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Control algorithm for multiscale flow simulations of water

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We present a multiscale algorithm to couple atomistic water models with continuum incompressible flow simulations via a Schwarz domain decomposition approach. The coupling introduces an inhomogeneity in the description of the atomistic domain and prevents the use of periodic boundary conditions. The use of a mass conserving specular wall results in turn to spurious oscillations in the density profile of the atomistic description of water. These oscillations can be eliminated by using an external boundary force that effectively accounts for the virial component of the pressure. In this Rapid Communication, we extend a control algorithm, previously introduced for monatomic molecules, to the case of atomistic water and demonstrate the effectiveness of this approach. The proposed computational method is validated for the cases of equilibrium and Couette flow of water.

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Water is the most important solvent and an ubiquitous component of biological systems. The interaction of water with its environment and the ability to capture its behavior at all scales from atomistic to continuum is challenging and has stimulated much interest in multiscale computational approaches, beginning with the work of Clementi.1

Multiscale methods, coupling continuum models to atomistic descriptions, have been largely developed for fluids described in the atomistic regime by Lennard-Jones potentials. We may distinguish multiscale methods by the way information is exchanged between the two descriptions: by a direct flux exchange and by a Schwarz alternating method. A number of recent works has presented extensions to more realistic systems involving, for example, polyatomic, polar molecules such as water. Multiscale simulations of liquid water include the works of de Fabritiis et al. [3] who employed the flux exchange scheme and Praprotnik et al. [10] who presented a spatially adaptive molecular resolution procedure to transition from a coarse grained to an all-atom representation.

In this Rapid Communication, we present an application of the Schwarz alternating method [8,9] to perform multiscale simulations of liquid water. The key advantage of this approach, over flux-based schemes, is that it does not require the calculation of the pressure tensor; a quantity that can be difficult to compute accurately due to limited spatial or temporal resolution. At the same time, Schwarz algorithms require several iterations in order to ensure an approximate conservation of mass and momentum between the different descriptions whereas flux-based schemes are algebraically exact. Both schemes encounter a difficulty with density oscillations attributed to the removal of periodic boundary conditions (BCs) in the atomistic domain. Flux-based schemes employ a buffer region that should be sufficiently large to ensure bulk properties in the atomistic region [3]. In Schwarz algorithms, a boundary force is introduced, along with a specular wall, to account for the missing component of the virial pressure and to eliminate these density oscillations. In this Rapid Communication, we determine the boundary force for multiscale simulations of liquid water, by extending the control algorithm, previously developed for monatomic liquids [8] to the case of water.

We begin with a summary of the general Schwarz iteration procedure (see Fig. 4). In this approach, a solution of the continuum velocity field is obtained first subject to external (outer boundary) and internal (from the atomistic region) boundary conditions. This is followed by a solution of the atomistic scale equations, usually Newton’s equations of motion as embodied in molecular-dynamics (MD) methods. This is implemented in the following five steps. (a) The interactions between atoms are computed, including the boundary force Fm. (b) The velocity boundary condition obtained from the continuum solution is imposed on the atomic degrees of freedom. (c) The atomistic-continuum interfaces are moved with the local continuum velocity normal to the interface to allow flow inward and outward of the atomistic domain; the atoms that have crossed the interfaces are bounced and the interface positions are reset to the initial values to keep a constant frame of reference. (d) The particles that have left the atomistic domain are reinserted [11,12]. (e) A velocity boundary condition, as measured in the atomistic domain, is constructed for the continuum region and used for the next iteration. The steps are then repeated again, starting with the solution of the continuum velocity field.

In the present algorithm, the atomistic region is described by MD simulations subject to nonperiodic boundary conditions (NPBCs). The position r=(x,y,z) and velocity u=(ui,vi,wi) of the i-th particle evolve according to the Newton’s equation of motion,

\[
\frac{d}{dt} r_i = u_i(t),
\]

\[
m_i \frac{d}{dt} u_i = F_i = - \sum_{j \neq i} \nabla U(r_{ij}),
\]

where mi is the mass and Fi is the force on particle i. The interaction potential U(rij) models the physics of the system.
Here we consider liquid water modeled using the rigid simple point charge/extended water model by Berendsen et al. [13], with an O-H bond length of 1 Å and a H-O-H angle of 109.47° constrained using the SHAKE algorithm [14]. The long-range electrostatic interactions are treated using the reaction field method [15]. The nonbonded interactions are computed using a molecular cutoff \( r_c \) of 1.0 nm and the equations are integrated using a leap-frog scheme with a time step \( \delta t = 2 \) fs.

We impose NPBC with a boundary force \( F_m \) to exert the correct mean virial pressure on the MD system [8]. A spherical boundary is used to prevent molecules from leaving the atomistic domain and to impose the ideal kinetic part of the system pressure. The collisions are detected in a moving frame of reference. At the end of the time step, the moving boundaries are reset to their initial position and particles that end up outside the computational domain are then reinserted. We start by applying a zero external boundary force. The density is sampled in 3 ps time intervals and we employ a filter to reduce the signal noise. The filter reduces the required sampling size and hence improves the convergence properties.

The density \( \rho_{m}^{n} \) is measured with a spatial resolution \( (\Delta x) \) of 0.025 nm in time intervals of 3 ps and processed twice through a Gaussian filter to obtain \( \rho_{m}^{n} \):

\[
\rho_{m}^{n}(x) = \frac{1}{\epsilon^2} \int \int \left[ \rho_{m}^{n'}(x') e^{-(x-x')^2/\epsilon^2} dx' \right] e^{-(x-x')^2/\epsilon^2} dx',
\]

where \( \epsilon = 2 \Delta x \). The cutoff used for the discrete evaluation of the convolution is 3 \( \Delta x \) [9]. We then evaluate the error in the fluid density as

\[
u^{n+1/2} = \nu^{n+1/2} - 2(\nu^{n+1/2} - u_{b}),
\]

\[
\chi^{n+1} = \chi^{n} + t \nu^{n+1/2} + (\delta t - t') \nu^{n+1/2}.
\]
constant target density, and

We let

where

and amplify this with a factor

for each

boundary because the magnitude of the density disturbances

between the dipole of each water molecule with the normal to the

direction at distances from the boundary:

where

is the distance to the boundary,

is the measured filtered value.

when the external force is not acting on the system, we

observe up to 60% density fluctuations. The results of applying

the control shown in Fig. 2 demonstrate that our approach eliminates these density oscillations. The value of

in the algorithm [9]. With

, the method has converged when the

convergence to

achieving after 0.4 ns. The integral

FIG. 4. (Color online) Schematic representation of the hybrid simulation for the Couette flow, indicating the atomistic region treated with

MD, the continuum region, and the motion of the boundary far from the atomistic region (with velocity \( \pm v \)).

is reduced as the distance increases. The boundary force is

finally computed as

and applied to the center of mass of each water molecule (see

Fig. 2). We consider that the method has converged when the

root mean square of the error

is less than a prescribed value, here 1%. The controller is

continuously acting on the system and

is computed in time

intervals of 90 ps.

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FIG. 3. The probability distribution of the cosine of the angle \( \phi \)

between the dipole of each water molecule with the normal to the

x direction at distances from the boundary: (a) 0.1 nm, (b) 0.2 nm, (c)

0.4 nm, and (d) 0.6 nm.

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x direction at distances from the boundary: (a) 0.1 nm, (b) 0.2 nm, (c)

0.4 nm, and (d) 0.6 nm.
As a further test, we apply the control algorithm to the case of Couette flow. A schematic representation of the flow geometry is shown in Fig. 4. The size of the computational domain is $12 \times 3 \times 3$ nm$^3$. A resolution of $0.5 \times 3 \times 3$ nm$^3$ is used to sample the velocities that serve as a BC for the continuum solver. The flow is imposed by moving the upper wall with a velocity $v = 0.1$ nm/ps and the lower one with $-v$. In the hybrid approach, we apply the Schwarz alternating method with an overlap region of four cells (2 nm) on each side, with the continuum-atomic overlap in the regions $-3$ nm $< x < -1$ nm and $1$ nm $< x < 3$ nm. Details about the exchange of boundary conditions between the MD and the continuum region described by incompressible Navier-Stokes (NS) equations can be found in Ref. [8]. In the present case, the solution to the NS equations is a linear streamwise velocity profile. The MD subdomain in the hybrid case has the dimensions $6 \times 3 \times 3$ nm$^3$ (12 boxes in $x$ and one box in $y$ and $z$ directions). In one cycle of the hybrid algorithm, we impose the BC from the continuum to the MD and subsequently sample the velocities for 80 ps to extract the BC for the continuum. We sample the results for 1 ns and show the velocity profiles obtained from the continuum and hybrid simulations in Fig. 5; the two sets of values are in excellent agreement.

In conclusion, we have presented a control algorithm to eliminate density fluctuations in the coupling of atomistic models with continuum descriptions of liquid water. A dynamic controller based on the errors measured in the local fluid density (that may reach up to 60%) provides an appropriate boundary forcing which applies the correct virial pressure to the system. The algorithm was applied to water at rest, in which case it eliminates the density oscillations, and to Couette flow, in which case it recovers the linear profile of the velocity field. Ongoing work aims to develop controllers that can eliminate the spurious orientation of the water molecules near the atomistic-continuum interface and the extension of the coupling to fully three-dimensional configurations.