Nearly Exact Solution for Coupled Continuum/MD Fluid Simulation

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Abstract. A general statistical approach is described to couple the continuum with molecular dynamics in fluid simulation. Arbitrary thermodynamic field boundary conditions can be imposed on an MD system while minimally disturbing the particle dynamics of the system. And by acting away from the region of interest through a feedback control mechanism, across a buffer zone where the disturbed dynamics are allowed to relax, we can eliminate that disturbance entirely. The field estimator, based on maximum likelihood inference, serves as the detector of the control loop, which infers smooth instantaneous fields from the particle data. The optimal particle controller, defined by an implicit relation, can be proved mathematically to give the correct distribution with least disturbance to the dynamics. A control algorithm monitors the estimated current fields with the desired fields at the boundary and modifies the behavior of the particle controller far way, until they eventually agree. This method, combined with a continuum code in a Schwarz iterative domain-decomposition formalism, provides mutually consistent solution for steady-state problems, as particles in the MD region of interest has no way to tell any difference from reality. Finally, we explain the importance of incorporating a higher order single-particle distribution function in the sense of Chapman-Enskog development for shear flow.

1. Introduction

Molecular dynamics (MD) plays a unique role in the simulation of fluids by virtue of its ability to offer insights into atomic-level structure and dynamics that cannot be obtained from continuum calculations. Because only a microscopic region of the fluid can be studied in this manner, there is considerable interest to develop hybrid atomistic-continuum methods. Though this problem has been well recognized [1], there still appears to be no completely satisfactory solution.

Two notable attempts have been made recently to rectify this situation, both invoking the use of an overlapping region but differing in how the molecular and continuum descriptions are to be made compatible. O'Connell and Thompson [2] proposed to constrain the dynamics of atoms in the hybrid layer between the MD and continuum regions to ensure continuity of property averages across the coupling region. Hadjiconstantinou and Patera [3] cast their formulation in the framework of Alternating Schwarz method [4, 5] and treated the matching in terms

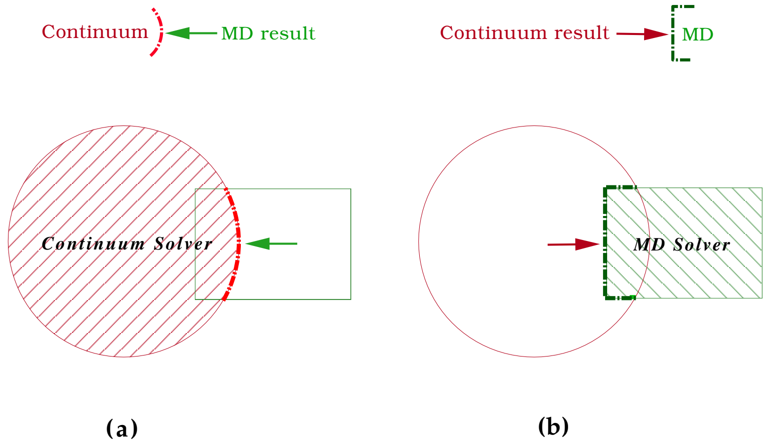

Figure 1. Alternating Schwarz formalism to get joint solution in two subdomains handled by different solvers. The atomistic subdomain will be treated by MD simulation, while the other subdomain will be treated by an appropriate continuum solver. In the iteration, two kinds of boundary condition (BC) coupling methods are needed: (a) MD result $\rightarrow$ continuum BC, (b) continuum result $\rightarrow$ MD BC.

of refining the boundary conditions imposed on each of the overlapping subdomains through an iterative process (see Fig. 1).

Our formalism [6, 7] is also based on the Alternating Schwarz method, but paying more attention to the microscopic physics. In order to iterate between continuum and MD solutions, one needs (a) to infer the macroscopic fields that accurately represent the particle result in an MD simulation, to be plugged into a continuum solver as BC, and (b) to perform the inverse, i.e., make sure that particles in an MD simulation do correspond to a set of prescribed macroscopic field(s) at the boundary, and that is achieved at a cost of as little artificial disturbance to the particle dynamics as possible.

A method for (a) has been developed in our first paper [6] in the form of an algorithm called the Thermodynamic Field Estimator (TFE). A method for (b) was briefly described in our second paper [7], and will be reviewed in sections 2 and 3. It is composed of two stages. In the first stage, we define a particle velocity transformation called the Optimal Particle Controller (OPC), to be imposed on the boundary of the MD sub-domain, $\partial\mathcal{C}$, in order to achieve the desired field BC on $\partial\mathcal{C}$. We regard this particular transformation as optimal in the sense that the resulting artificial disturbance to the particle dynamics, as measured in terms of the squared difference in the particle velocities before and after the transformation, is minimal. In the second stage, we say that if one is willing to pay an extra price computationally and not directly act on $\partial\mathcal{C}$ itself, but on an action region $\mathcal{A}$ separated from $\partial\mathcal{C}$ by a buffer zone $\mathcal{B}$, then the particle dynamics will be fully restored at $\partial\mathcal{C}$. Meanwhile one can still impose the desired BC on $\partial\mathcal{C}$ using a feedback-control mechanism, by modifying the actions in $\mathcal{A}$. This is what we call the Extended Boundary Condition (EBC), which we believe gives exact solution to the problem of (b). Combining (a) and (b) in the Alternating Schwarz formalism (Fig.1), one could obtain exact unified solutions for most steady-state fluid problems.

In section 4, we explain the importance of adopting a more accurate single-particle distribution function, other than the lowest-order, local Maxwellian distribution, that is especially critical when direct OPC (stage one) method is used to impose the BC in (b). By looking at the Chapman-Enskog development in kinetic theory, we work out an approximate solution to solve the most outstanding problems.

2. Optimal Particle Controller (OPC)

The theoretical challenge to linking continuum with MD is that the two descriptions deal with different degrees of freedom. The continuum description deals with fields such as the density field $\rho(\mathbf{x})$, velocity field $\bar{\mathbf{v}}(\mathbf{x})$ and the temperature field $T(\mathbf{x})$, while MD deals with discrete particles, their positions $\mathbf{x}_i$ and velocities $\mathbf{v}_i$. And they are also different in evolution equations: the fields evolve by a set of partial differential equations like the Navier-Stokes equation, while the particles evolve by many-body Newton's equation. The bridge linking the two, we think [3, 6, 7], should be the single-particle distribution function

$$\begin{aligned} dP &= f\left(\mathbf{x},\mathbf{v}|\rho(\mathbf{x}),T(\mathbf{x}),\bar{\mathbf{v}}(\mathbf{x})\right)d\mathbf{x}d\mathbf{v} \\ &\approx \frac{\rho(\mathbf{x})d\mathbf{x}}{(2\pi T(\mathbf{x}))^{3/2}}\exp\left(-\frac{|\mathbf{v}-\bar{\mathbf{v}}(\mathbf{x})|^2}{2T(\mathbf{x})}\right)d\mathbf{v}+f^{(2)}d\mathbf{x}d\mathbf{v}, \end{aligned} \quad (1)$$

a probability distribution in $\{\mathbf{x}_i,\mathbf{v}_i\}$ parameterized by the smooth fields $\rho(\mathbf{x}), \bar{\mathbf{v}}(\mathbf{x}), T(\mathbf{x})$. $f^{(2)}$ denotes the second-order correction to the leading-order local Maxwellian distribution when the fields have gradients. For a coupling scheme to work, (1) must be true in the overlap region, i.e., $\rho(\mathbf{x})$, $\bar{\mathbf{v}}(\mathbf{x})$, $T(\mathbf{x})$ must be well-defined in the sense of (1) and having small gradients, because otherwise it makes no sense to couple in the first place - both solvers in (Fig.1) should be valid in the overlap region, and the Navier-Stokes equation can be *derived* from the particle description in the Chapman-Enskog development [9] assuming (1).

In a previous paper [6] we have shown how to infer the macroscopic fields in an MD region of interest $\mathcal{C}$ from the current particle data, using a technique called thermodynamic field estimator (TFE). Now,

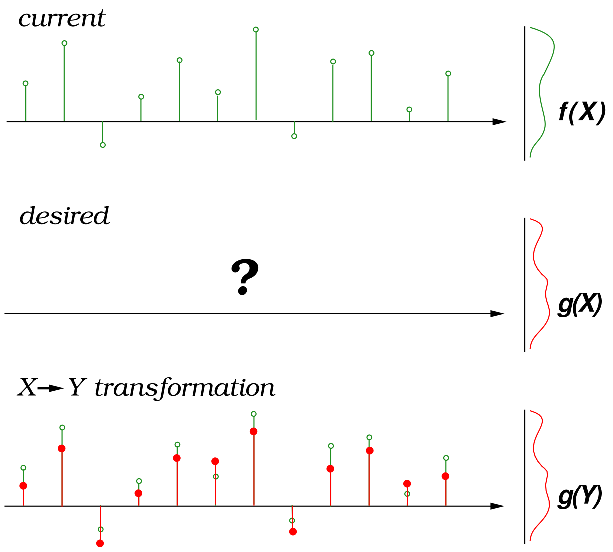

Figure 2. Given that an incoming sequence $\{X\}$ (circles) satisfy a *current* distribution $f(X)$, but one would like to change $\{X\}$ to $\{Y\}$ (dots) such that Y satisfy the *desired* distribution $g(Y)$, what $\mathcal{T}: X \to Y$ transformation should one use? Note that the average difference between circles and dots represent the magnitude of the artificial action one applies onto the atoms.

suppose one has inferred the *current* fields to be $\rho'(\mathbf{x})$, $T'(\mathbf{x})$, $\bar{\mathbf{v}}'(\mathbf{x})$ on $\partial\mathcal{C}$ using TFE, but actually wants the fields to be $\rho(\mathbf{x})$, $T(\mathbf{x})$, $\bar{\mathbf{v}}(\mathbf{x})$, how should one modify the particle coordinates such that the desired distribution is achieved? That is, say there is a random variable sequence $\{X_n\}$ satisfying distribution $f(X)$, but we want them to satisfy another distribution g, so we begin to replace X_n's by Y_n's such that $\{Y_n\}$ will satisfy $g(Y)$, what should be the *optimal* $\mathcal{T}: X_n \to Y_n$ transformation?

We propose the criterion for optimality to be the minimization of

$$B = \sum_n |\mathbf{v}_n^{\mathrm{aft}} - \mathbf{v}_n^{\mathrm{bef}}|^2, \tag{2}$$

where $\mathbf{v}_n^{\mathrm{bef}}$ and $\mathbf{v}_n^{\mathrm{aft}}$ are the velocities of the n^{th} particle before and after the $\mathcal{T}$ transformation, on the constraint that $\mathbf{v}_n^{\mathrm{aft}}$ now satisfy the desired single-particle distribution described by fields $\rho(\mathbf{x})$, $T(\mathbf{x})$, $\bar{\mathbf{v}}(\mathbf{x})$. $\mathcal{T}$ is then called the Optimal Particle Controller (OPC). One may choose to operate $\mathcal{T}$ in a finite-volume region outside of $\mathcal{C}$, or just on a certain boundary such as $\partial\mathcal{C}$, which are called bulk or boundary OPC, respectively. The subtlety of boundary OPC involving conditional probability is already discussed in a previous paper [7].

We believe that, in general, a unique OPC exists for a given problem. Especially, we show that for one- or decoupled multi-dimensional (factorizable) distributions, the following transformation is OPC,

$$X \to Y : \int_{-\infty}^{X} f(\xi) d\xi = \int_{-\infty}^{Y} g(\xi) d\xi, \tag{3}$$

where one solves the implicit equation for Y, given each X. It can be checked that Y indeed conforms to distribution g if the input random variable X conforms to f. The proof that (3) is OPC in 1D with discussions on general 3D boundary OPC is to be given in our next paper [8].

3. Extended Boundary Condition

In section 2 a method was proposed to impose the field boundary condition in an MD simulation that results in the least disturbance to the particle dynamics in the sense of (2). Nevertheless the disturbance still exists for particles in the skin region near the boundary, which can be shown to be proportional to the rate of dissipation in the system. In this section we will formulate a procedure which eliminates that disturbance entirely at the specific boundary and in the interior, thus providing an *exact* solution to the problem. Since any artificial action necessarily alters the particle dynamics in the vicinity where it is imposed, the best one can do is to act some distance away from the intended boundary and cause the macroscopic field at the boundary to be what is prescribed.

This can be done using a three-region approach which we will call the Extended Boundary Condition (EBC), shown in Fig.3, and through a feedback control mechanism. The physical region of interest, $\mathcal{C}$ (core), is surrounded by a buffer zone $\mathcal{B}$. Artificial actions are applied on an outer MD region $\mathcal{A}$ which is sufficiently separated from $\mathcal{C}$, its aim being to induce the prescribed fields on the core boundary $\partial\mathcal{C}$. Due to molecular chaos in fluids, short wavelength perturbations on particle dynamics will decay over a distance of a few collisional mean free paths, thus setting a lower limit for $\mathcal{A}$-$\mathcal{C}$ separation; only the hydrodynamic variables which correspond to the three collisional invariants will still have influence on $\partial\mathcal{C}$. The action in $\mathcal{A}$ is in general a matter of choice, but here we will use the boundary OPC introduced in section 2 because it minimizes the artificial disturbance, thus $\mathcal{B}$ could be as thin as possible. The thermodynamic field estimator (TFE) serves as the detector of the control loop, inferring current fields on $\partial\mathcal{C}$ based on particle data from the entire $\mathcal{C}$ region. A control algorithm compares the inferred current field with the prescribed field on $\partial\mathcal{C}$, and gives instructions to the particle controller.

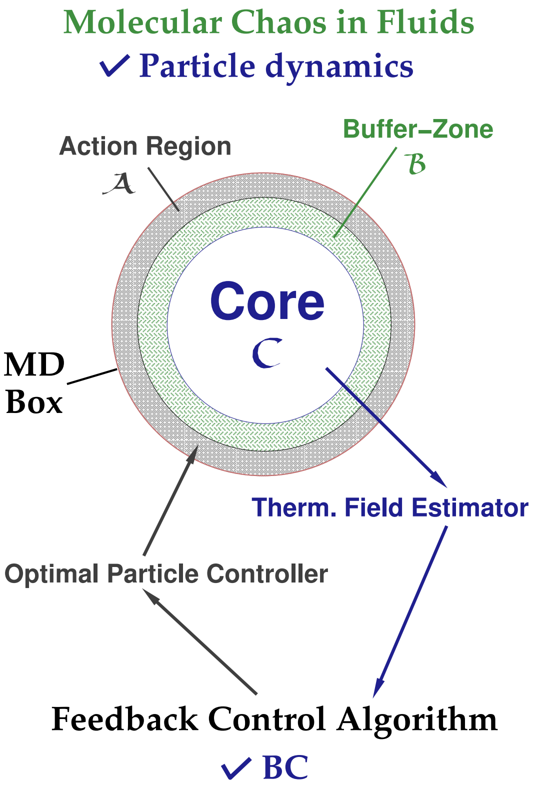

Figure 3. Schematic diagram of the Extended Boundary Condition (EBC), which incorporates a field estimator, a particle controller and a feedback control algorithm.

Note that although $\mathcal{A}$ and $\mathcal{B}$ are discrete-particle regions governed by molecular dynamics, as the core $\mathcal{C}$, they are not real, physical domains that appear in the Schwarz domain decomposition diagram in Fig. 1. They are only numerical constructs in the MD sub-step of the iteration, merely to ensure that the physical region of interest, $\mathcal{C}$, has the correct field boundary conditions and evolves according to natural particle dynamics. Properly implemented, it is a powerful tool for studying fluid systems because a fluid atom in $\mathcal{C}$ has no way to tell *any* difference from reality.

Compared with directly applying OPC to $\partial\mathcal{C}$ without a buffer zone, EBC has two advantages: 1. It practically generates no disturbance to the core region. 2. We do not need to worry about $f^{(2)}$ or higher-order terms, which will be generated automatically at $\partial\mathcal{C}$. The drawback is that for general BC, multi-variable feedback control is needed, which is quite a challenging task.

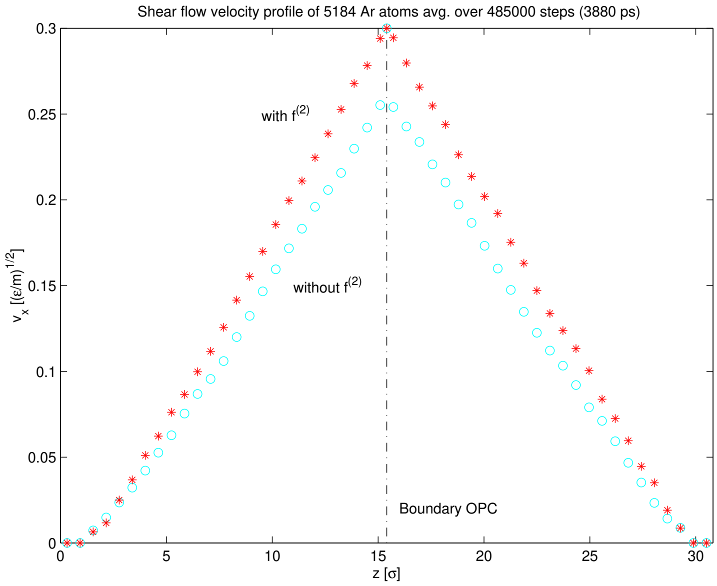

Figure 4. Result of applying direct boundary OPC with and without $f^{(2)}$ in a simple shear flow scenario.

4. Improved Single-Particle Distribution Function

Our discussion in section 2 is general with respect to the single-particle distribution function f. That is, we only assume f exists, and its leading order is the local Maxwellian distribution $f^{(0)}$, without insisting on a certain form of $f^{(2)}$. Indeed, it is all right to just use $f^{(0)}$ if EBC is used to impose the continuum BC, since $f^{(2)}$ and higher-order terms will then be present naturally at $\partial\mathcal{C}$. But, if one directly imposes OPC on $\partial\mathcal{C}$ as BC, he should be more careful about $f^{(2)}$ to at least ensure the continuity of stress and heat current across $\partial\mathcal{C}$. An example is shown in Fig. 4, where direct boundary OPC is imposed on the center plane to achieve a shear flow speed of 0.3, in an MD simulation of 5,184 LJ6-12 atoms at a reduced temperature of 1.1, with solid walls on both sides[1]. However, if one just uses $f^{(0)}$ to derive and implement a boundary OPC, the flow speed will not reach the desired value but actually drop to 0.25.

A simple mechanical reason for this behavior is that $f^{(0)}$, the local Maxwellian distribution, does not carry any shear stress (or heat current) since the net flux of momentum (or energy) across any small area vanish whenever the velocity distribution is isotropic. On the other

[1] This setup can be used to study the Couette flow with continuum-atomistic coupling, as part (b) of the iteration in Fig. 1.

hand, shear stress and $f^{(2)}$ do exist in the fluid bulk as in Fig. 4, and atoms near the center plane, newly assigned the low-order distribution $f^{(0)}$, have to satisfy stress continuity by sacrificing parts of their own inertia. Thus $f^{(2)}$ is achieved by atoms leaving the center plane into the "bulk", at the cost of $\bar{v}_z$.

The classical description one has on $f^{(2)}$ is the Chapman-Enskog development [9] in kinetic theory. Although that theory is meant for gases, we nevertheless use it for inspiration in liquids. The big difference between a liquid and a gas is that the interatomic interaction is a dominant contribution to fluxes (shear stress, heat current) in a liquid, whereas it is relatively less importance in a gas. Thus in liquids, spatial correlation like $g(\mathbf{r})$ should contribute roughly the same to fluxes as velocity distribution $f^{(2)}$. However, we are only able to modify $f^{(2)}$, a single-particle rather than a two-particle distribution, in our present formalism. The plausibility argument for doing so would be a mechanical one: so long as we satisfy flux continuity at the interface by assigning enough fluxes to $f^{(2)}$, the internal conversion between $f^{(2)}$ and $g(\mathbf{r})$ can be carried out in a relatively "peaceful" manner in the skin region near the boundary without causing great error to other properties, unlike the former scenario where *not* enough flux is assigned to $f^{(2)}$, and the fluids have to pay for it by themselves.

In the Chapman-Enskog development, the velocity distribution is expanded as

$$f = \frac{\rho(\mathbf{x})}{(2\pi T(\mathbf{x}))^{3/2}} \exp\left(-\frac{|\mathbf{v}-\bar{\mathbf{v}}(\mathbf{x})|^2}{2T(\mathbf{x})}\right)(1+\phi^{(2)}) + ..., \tag{4}$$

which is then plugged into the Boltzmann equation. It happens that there exists exact solution for a so-called quasi-Maxwell model [9], where $\phi^{(2)}$ turns out to be a linear sum of several terms,

$$\phi^{(2)} = \phi^{(2)}_{shear} + \phi^{(2)}_{heat} + ... \tag{5}$$

For the purpose of this discussion we will only consider the shear flow term,

$$\phi^{(2)}_{shear} = \frac{-\mu}{\rho(\mathbf{x})T(\mathbf{x})^2}(\mathbf{v}-\bar{\mathbf{v}}(\mathbf{x}))^T\mathbf{D}(\mathbf{x})(\mathbf{v}-\bar{\mathbf{v}}(\mathbf{x})), \tag{6}$$

where

$$D_{\alpha\beta}(\mathbf{x}) = \frac{1}{2}\left(\frac{\partial\bar{v}_\alpha}{\partial x_\beta} + \frac{\partial\bar{v}_\beta}{\partial x_\alpha}\right) \tag{7}$$

is the fluid strain rate tensor, and

$$\mu = -\frac{(2T)^{1/2}}{2\sigma\lambda_{02}} \tag{8}$$

is fluid shear viscosity, expressed in terms of the collisional cross-section σ and some eigenvalue λ_{02}.

We then approximate $1+\phi^{(2)}_{shear}$ in (4) by $\exp(\phi^{(2)}_{shear})$, a common maneuver in perturbation expansion, to show that the only apparent effect of $\phi^{(2)}_{shear}$ is to distort the velocity distribution f from an isotropic Gaussian distribution $f^{(0)}$ to a *tilted* Gaussian distribution. In the simple shear flow scenario depicted in Fig. 4, the principal axes of the tilted Gaussian are $\{(\bar{v}_x+\bar{v}_z)/\sqrt{2}, \bar{v}_y, (\bar{v}_x-\bar{v}_z)/\sqrt{2}\}$, as

$$(v_x-\bar{v}_x(z), v_y, v_z)\cdot[\begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \longrightarrow \begin{pmatrix} 1 & 0 & \gamma \\ 0 & 1 & 0 \\ \gamma & 0 & 1 \end{pmatrix}]\cdot\begin{pmatrix} v_x-\bar{v}_x(z) \\ v_y \\ v_z \end{pmatrix}, \quad (9)$$

in the exponent, with off-diagonal coupling coefficient $\gamma(\mathbf{x})$ defined to be

$$\gamma(\mathbf{x}) \equiv \frac{\tau_{xz}(\mathbf{x})}{\rho T}. \quad (10)$$

The merit of (10) is that it is readily computable on the fly in a coupled continuum-MD simulation, without any other extra parameters. And one can easily check that by including off-diagonal coupling in (4), the stress carried by f is exactly $\tau_{xz}(\mathbf{x})$, which is demanded by mechanical equilibrium. To include $\gamma(\mathbf{x})$ in the bulk OPC is very easy: f is still factorizable in the $\{(\bar{v}_x+\bar{v}_z)/\sqrt{2}, \bar{v}_y, (\bar{v}_x-\bar{v}_z)/\sqrt{2}\}$ frame, so (3) can be readily used. The boundary particle controller is a lot more complicated because the boundary velocity distribution is not a Gaussian [7] and cannot be factorized. It is not clear yet what transformation is the OPC in that case. Some preliminary tests suggest that by giving each atom which hits the z-plane an extra $-2\gamma(\mathbf{x})v_z$ in v_x in addition to the boundary OPC as if $f^{(2)}$ is not present, the results can be satisfactory, as shown in Fig. 4.

Finally, we mention that in the spirit of the Chapman-Enskog expansion, it is all right to use $f^{(0)}$ in the TFE to estimate the current fields $\rho(\mathbf{x})$, $T(\mathbf{x})$, $\bar{\mathbf{v}}(\mathbf{x})$, calculate the spatial gradients, and then impose direct $f^{(0)}+f^{(2)}$ OPC using Chapman-Enskog $f^{(2)}$ with *those* gradients. It achieves the same level of accuracy as initially using $f^{(0)}+f^{(2)}$ TFE.

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