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Optimal particle controller for coupled continuum/MD fluid simulation [1]

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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. The importance of incorporating a higher order single-particle distribution function in light of the Chapman–Enskog development is demonstrated for shear flow.

Keywords: Please provide 3–5 keywords

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 [3–5] cast their formulation in the framework of alternating Schwarz method [6,7] and treated the matching in terms of refining the boundary conditions imposed on each of the overlapping subdomains through an iterative process.

Our implementation [8–10] 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 boundary conditions, and (b) to perform the inverse, i.e., make sure that particles in an MD simulation do correspond to a set of prescribed macroscopic fields 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 [8] in the form of an algorithm called the thermodynamic field estimator (TFE). A method for (b) will be introduced in Section 2. 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 boundary conditions. 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 Section 3, we explain the importance of adopting a more accurate single-particle distribution function, other than the lowest-order Maxwellian distribution. By looking at the Chapman–Enskog development in kinetic theory, we work out an approximate solution to solve the outstanding problems.

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2. Optimal particle controller

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 [3–5,8–10] is likely to be the single-particle distribution function,

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

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. The Navier–Stokes equation can then be derived from the Chapman–Enskog development [12].

In a previous paper [8] 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, 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 phase space 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 replace X_n's by Y_n's such that $\{Y_n\}$ will satisfy $g(Y)$, what should be the *optimal* $\mathcal{T}\colon X_n \to Y_n$ transformation (Fig. 1)?

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 nth particle before and after the $\mathcal{T}$ transformation, under 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 subtleties of boundary OPC involving conditional probability are already discussed in a previous paper [9,10].

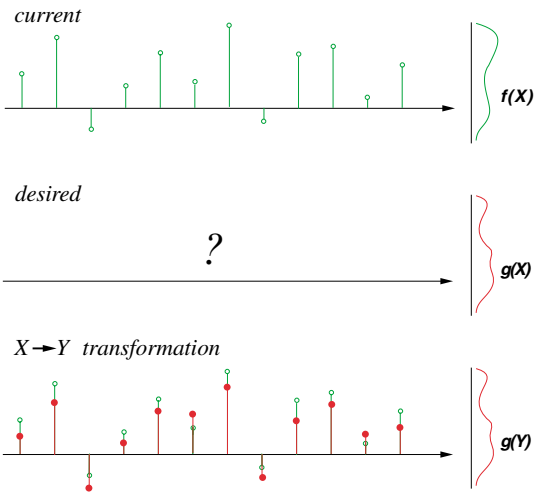

Fig. 1. Given that an incoming sequence $\{X\}$ (circles) satisfies a *current* distribution $f(X)$, but one would like to change $\{X\}$ to $\{Y\}$ (dots) such that Y satisfy the desired distribution $g(Y)$, what $\mathcal{T}\colon X \to Y$ transformation should one use? Note that the average difference between circles and dots represent the magnitude of the artificial action one applies onto the atoms.

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,

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

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 a longer paper [11].

3. Second-order single-particle distribution functions

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 the so-called extended boundary condition [9,10] is used to impose the boundary conditions, since $f^{(2)}$ and higher-order terms will appear automatically at $\partial\mathcal{C}$. But, if one directly imposes OPC on $\partial\mathcal{C}$ as boundary conditions, 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. 2, where direct boundary OPC is imposed on the center plane to achieve a shear flow speed of 0.3, in an MD simulation of 5184 LJ6-12 atoms at a reduced temperature of 1.1, with

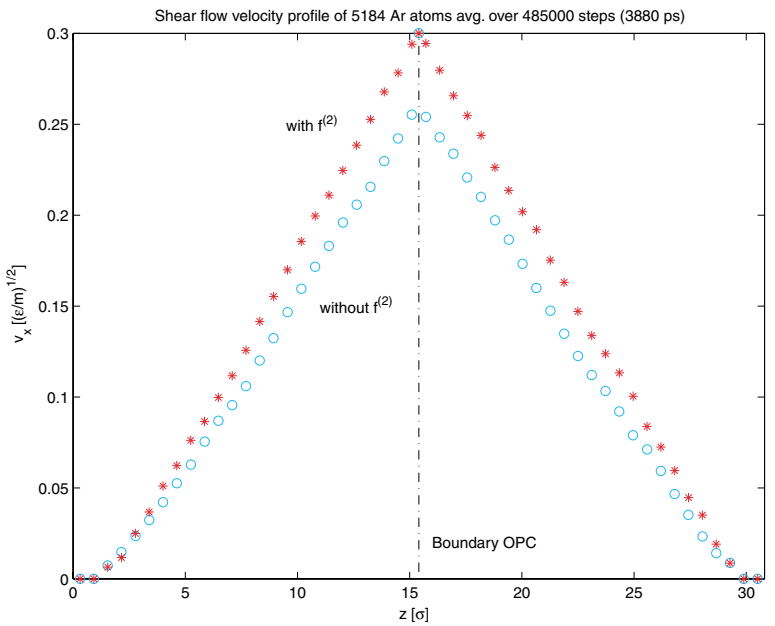

Fig. 2. Result of applying direct boundary OPC with and without $f^{(2)}$ in a simple shear flow scenario.

solid walls on both sides. 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 hand, shear stress and $f^{(2)}$ do exist in the fluid bulk as in Fig. 2, 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. $f^{(2)}$ is only approached gradually as atoms leave the center plane and come into the bulk, at the cost of $\bar{v}_z$.

The classical description on $f^{(2)}$ is the Chapman–Enskog development [12]. Although that theory is meant for gases, we nevertheless use it for 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 non-disturbing manner in the skin region near the boundary, unlike the case 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

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

which is then plugged into the Boltzmann equation. It happens that there exists exact solution for a so-called quasi-Maxwell model [12], where $\phi^{(2)}$ turns out to be a linear sum of several terms,

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

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

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

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 kernel eigenvalue λ_{02}.

We then approximate $1+\phi^{(2)}_{\text{shear}}$ in (4) by $\exp(\phi^{(2)}_{\text{shear}})$ to show that the leading-order effect of $\phi^{(2)}_{\text{shear}}$ 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. 2, 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

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

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

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

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 [9,10] 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 are satisfactory, as shown in Fig. 2.

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 $f^{(2)}$ using those gradients. It achieves the same level of accuracy as initially using $f^{(0)} + f^{(2)}$ TFE.

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