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HETEROGENEOUS ATOMISTIC-CONTINUUM REPRESENTATIONS FOR DENSE FLUID SYSTEMS*

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We present a formulation and numerical solution procedure for heterogeneous atomistic-continuum representations of fluid flows. The ingredients from atomistic and continuum perspectives are non-equilibrium molecular dynamics and spectral element, respectively; the matching is provided by a classical procedure, the Schwarz alternating method with overlapping subdomains. The technique is applied to microscale flow of a dense fluid (supercritical argon) in a complex two-dimensional channel.

Keywords: Schwarz; Matching; Molecular Dynamics; Hybrid Continuum-Molecular Dynamics.

1. Introduction

Many important problems in materials processing involve a range of scales from the atomistic to the continuum. Purely atomistic representations are clearly computationally intractable because of the number of particles required to represent macroscale phenomena[1]; conversely, purely continuum approaches often neglect certain critical microscale phenomena that do not now, and many never, admit adequate macroscale constitutive characterization. New integrative frameworks are therefore required, not only of the well-established atomistic-*then*-continuum variety, but also of the atomistic-*with*-continuum variety. Our interest is in the latter, in particular, in gas-*liquid*-solid systems subject to the full range of mechanical, thermal, and, ultimately, chemical interactions.

Heterogeneous atomistic-continuum representations have been successfully developed for problems in both solid mechanics and fluid mechanics. In fluid mechanics, several domain decomposition approaches have been proposed and implemented.[2] Most methods developed to date are, however, restricted to gas

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flows, in that both the particle treatment (e.g., Direct Simulation Monte Carlo techniques, discretizations of the Boltzmann equation) and the interface treatment (e.g., variations of the Marshak condition) are directly applicable only to dilute systems.[3–5] Although these concepts can perhaps be extended to dense fluids, we have chosen a slightly different strategy that does not require explicit calculation of fluxes. Our approach is, in fact, more closely related to heterogeneous representations of solid mechanics problems,[6,7] such as the study by Kohlhoff *et al.*,[6] and to a recently published fluid mechanical study by O'Connell and Thompson.[8]

In the formulation proposed by Kohlhoff *et al.*, the physical domain is decomposed into two overlapping regions, or equivalently, three regions: an atomistic domain treated by molecular dynamics (MD), a continuum domain treated by finite elements, and an atomistic-continuum overlap region. Displacement boundary conditions in the overlap region provide globally consistent stress fields — presuming that the atomistic description is consistent with the continuum constitutive model of the finite element analysis. For this effectively zero-temperature structural problem, in which the atoms are bound to lattice sites, displacements can be matched on an individual atom-to-finite element node basis; in contrast, for fluid systems, or fluid-solid systems, thermal considerations and material motion preclude such a Lagrangian identification, and an appropriate Eulerian generalization is required.

The recent work of O'Connell and Thompson[8] addresses the problem of material motion by introducing an "Eulerian" overlap region. An important conclusion of Ref. 8 is that stress continuity can be achieved *without* explicitly matching (or imposing) fluxes. Our method also exploits this result, and also employs an overlap region. However there are several important differences between the two approaches. First, O'Connell and Thompson invoke constrained Lagrangian dynamics in the overlap region; we find that a simpler "Maxwell Demon" leads to slightly better results. Second, the technique of O'Connell and Thompson, as demonstrated in Ref. 8, is one-dimensional. Many new issues arise in higher space dimensions, in particular when there is mean (continuum) flow across the atomistic-continuum boundary; we treat these issues by the introduction of a particle reservoir.

Third, and most importantly, as reported in Ref. 8, the algorithm of O'Connell and Thompson matches the atomistic and continuum representations on a time step-by-time step basis, with the time step imposed by the molecular dynamics representation. This necessitates many continuum evaluations that, in higher dimensions, and in particular for flows that evolve over continuum timescales, will result in prohibitive computational costs. In some sense, the O'Connell and Thompson technique decouples lengthscales, but not timescales; our approach does decouple timescales, requiring only $O(1)$ (in practice, $O(10)$) continuum solutions in order to achieve a steady state. At present our technique is only appropriate for steady problems, although we believe unsteady problems evolving over continuum timescales can also be treated.

Finally, we remark that, although the O'Connell and Thompson technique permits analysis of detailed atomistic-scale temporal evolution, adequate statistics require averages over sufficiently long timescales, and thus accuracy and

resolution are coupled. Specifically, the results of Ref. 8 suggest that only timescales which correspond to quasi-static evolution on the atomistic scale can be accurately resolved. This is consistent with linear response theory,[9] according to which a system "makes use" of fluctuations about a quasi-equilibrium state to alter this state in response to a forcing input.

In Sec. 2 we present the heterogeneous algorithm, and describe the matching method and the imposition of MD boundary conditions; in Sec. 3, we present an illustrative application.

2. Heterogeneous Algorithm

We describe an example of the formulation and numerical solution of a heterogeneous atomistic-continuum representation of a dense fluid system: flow in an "obstructed" channel. It should be clear from the presentation, however, that the approach is quite general. The ingredients are, from the atomistic side, non-equilibrium molecular dynamics,[10] and from the continuum side, spectral element solution[11]; the matching is provided by a classical procedure — recently resurrected in the domain decomposition framework[12] — the Schwarz alternating method with overlapping subdomains (Sec. 2.1). A critical prerequisite is the ability to impose arbitrary boundary conditions on a molecular dynamics domain (Sec. 2.2).

Unless otherwise stated, all quantities will be expressed in reduced units using $\sigma = \sigma_{Ar} = 3.4$ Å for length, $m_{Ar} = 40$ amu for mass, $\varepsilon_{Ar}/k_b = 119.8$ K for temperature, and $\tau = (m_{Ar}\sigma_{Ar}^2/48\varepsilon_{Ar})^{1/2} = 3.112e(-13)s$ for time. Here σ_{Ar} and ε_{Ar} are the parameters of the Lennard–Jones (LJ) potential for argon,[10] m_{Ar} is the mass of the argon atom, and τ is the characteristic time for argon.

2.1. *Schwarz coupling approach*

The matching method is described here for the case of a one-dimensional problem; extension to higher dimensions directly follows. Given a problem to be solved by continuum techniques for $0 < x < b$ and molecular techniques for $b < x < L$, we define the continuum subdomain as $0 < x < b$ and the molecular subdomain as $a < x < L$, where $a < b$; $a \leq x \leq b$ is the overlap region where both models are presumed valid. Given boundary conditions at $x = 0$ and $x = L$, a solution is first assumed in one of the subdomains, say, the molecular. This solution at $x = b$ ($\in [a, L]$) serves as a boundary condition on the continuum subdomain, permitting calculation of the "first continuum iterate." This first continuum iterate, in turn, provides a boundary condition at $x = a$ for the molecular dynamics (MD) simulation. Finally, the resulting "first MD iterate" yields a new boundary condition at $x = b$ for the continuum subdomain. This alternating procedure is then repeated until convergence.

Convergence can be proven for a large class of problems when both subdomains are treated by continuum methods.[12] Convergence should, thus, also be obtained for a heterogeneous description if the continuum and molecular models are equivalent in the overlap region. However, additional "steady-state" errors will arise in the

heterogeneous case due to, firstly, discrepancies in the transport coefficients in the two subdomains, secondly, noise introduced by MD statistical fluctuations, and thirdly, smoothing of the MD data prior to imposition of the boundary conditions on the continuum iterate. The convergence *rate* may also be affected by these new sources of error.

This overlapping Schwarz method is chosen because it avoids the use, and hence imposition, of fluxes in the matching of solutions on different subdomains. Assuming that the transport coefficients are correctly matched, flux (e.g., stress) continuity is automatically ensured by the agreement in the overlap region of the converged continuum and atomistic representations. The method is also advantageous because it is "implicit," and thus decouples not only lengthscales, but also *timescales*: only steady-state MD simulations are required. Extension of the method to treat time-varying problems on the continuum timescale is thus possible, in which at *each time* t^n the (say, implicitly) integrated continuum iterate is matched to the previous MD iterate at t^n; given the macroscale timesteps and nanoscale MD domain, the MD calculations may be treated as quasi-static, thus avoiding expensive *coupled* time integration at the molecular timescales.

2.2. *Imposition of MD boundary conditions*

A prerequisite for our approach is a method for imposing general velocity and temperature boundary conditions on an MD simulation in a *general* geometry $\mathcal{D}$ with associated boundary $\partial\mathcal{D}$, as shown in Fig. 1. Note that for the purposes of this paper we shall consider MD regions in the *interior* of the flow, and thus all boundary conditions on the MD domain are of the artificial variety, that is, required solely for purposes of heterogeneous representation. We thus avoid the issue of molecular walls[1] which, although important, is a purely atomistic issue, and thus separate from the primary "matching" concerns of the current paper. We assume that the

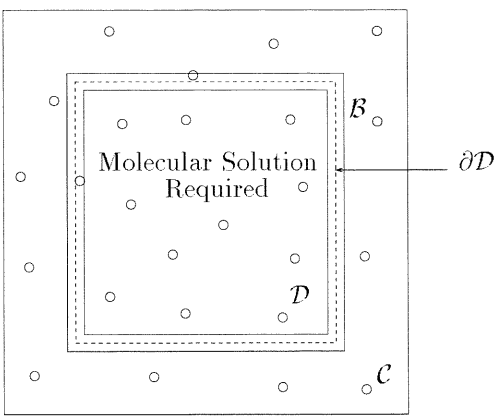

Fig. 1. Velocity and temperature boundary condition imposition.

velocity and temperature, as well as the gradient of the velocity and temperature, are known on $\partial\mathcal{D}$, and that we desire the steady-state MD solution everywhere inside $\mathcal{D}$.

The boundary conditions are imposed through the artifice of "reservoirs" of fluid that encompass the domain $\mathcal{D}$. In particular, we first introduce a region, or sheath, $\mathcal{B}$, of small but finite thickness δ, that surrounds $\partial\mathcal{D}$. While in residence in $\mathcal{B}$, particles are given, at each time step, a velocity drawn from a Maxwellian distribution with mean and variance consistent with the desired velocity and temperature of the fluid; the latter are constructed from a first order Taylor series expansion based on the known values of the velocity, temperature and velocity and temperature (normal) gradients on $\partial\mathcal{D}$. (We prefer the Taylor series approach over pointwise matching everywhere in $\mathcal{B}$ because the former is much less cumbersome and storage-intensive. For our Taylor series approach to be valid, δ must be small compared to the lengthscale over which the continuum solution varies appreciably.) Finally, we provide a continuous supply of molecules to and from $\mathcal{D}$ by enclosing $\mathcal{D} \cup \mathcal{B}$ in a larger, periodic domain $\mathcal{C}$; $\mathcal{C} \setminus (\mathcal{D} \cup \mathcal{B})$ acts as particle reservoir that, for incompressible problems, ensures that there is no *net* mass flow into $\mathcal{D}$. (Note $\mathcal{C}\setminus(\mathcal{D}\cup\mathcal{B})$ refers to $\mathcal{C}$ with $\mathcal{D}\cup\mathcal{B}$ removed.) Extension to substantially compressible systems has not yet been considered.

The errors introduced by this method of imposing boundary conditions originate in the not strictly correct dynamical state of the particles in $\mathcal{C}\setminus\mathcal{D}$, in particular $\mathcal{C} \setminus (\mathcal{D} \cup \mathcal{B})$, with which particles in $\mathcal{D}$ interact. The incorporation of information about the *gradient* of the required boundary conditions in $\mathcal{B}$ — not just the function values, as one might anticipate from the continuum case — addresses, and largely alleviates, this problem. If necessary, the width of $\mathcal{B}$, δ, can be chosen such that $\delta > \ell_c$, thus at least shielding particles in $\mathcal{D}$ from the reservoir $\mathcal{C}\setminus(\mathcal{B}\cup\mathcal{D})$; here, ℓ_c is the Lennard–Jones interaction-potential cut-off. The Maxwellian equilibrium distributions assumed in $\mathcal{B}$ constitute another source of error; this error will be small if the timescale, or equivalently lengthscale, over which the particle distribution functions relax to the correct non-equilibrium forms is small compared to the size of $\mathcal{D}$. Non-equilibrium simulations performed by the authors have confirmed earlier findings[13] that bulk equilibrium *and* non-equilibrium properties are, indeed, unaffected by Maxwellian distribution-based boundary conditions, the effects of which remain localized near the simulation boundary. Although distribution functions for dense fluids in non-equilibrium flows could be used,[14] the latter are not too well characterized, and thus empirical constructions would be required.

Several alternatives to the above "Maxwell Demon" (in fact a "two-sided" fair demon) method are possible. For example, we have also considered a method by which $\mathcal{B}$ is subdivided into smaller regions along $\partial\mathcal{D}$ in which the particles are continuously "rescaled" to the desired local mean velocity and temperature. In the latter rescaling process, all particles receive a velocity increment such that their mean velocity matches the desired value, and all velocity fluctuations about this mean are scaled such that the temperature of the subregion matches the requisite

boundary condition. This strategy is very similar to the constrained Langrangian dynamics technique of Ref. 8. We find that the "Maxwell Demon" approach, though less sophisticated, typically performs better than these more subtle constraints on particle motion. To our knowledge, neither method is proven to reproduce the correct physical dynamics,[15] and hence both will no doubt introduce local errors in the non-equilibrium distribution functions.

3. Validation

3.1. *Model problem statement*

In this section we describe the model problem with which we validate our techniques for imposing MD boundary conditions and for matching heterogeneous representations. The problem is flow in the "obstructed" channel domain Ω, a sketch of which is shown in Fig. 2, with parabolic inflow velocity profile, no stress ("outflow") boundary conditions downstream, and no-slip velocity conditions at all solid walls. The channel length has $L = 300$ Å and height $H = 150$ Å. The flow is "obstructed" by a square block with side $l_b = 30$ Å residing in the middle of the channel.

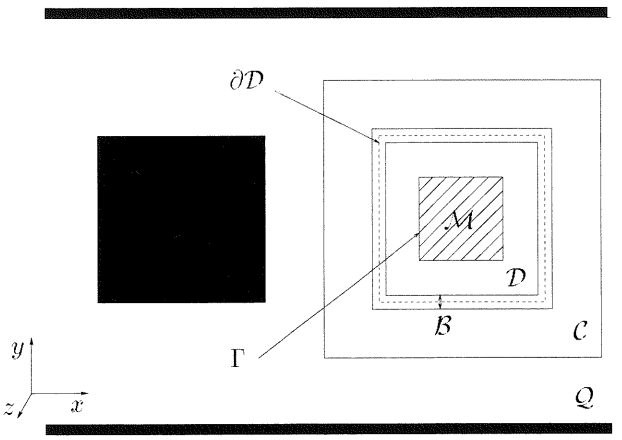

Fig. 2. Heterogeneous calculation domain decomposition (not to scale). The actual ratio of the volume of $\mathcal{D}$ to the volume of the entire domain $\mathcal{Q}$ is 1/20, and in practice could be much smaller.

A fully continuum spectral element discretization of the incompressible Navier–Stokes equations serves as the reference exact solution. The Reynolds number $R = \rho\overline{U}H/\mu$ is 3.7 based on the channel height and imposed mean flow velocity $\overline{U} = 52.1$ m/s. The viscosity $\mu = 3e-4$ Kg/ms of argon at the specified temperature $T = 110$ K and density $\rho = 1420$ Kg/m^3 is known experimentally,[16] and independently confirmed by the non-equilibrium MD simulations of the authors and others.[13] We assume on the continuum side that the flow is incompressible, isothermal, and has constant-property; we can make no such "assumptions" on the molecular side, as described below.

The region of the flow is represented by the MD calculation where $\mathcal{D}$ is the wake region behind the square block in the middle of the channel, as shown in Fig. 2; $\mathcal{D}$ measures $(\ell_{\mathcal{D}} \times \ell_{\mathcal{D}} \times 8.82\sigma)$ with $\ell_{\mathcal{D}} = 8.82\sigma = 30$ Å. This MD domain is chosen because the "wake" region behind the block is strongly nonparallel, and thus serves as a good test for the method, yet is far enough from all walls that continuum behavior should obtain. The latter, of course, does not make for an interesting "real" application, but does allow for a controlled study of the approach. In a real application, the MD simulation would be applied in a region $\mathcal{D}$ in the interior of which the continuum description is presumably not adequate.

The actual MD calculation is performed on a box $\mathcal{D}'$, *dynamically similar* to $\mathcal{D}$, which measures $\ell'_{\mathcal{D}'} \times \ell'_{\mathcal{D}'} \times 8.82$, with $\ell'_{\mathcal{D}'} = 11.47\sigma = 39$ Å. Denoting the (say maximum streamwise) velocity on $\partial\mathcal{D}$ dictated by the continuum as $u_{\mathcal{D}}$, we impose on $\partial\mathcal{D}'$ the scaled velocity $u'_{\mathcal{D}'} = (\ell_{\mathcal{D}}/\ell'_{\mathcal{D}'})u_{\mathcal{D}}$. Since we specify the same material $(\varepsilon_{Ar}, \sigma_{Ar})$ and thermodynamic state $(T' = T, \rho' = \rho)$ in both $\mathcal{D}$ and $\mathcal{D}'$, $\mu' = \mu$, and it then follows that, in the incompressible limit, the flows in $\mathcal{D}$ and $\mathcal{D}'$ are dynamically similar. Note we choose $\ell'_{\mathcal{D}'} > \ell_{\mathcal{D}}$ and hence $u'_{\mathcal{D}'} < u_{\mathcal{D}}$ to reduce compressibility and temperature variation due to viscous heating, as both these effects are absent in our continuum model. However, we cannot choose $\ell'_{\mathcal{D}'}$ to be too large or $u'_{\mathcal{D}'}$ too small as the number of molecules will increase prohibitively, and the signal-to-noise ratio will degrade, respectively. To complete the specification of the MD problem, we choose $\delta' = 0.88\sigma$ for the thickness of $\mathcal{B}'$, and a box of extent $(20.3\sigma \times 20.3\sigma \times 8.82\sigma)$ for $\mathcal{C}'$. The interaction potential cutoff is set to the conservative value $\ell'_c = 3\sigma$. Note that we shall present quantities in "original" units, that is, we rescale all MD results obtained on $\mathcal{D}'$ back to $\mathcal{D}$ prior to presentation.

Figure 2 shows the domain of the continuum part of the calculation, $\mathcal{Q} = \Omega \backslash \mathcal{M}$, which is everything *outside* the "MD-only" hatched region $\mathcal{M}$; the MD region of the calculation, $\mathcal{D}$, and associated reservoirs $\mathcal{B}$ and $\mathcal{C}$; and the overlap region, $\mathcal{Q} \cap \mathcal{D}$. Recall that we piece together the solution as continuum in $\Omega \backslash \mathcal{D}$, molecular in $\mathcal{M}$, and continuum *or* molecular in $\mathcal{Q} \cap \mathcal{D}$. The boundary conditions on the continuum spectral element incompressible Navier–Stokes calculation are known on all portions of $\partial\mathcal{Q}$ save $\Gamma = \partial\mathcal{M}$, where the conditions are provided by the MD calculation as part of the Schwarz procedure. Reciprocally, the MD domain obtains boundary conditions for $\partial\mathcal{D}$ from the continuum solution on $\mathcal{B} \subset \mathcal{Q}$.

The width of the overlap region $\mathcal{Q} \cap \mathcal{D}$ in these calculations is 2.94σ. The thickness of the overlap region significantly affects the convergence rate, and must be some nonnegligible fraction of the extent of $\mathcal{D}$ to ensure rapid convergence. In practice, the overlap region should not be taken as too large, since the number of molecules increases rapidly; for the domain chosen here, there are 245 molecules in $\mathcal{M}$, 980 molecules in all of $\mathcal{D}$, and 3060 molecules in all of $\mathcal{C}$. In real applications, the MD-only region $\mathcal{M}$ will be selected first, with $\mathcal{D}$ (and hence the overlap region), $\mathcal{B}$, and $\mathcal{C}$ then determined by numerical considerations.

Although the continuum calculation is two-dimensional, the MD simulation is three-dimensional. However we consider Γ to be effectively defined in the (x, y)

plane, since only z-averaged information passes between the two representations. Note that the MD-derived field variables on Γ are smoothed — fitted by a low order (here third order) polynomial — prior to imposition on the subsequent continuum iterate. To ensure that the continuum boundary data satisfies zero-mass-flux through Γ, the MD smoothed data on Γ is further projected onto a zero mass flux field by subtracting $\oint_\Gamma \mathbf{u} \cdot \hat{n} dl / \oint_\Gamma dl$ from the normal velocity on Γ, where $\mathbf{u}$ and $\hat{n}$ are the velocity and outward normal on Γ, and dl is a differential line element.

3.2. *Results*

3.2.1. *Boundary condition imposition*

To validate the method of imposing boundary conditions described in Sec. 2.2, the exact (spectral element) solution for our model problem is used on $\partial\mathcal{D}$ as the boundary condition for an MD simulation, and the resulting MD predictions in the interior of $\mathcal{D}$ are then compared to the reference fully continuum solution.

Figure 3 shows good agreement between the reference solution and the MD results, even well away from the region $\mathcal{B}$ in which the MD simulation receives boundary data. The MD simulations are integrated to a statistical accuracy of approximately ± 3 m/s; averaging here, and in the Schwarz iteration of the next section, is performed over $(1.15\sigma \times 1.15\sigma \times 8.82\sigma)$ pencils in space, and 4800τ intervals in time, after reaching a stationary state in approximately 1000τ.

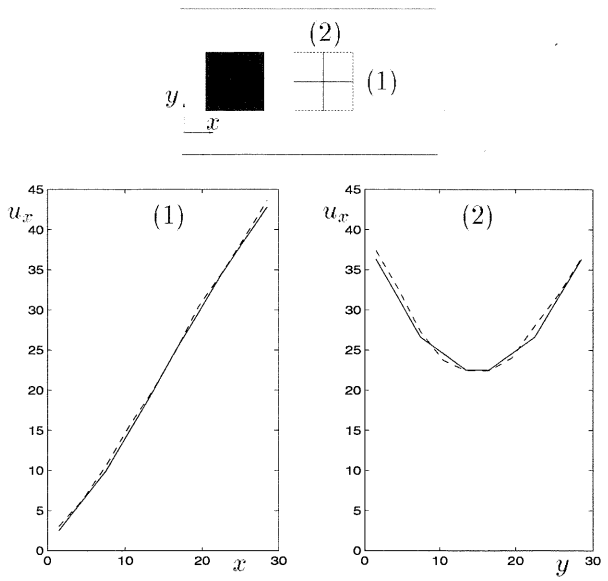

Fig. 3. Comparison between fully continuum (solid) and MD (dashed) solutions in the interior of $\mathcal{D}$. The velocities are in m/s and the lengths x, y in Å. The graphs correspond to the constant-y (1) and constant-x (2) slices indicated above.

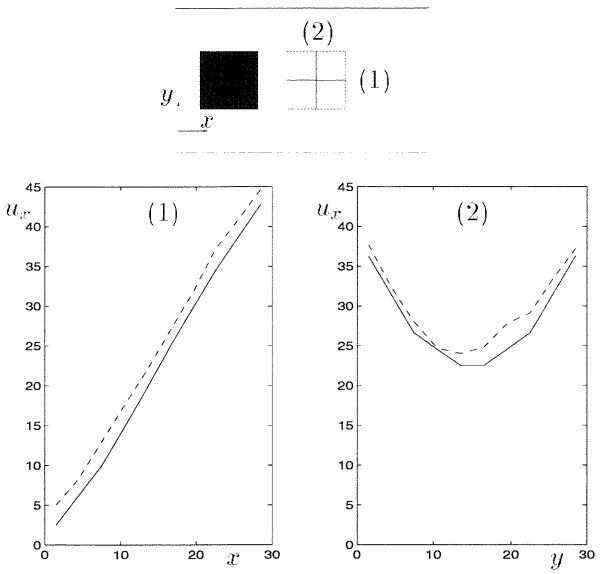

Fig. 4. Comparison between fully continuum (solid) and heterogeneous (dashed) Schwarz solutions in the interior of $\mathcal{D}$. The velocities are in m/s and the lengths x, xy in Å. The graphs correspond to the constant-y (1) and constant-x (2) slices indicated above.

3.2.2. *Heterogeneous calculation*

The Schwarz iteration is initiated by assuming a zero (mean velocity) solution in $\mathcal{D}$, and hence zero velocity boundary conditions on Γ. Each full iteration requires, on an HP 735 workstation, 5 minutes for the 3500 degree-of-freedom steady spectral element solution and 18 hours for the 3060-atom MD simulation. (For this "experimental study," we choose the interaction potential cut-off, overlap region, particle reservoir $\mathcal{C}$, and number of time steps very conservatively; more practical choices for these parameters would reduce the MD simulation time to roughly 10 hours/iteration.) Note, however, that one time step of the 180000-time step MD simulation requires only 0.5 seconds, and thus a technique that demanded continuum evaluation on the MD-timescale would be enormously expensive — a 10-year computation. (For such a small imposed time step, we could consider a less expensive fully explicit compressible continuum simulation, however the computational cost would still be formidable.) Needless to say, even our approach is not yet particularly efficient given the intrinsic profligacy of the MD computations.

Iterative convergence is obtained in approximately 20 iterations. The accuracy achieved, although acceptable, is reduced with respect to that obtained in Sec. 3.2.1 (see Fig. 4); we attribute this degradation to the response (amplification in this case) of the Schwarz method to the MD statistical fluctuations, the difference in transport coefficients in the two models, and the error introduced by the MD boundary condition imposition. We do not yet have a complete understanding of this

amplification, nor proposals to improve accuracy. Nevertheless, the results indicate that our approach to heterogeneous representations can, indeed, capture the dynamics of "complex" multidimensional flows, and at a cost which scales as the cost (or $O(10)$ the cost) of the constituent MD computation.

Acknowledgments

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