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MULTISCALE SIMULATIONS USING PARTICLES

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Summary We outline a multiscale algorithm for the simulation of dense fluids that couples molecular dynamics and continuum fluid dynamics. The coupling between the two models is realized by a common overlap domain and by the alternating Schwarz method. We prefer this method to those based on direct flux exchange, since the accurate estimation of the fluxes requires much more statistics. The boundary conditions on the atomistic region are imposed using stochastic walls to thermalize and confine the molecular dynamics system, and the USHER algorithm to insert new atoms. The forcing of the continuum is achieved using appropriate source terms in the momentum equations. To test the algorithm, we study the flow of liquid argon past a carbon nanotube.

INTRODUCTION

The study of fluid mechanics at the nanoscale level [1] has received wide attention due to its importance for the understanding and development of biosensors. From the computational side, classical molecular dynamics (MD) have so far been the most useful tool to characterize phenomena such as wetting, hydrophobicity and boundary conditions. One system of particular interest with regard to biosensors is carbon nanotubes in water, which has recently been studied in some detail [2, 3]. Despite the success of molecular dynamics, its limitations in terms of accessible length and time scales are currently of the order of $10\,\mathrm{nm}^3$ and $10\,\mathrm{ns}$. In order to allow the computational analysis of nanoscale systems integrated in microfluidic environments, a multiscale approach is indispensable Here, we present an algorithm to impose arbitrary, non-periodic boundary conditions on MD simulations of dense fluids. We show how this algorithm couples standard molecular dynamics codes with commercial computational fluid dynamics packages. As a first application we consider a flow of liquid argon around a carbon nanotube (CNT), where only the CNT and its interaction with the nearby argon are modeled using MD, cf. Fig. 2, and where the continuum part is described by the isothermal, incompressible, Navier-Stokes (NS) equations.

THE COMPUTATIONAL METHOD

The central part of any hybrid scheme is the coupling of the atomistic and continuum regions. One can distinguish between two fundamental types of coupling, the former aim at matching the fluxes of mass, momentum, and energy at the interface [4, 5] and, the others are based on overlapping cells where the mass density ρ, the mean velocity $\boldsymbol{u}$, and the temperature T are imposed. The latter schemes have two main advantages over flux based schemes, namely that they decouple time scales [6] and that densities are substantially easier to sample than fluxes.

The Schwarz algorithm

The algorithm is based on the Schwarz iteration scheme [6], i.e., the computational domain consists of continuum cells C, particle cells A, and overlap cells X, where both descriptions are valid, cf. Fig. 1 The following iteration is run until

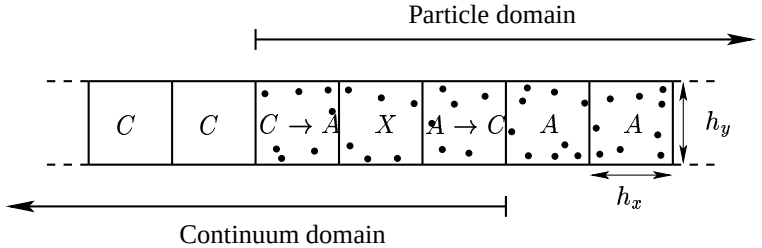

Figure 1. One dimensional sketch of a computational domain. There are pure continuum (C) and particle cells (A), as well as overlap cells $C \to A$, X, and $A \to C$, where both descriptions are valid. The Schwarz iteration by turns imposes the values measured in the $C \to A$ cells on the atomistic region and the values measured in the $A \to C$ cells on the continuum region until the solution converges in X.

the solution is converged in the overlap region.

Step 1) Run an MD simulation until a steady state is reached, with $(\rho, \boldsymbol{u}, T)$ imposed in the $C \to A$ cells.

Step 2) Solve the continuum equations with the steady state solution of the MD in the $A \to C$ cells as boundary conditions.

The atomistic part

The main difficulties in imposing the continuum field values on the atomistic system in the $C \to A$ domain are the introduction and removal of particles but also the altered dynamics of the molecules in the $C \to A$ region due to the

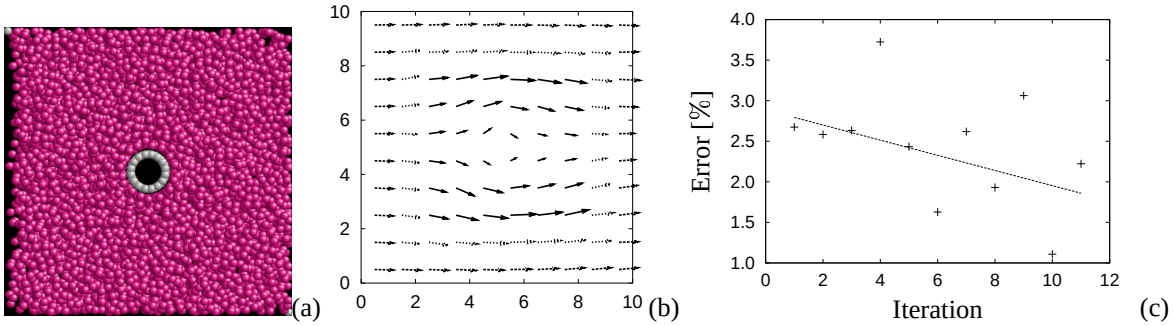

Figure 2. Flow of argon around a carbon nanotube. (a): A snapshot of the atomistic system. (b): The converged velocity field in the atomistic domain. (c): Convergence of the velocity field imposed on the edge of the atomistic domain. The error measure is defined in Eqn. (**??**). The solid line is a linear fit to the data.

"missing particles" exteriour to the atomistic domain. For a monatomic Lennard-Jones fluid, the first problem is solved using the USHER algorithm [7]. For the second problem, we use the notion of stochastic boundary condition introduced to simulate the contact of an MD system with a thermal reservoir [8].

The continuum part

The continuum system is described by a finite volume discretization of the isothermal, incompressible, Navier-Stokes (NS) equations in the entire computational domain including the atomistic region. By the complete overlay, we avoid explicit internal interfaces in the continuum system and instead utilize body forces to impose the average molecular velocity field. We note that the external boundary conditions of the continuum system are not limited to cyclic conditions, but can take any for the flow solver admissible form.

RESULTS

To test the proposed algorithm, we have interfaced a commercial Navier-Stokes solver (StarCD [9]), with an in-house molecular dynamics code (FASTTUBE [10]). In the study the carbon nanotube is modeled as a rigid structure which interacts with the argon through a Lennard-Jones potential. The size of the finite-volume cells are chosen to match the size and location of the bins used in the molecular system for the averaging and for the imposition of the boundary conditions. In the present study we use cells of size $1.0 \times 1.0 \times 4.26\,\mathrm{nm}$ to secure a sufficient averaging and forcing of the molecular system. The continuum mesh consists of $30 \times 30 \times 1$ mesh points. The boundary conditions of the continuum system are chosen to be periodic in y and z. A flow is imposed along the x-direction with an inlet velocity of $u_x = 100\,\mathrm{ms}^{-1}$ at the left boundary and an outlet boundary condition at the right boundary. The atomistic domain extends over $10 \times 10 \times 4.26\,\mathrm{nm}$ (including the $C \to A$ cells) and contains 5600 argon atoms. A snapshot of the atomistic domain is given in Fig. 2a. In every iteration of the atomistic domain, we let the system adapt to the new boundary conditions for $20\,\mathrm{ps}$, and we subsequently sample the system during $80\,\mathrm{ps}$. We quantify the convergence by measuring the change (E) in the velocity field between each main iteration. In Fig. 2c, we show this measure as a function of the iteration number. The statistical error in sampling the velocity in the atomistic region is estimated to be $\approx 2.5\%$. Therefore we conclude that the solution has nearly converged after the first iteration, cf. Fig. 2c.

CONCLUSION

We have presented a hybrid multiscale algorithm for the coupling of atomistic and continuum systems. It is based on the Schwarz domain decomposition technique, the Usher algorithm for particle insertions, and stochastic walls.

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