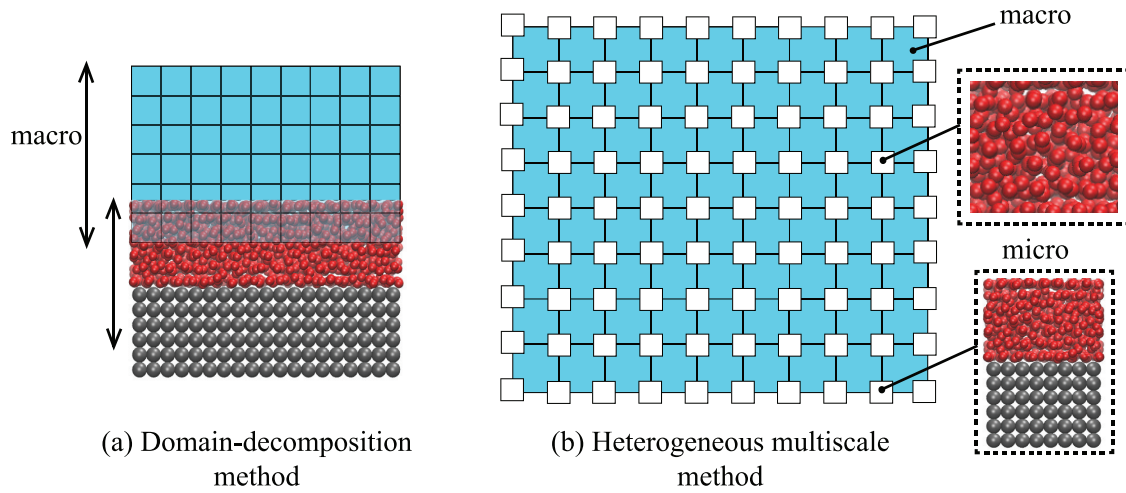
(b) Heterogeneous multiscale method

Figure 1: Schematics of (a) the domain decomposition method, and (b) the HMM.
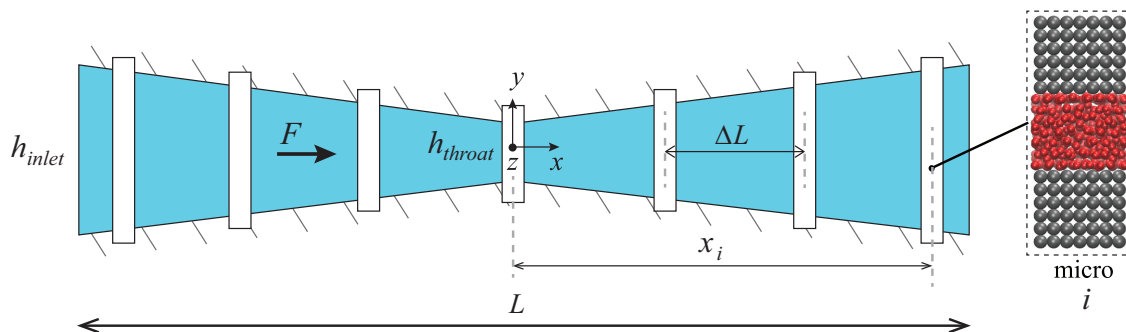

Figure 2: Illustration of our multiscale technique applied to a converging/diverging nanochannel case. Scale-separation is exploited in the $x-$direction by distributing MD sub-domains with a top and bottom wall (molecular type) and coupled in a multiscale framework by applying hydrodynamic constraints.

The method exploits directional scale separation (in the figure, denoted by the x−direction) to simulate multiscale flows through channels of generally long aspect ratios.

2   Simulation method

The multiscale method we present here is for tested on an axially-periodic converging/diverging channel flow, driven by a gravity-type force. The full MD simulation of the case requires a constant force $\mathbf{f}^{ext} = F\hat{\mathbf{n}}_x$ applied to all liquid molecules in the domain, where $\hat{\mathbf{n}}_x$ is the direction vector in the axial x-direction. Cyclic boundary conditions are applied at the inlet and outlet in order to impose an axially repeating geometry.

The multiscale setup of this case is illustrated in Fig 2. There are Π MD sub-domains defined in the multiscale simulation, each with different x_i−positions, where i $(1,\ldots,\Pi)$ is an arbitrary MD box. The length of the channel is denoted by L and the separation between MD sub-domains is $\Delta L(x)$. For purposes of simplicity, we assume that the channel is symmetric so that Π only covers one side (converging *or* diverging) of the channel such that the channel height of the ith MD simulation box is given by:

$$h_i = h_{throat} + \frac{h_{throat}(i-1)}{\Pi - 1}, \qquad (1)$$

where h_{throat} is the height of the throat at which the converging and diverging parts of the channel meet.

2.1   Molecular dynamics

The micro simulations are described by molecular dynamics [11, 12]. We use mdFoam [13–15], a parallelised non-equilibrium MD solver that is open-source and available to download from [16]. Molecule positions $\mathbf{r}_k(t) = (x_k, y_k, z_k)$, and velocities $\mathbf{v}_k(t) = (u_k, v_k, w_k)$ evolve according to Newton's equation of motion:

$$\frac{d}{dt}\mathbf{r}_k = \mathbf{v}_k(t), \qquad (2)$$

$$m_k \frac{d}{dt}\mathbf{v}_k = \mathbf{f}_k + \mathbf{f}^{ext}, \qquad (3)$$

where $k = (1, \ldots, N)$ is an arbitrary molecule in the system, m_k is the molecule mass, $\mathbf{f}^{ext}$ is an external force, and $\mathbf{f}_k$ is the total force due to interacting molecule neighbours:

$$\mathbf{f}_k = \sum_{j=1(\neq k)}^{N} \nabla U(r_{kj}). \qquad (4)$$

The interaction potential $U(r_{kj})$ between liquid-liquid and solid-liquid molecules governs the physics of the flow. In this paper we simulate monatomic liquid argon using the 12-6 Lennard Jones (LJ) potential:

$$U_{12-6}(r_{kj}) = 4\epsilon \left[\left(\frac{\sigma}{r_{kj}}\right)^{12} - \left(\frac{\sigma}{r_{kj}}\right)^{6} \right], \quad (5)$$

where $\epsilon = 1.6568 \times 10^{-21}$J and $\sigma = 0.34 \times 10^{-9}$m are the potential's characeristic energy and length scales, and $r_{kj} = |\mathbf{r}_k - \mathbf{r}_j|$ is the separation of two arbitrary molecules within a cut-off radius $r_{cut} = 4\sigma$.
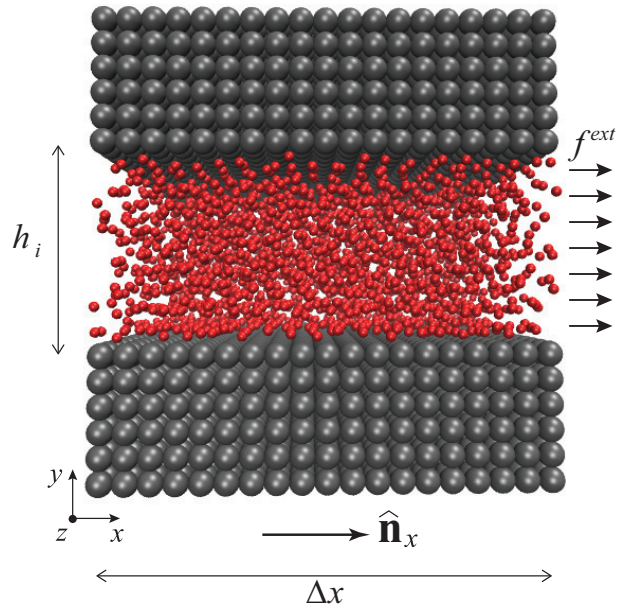

Figure 3: MD simulation box.

The MD simulation boxes for the multiscale method (see Fig 3) are of identical sizes $(\Delta X = \Delta Z = 20\sigma)$ except the cross-channel

height, which varies according to Eqn. (1). The walls of thickness, 6σ are modelled as 'frozen' wall molecules that interact with the liquid using the same LJ parameters. Periodic boundaries are applied in the $x-$ and $z-$directions of the simulation box, and an external force $\mathbf{f}^{ext} = f^{ext}\hat{\mathbf{n}}_x$ applied to all liquid molecules drives the desired flow. A Berendsen thermostat [17] is imposed on the MD box that sets the temperature at $T = 87K$, in order to remove heat generated by the work done by external forces.

2.2 Hybrid approach

There are three macroscopic conditions that constrain the micro MD simulations:

1. periodicity,

2. continuity, and

3. momentum conservation.

2.2.1 Momentum conservation

For the steady and incompressible case we are considering in this paper, the conservation of momentum equation is:

$$\rho F \mathbf{n}_x = -\nabla p + \nabla \cdot \sigma, \qquad (6)$$

where F is the axial applied force, ρ is the number density, p is the pressure and σ is the shear stress.

In the MD micro elements the momentum balance is:

$$\rho \mathbf{f}^{ext} = \nabla \cdot \sigma. \qquad (7)$$

The pressure gradient term in Eqn. (6) does not feature in Eqn. (7) due to the streamwise periodicity of the micro MD domains. It is highly complex to prescribe such a pressure gradient directly, and so here we impose it indirectly through the external forcing:

$$\rho \mathbf{f}^{ext} = \rho F \hat{\mathbf{n}}_x + \nabla p. \qquad (8)$$

Substituting Eqn. (8) into Eqn. (7) yields the desired momentum Eqn. (6). If we integrate Eqn. (8) over the channel length, the pressure gradient term is removed (because we have imposed periodicity at inlet and outlet), and we are left with a convenient constraint on the external force, f^{ext}:

$$\frac{1}{L} \int_0^L \mathbf{f}^{ext} dx = F\hat{\mathbf{n}}_x = \mathbf{F}, \qquad (9)$$

where L is the length of the channel. Equation (9) states that the spatial mean of the computational forces in each micro simulation, must be kept equal to the physical gravity force, $\mathbf{F} = \bar{\mathbf{f}}^{ext}$.

2.2.2 Continuity

Continuity requires that the mass flow rate, $\dot{m}$, must be the same for all the micro elements. This can be enforced by appropriately modifying the axial variation of $f^{ext}(x_i)$ (while ensuring that the mean value of the force is equal to F, in accordance with above).

Since force is roughly proportional to mass flow rate, a simple proportional estimator for the correct axial variation can be used, which we outline in the multiscale algorithm described next.

2.3 Multiscale procedure

The solution to the multiscale problem is an iterative one, which involves solving the continuity and momentum conservation equations across all the detached MD sub-domains. The iterative procedure is outlined in the following steps:

1. Assign an initial force $f_i^{(n=0)} = F$ to all Π micro simulations. (Note the direction of the force points in the x-direction: $\mathbf{f}_i^{ext} = f_i\mathbf{n}_x$).

2. Simulate all Π simulations for τ_m molecular time-integration steps, until steady-state is reached.

3. Measure the mean mass flow rates $\dot{m}_i$ over S statistical number of samples:

$$\dot{m}_i = \frac{1}{S\Delta X} \sum_{t=1}^{S} \sum_{k=1}^{N_i} m_k \mathbf{v}_k(t) \cdot \mathbf{n}_x, \quad (10)$$

where N_i are the number of molecules in the ith MD simulation, $m_k, \mathbf{v}_k(t)$ are the molecules mass and instantaneous velocity respectively and ΔX is the $x-$direction size of the MD box.

4. Take the mass flux from the smallest channel height and set it as the target mass flux, $\dot{m}_T = \dot{m}_s(h_s \leq h_i)$.

5. At the new iteration step, $n = n+1$, compute the new forces that satisfy mass and momentum conservation:

$$f_i^* = f_i^{(n)}\frac{\dot{m}_T}{\dot{m}_i}, \qquad (11)$$

$$\Delta F = F - \frac{1}{L}\int_0^L f_i^* dx, \qquad (12)$$

$$f_i^{(n+1)} = f_i^* + \Delta F, \qquad (13)$$

where L is the length of the channel and ΔF is a measure of the global force convergence.

6. Repeat from 2, until convergence (i.e. $\Delta F = 0$, and $\dot{m}_i = \dot{m}_T$). For practicality, convergence is assumed when ΔF varies between $\pm 0.02 \times 10^{-12}$N over several iterations.

3   Results

For the purpose of this paper we consider a nano-scale converging/diverging channel of length $L = 60\sigma$. The multiscale simulation consists of 5 streamwise distributed MD subdomains, which are reduced to 3 because the converging/diverging channel is symmetric at the throat; only channel heights from inlet to throat are taken. The channel heights of the micro-simulations are 8σ, 6σ and 4σ respectively, which are separated uniformly by a spacing of $\Delta L = 15\sigma$.

A full MD simulation of the same test case is also carried out in order to validate the multiscale simulations, see Fig. 5. A gravitational-like force, $f^{ext} = 1.22 \times 10^{-12}$N is applied to generate flow across the full MD system. Five

measurement planes are placed in the streamwise channel, with same 15σ spacing as in the multiscale simulation, for direct measurements of mass flux. The net mass flow rate at a plane is measured by averaging the number of molecules which cross it over a prescribed time period; molecules which cross in the positive $x-$direction are counted as positive and those which cross in the opposite direction are counted as negative. The average mass flow rate was calculated to be $(67.81 \pm 1.5\times)10^{-12}$ kg/s.
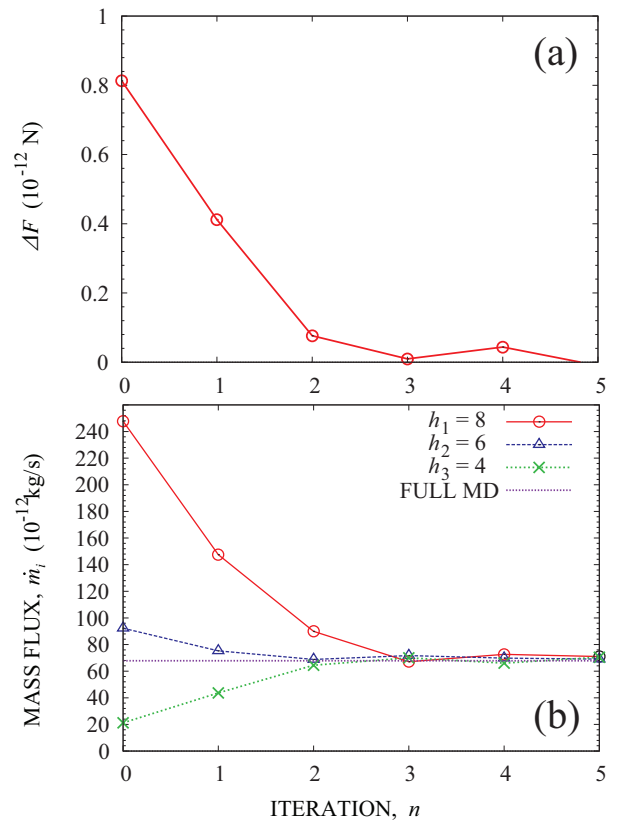

Figure 4: Convergence results from the multiscale procedure: (a) global force variations and (b) mass fluxes from i MD boxes, against iteration number n.

Results from the multiscale simulation show that convergence is obtained over a very short number of iterative steps ($n \sim 3-4$), as seen in Fig. 4(a) for the force term, ΔF in Eqn. (12). Figure 4(b) also verifies that the mass flux in the three individual MD boxes converge to the value calculated in the full MD simulation.

We plot the spatial variation of forces and

mass fluxes in the streamwise direction of the converging/diverging channel in Fig. 6. We see that the mass fluxes converge to a uniform value in all streamwise x_i positions, coinciding with the planar flux measurements from the full MD simulation. Furthermore, Fig. 6(b) shows the converged non-uniform axial force distribution $f^{ext}(x_i)$ for enforcing continuity in all micro elements.

To assess the computational advantage of our multiscale method over full MD we consider the following criteria based on spatial and temporal decoupling. Separation of spatial scales will yield a computational advantage if $\Delta L/L \ll 1$ (i.e. the spacing of micro simulations relative to the total length of the channel), $\Delta X/\Delta L \ll 1$ (overlapping micro simulations in the $x-$direction), and $2\Delta Z(h_{inlet} - h_{throat})/L \ll 1$ (the rate of variation of channel cross sectional area). In our test case we get the following values respectively, 0.25, 1.33 and 0.42.

Time-scale separations may be assessed by evaluating the relaxation time of the flow to reach steady-state after the external force is imposed. The basic criteria should then be $\tau_{micro}n/t_{macro} \ll 1$. For the full MD simulation $t_{macro} \sim 1.3$ns, while for the small MD boxes $\tau_{micro} \sim 0.1$ns (and another 0.1ns to achieve the necessary statistics when computing mass flux). The value for time-scale separation is 0.46.

Although most of the scale-separation values are below 1 (except the criteria for overlapping micro simulations), they are still not much less than 1, to achieve desirable speed up. We expect that by increasing the channel length L and keeping the height at the inlet/outlet and throat the same, we would get much larger scale separations, and therefore much larger computational savings using our multiscale approach.

4   Conclusions

We have presented a new multiscale iterative algorithm for coupling hydrodynamics and molecular dynamics through constraints of continuity and momentum. The iterative scheme was tested on an axially-periodic converging/diverging nanochannel, with a flow driven by a gravity-type force. The axial mass flow rate in the multiscale simulation agreed well with measurements taken from a full MD simulation of the same test case. Although small computational savings were observed for the small case considered in this paper, it is predicted that the full potential of this technique will be seen in larger (more realistic) systems, where substantial scale separations exist. The application of the method on GPUs

The multiscale method described in this paper is applicable to arbitrary-shaped nanochannels, so long that there exists spatial scale variation in one direction. The method may be used to study the fast transport of water in micro-scale long carbon-nanotubes of nanoscale diameters [18], or to study bifurcation laws for designing micro- and nano-fluidic devices [19].

5   Acknowledgments

6   References

[1] J. Koplik and J. R. Banavar. Continuum deductions from molecular hydrodynamics. *Annual Review of Fluid Mechanics*, 27:257–292, 1995.

[2] K. P. Travis and K. E. Gubbins. Poiseuille flow of Lennard-Jones fluids in narrow slit pores. *Journal of Chemical Physics*, 112(4):1984–1994, Jan 2000.

[3] S. A. Somers and H. T. Davis. Microscopic dynamics of fluids confined between smooth and atomically structured solid surfaces. *Journal of Chemical Physics*, 96(7):5389–5407, 1992.
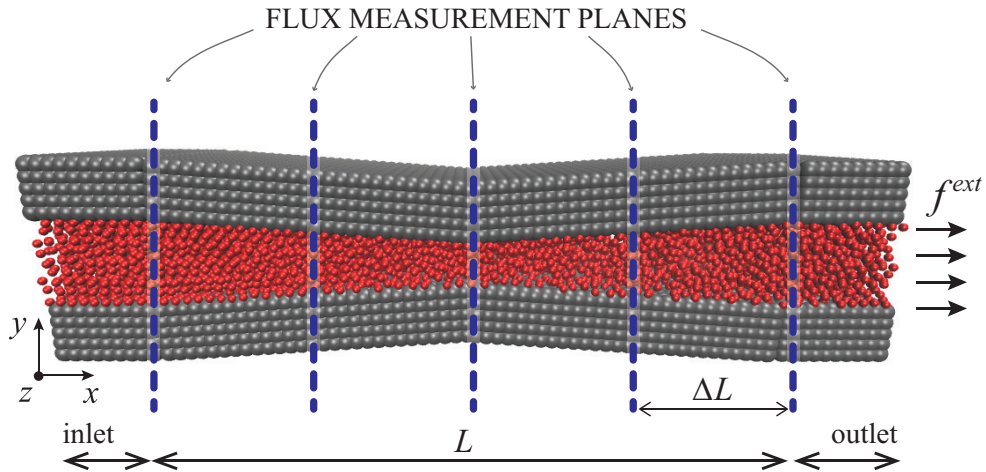
Figure 5: Full MD simulation setup of the converging/diverging nanochannel case.
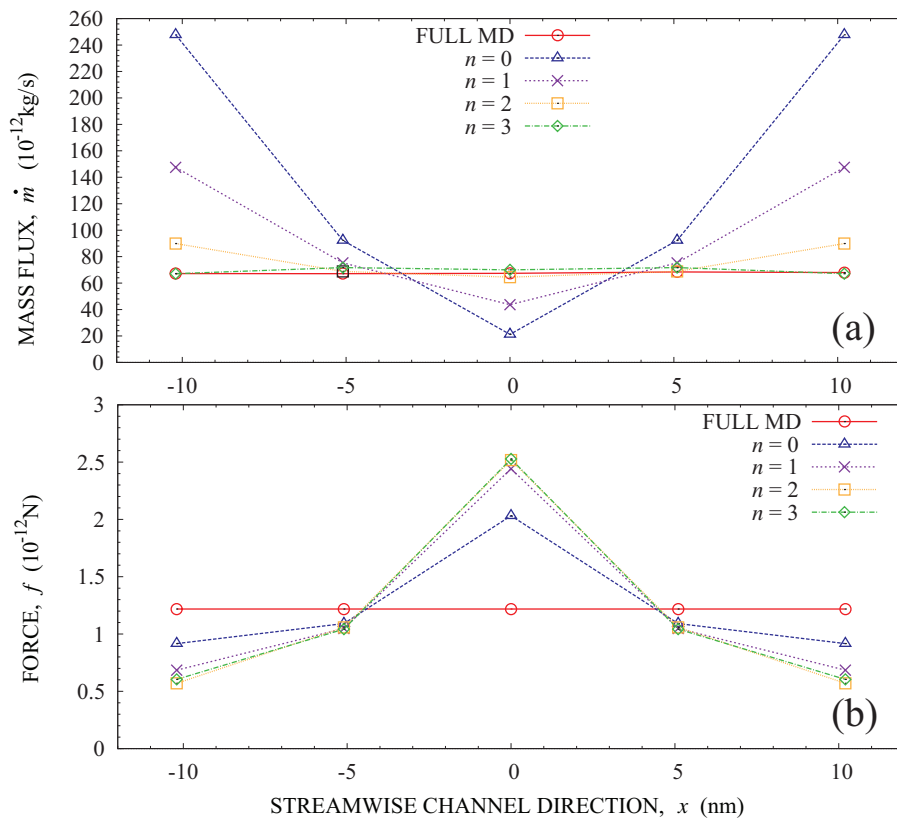

Figure 6: Streamwise results from the multiscale procedure showing convergence of: (a) uniform mass flux and (b) non-uniform force variations. Comparisons are made to the full MD simulation.

[4] H. Okumura and D. M. Heyes. Comparisons between molecular dynamics and hydrodynamics treatment of nonstationary thermal processes in a liquid. *Physical Review E*, 70(6):061206, 2004.

[5] S. T. O'Connell and P. A. Thompson. Molecular dynamics–continuum hybrid computations: A tool for studying complex fluid flows. *Physical Review E*, 52:R5792–R5795, December 1995.

[6] N. Hadjiconstantinou and A. Patera. Heterogeneous atomistic-continuum methods for dense fluid systems. *International Journal of Modern Physics C*, 8(4):967–976, 1997.

[7] X. B. Nie, S. Y. Chen, W.N. E, and M. O. Robbins. A continuum and molecular dynamics hybrid method for micro- and nano-fluid flow. *Journal of Fluid Mechanics*, 500:55–64, 2004.

[8] R. Delgado-Buscalioni and P. V. Coveney. Continuum-particle hybrid coupling for mass, momentum, and energy transfers in unsteady fluid flow. *Physical Review E*, 67(4):1–13, 2003.

[9] W. Ren and W. E. Heterogeneous multiscale method for the modeling of complex fluids and micro- fluidics. *Journal of Computational Physics*, 204(1):1–26, 2005.

[10] W. E, W. Ren, and E. Vanden-Eijnden. A general strategy for designing seamless multiscale methods. *Journal of Computational Physics*, 228:5437–5453, 2009.

[11] M. P. Allen and D. J. Tildesley. *Computer Simulation of Liquids*. Oxford University Press, 1987.

[12] D. C. Rapaport. *The Art of Molecular Dynamics Simulation*. Cambridge University Press, 2nd edition, 2004.

[13] G. B. Macpherson, M. K. Borg, and J. M. Reese. Generation of initial molecular dynamics configurations in arbitrary geometries and in parallel. *Molecular Simulation*, 33(15):1199–1212, December 2007.

[14] G. B. Macpherson and J. M. Reese. Molecular dynamics in arbitrary geometries: Parallel evaluation of pair forces. *Molecular Simulation*, 34(1):97–115, Jan 2008.

[15] M. K. Borg, G. B. Macpherson, and J. M. Reese. Controllers for imposing continuum-to-molecular boundary conditions in arbitrary fluid flow geometries. *Molecular Simulation*, 36(10):745, 2010.

[16] OpenFOAM. www.openfoam.org.

[17] H. J. C. Berendsen, J. P. M. Postma, W. F. van Gunsteren, A. Dinola, and J. R. Haak. Molecular dynamics with coupling to an external bath. *Journal of Chemical Physics*, 81:3684–3690, October 1984.

[18] W. D. Nicholls, M. K. Borg, D. A. Lockerby, and J. M. Reese. Water transport through (7,7) carbon nanotubes of different lengths using molecular dynamics. *Submitted to the Journal of Microfluidics and Nanofluidics*, 2011.

[19] R. Barber and D. Emerson. Optimal design of microfluidic networks using biologically inspired principles. *Microfluidics and Nanofluidics*, 4(3):179–191, March 2008.