IMPOSING FIELD BOUNDARY CONDITIONS IN MD SIMULATION OF FLUIDS: OPTIMAL PARTICLE CONTROLLER AND BUFFER ZONE FEEDBACK

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ABSTRACT

We formulate a method for imposing continuous field boundary conditions on an MD simulation with little or no disturbance to its dynamics. Our approach combines the previously developed thermodynamic field estimator, which extracts macroscopic fields from particle data, with a novel procedure, optimal in the sense of least disturbance, for imposing prescribed continuum field boundary conditions on the atomistic system. By means of feedback control and assuming molecular chaos in fluids, that disturbance can further be eliminated entirely, thus providing an exact solution for general steady-state fluid problems, where the desired fields at the real boundary is achieved by adjusting actions in a region *separated* from the real boundary by a buffer zone.

OPTIMAL PARTICLE CONTROLLER (OPC)

A basic problem in fluid simulation, which was first investigated by O'Connell and Thompson[1], then Hadjiconstantinou and Patera[3], and then by us[4, 5], is the following: we would like the particles in an MD simulation to satisfy the following distribution in their positions and velocities $\{\mathbf{x}_i, \mathbf{v}_i\}$,

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

at a prescribed boundary $\partial\mathcal{C}$, where $\rho(\mathbf{x})$, $T(\mathbf{x})$, $\bar{\mathbf{v}}(\mathbf{x})$ are the macroscopic density, velocity and temperature fields. For this to be sensible, $\partial\mathcal{C}$ should be in small-gradient regions where $\rho(\mathbf{x})$ etc. are well-defined; conversely, if that is true, the transition from particle to continuum description like the Navier-Stokes equation, can be shown to be exact by the Chapman-Enskog expansion, where $f^{(2)}$ is the the second-order correction to the local Maxwellian that contains the spatial gradients of these fields. So, both continuum and particle descriptions should be valid when close to $\partial\mathcal{C}$, and (1) is the link that allows us to couple the two.

In a previous paper[4] we have shown how to solve the opposite problem, that is how to *infer* the macroscopic fields in $\mathcal{C}$ from current particle data, using a technique named thermodynamic field estimator (TFE); the upshot is that we can accurately determine the macroscopic fields at an inner boundary $\partial\mathcal{U}$ inside $\mathcal{C}$ in real time even when the particle data contains considerable "thermal noise". And so $\partial\mathcal{U}$ provides the BC for a continuum solver in the classical domain decomposition and alternating Schwarz iteration formalism[2], where $\partial\mathcal{C} - \partial\mathcal{U}$ is the continuum-MD overlap region. Because of space limitation we will not further discuss the TFE here, nor the full Schwarz iteration implementation which we have carried out for Couette flow, but will focus on *just* the second part of the iteration, which is how to impose continuous field BC on $\mathcal{C}$. Interested readers should follow the pioneering paper of Hadjiconstantinou and Patera[3], and much detailed reports of our work in [4, 5]. Suffice it here to note that in order for the whole scheme to work to second order accuracy in (1), we can simply use the first term in (1) for TFE, ignoring $f^{(2)}$.

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 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 set $\{X_n\}$ satisfying distribution $f(X)$, but we want them to satisfy distribution $g(X)$, so we begin to replace X_n's by Y_n's hoping that $\{Y_n\}$ will satisfy $g(Y)$, what should be the optimal $X_n \to Y_n$ transformation?

One may ignore X_n and use only $g(Y)$ to resample the random variable Y_n. While this would indeed lead to a BC imposing scheme, it is quite conceivable that ignoring the current state will result in a procedure that strongly disturbs the particle dynamics. If decoupling the two distributions, or equivalently, the two sets of particle coordinates, is not a good idea, then one should look for a way to relate them such that the disturbance is minimized. We now introduce a quantitative measure of this disturbance:

$$B = \sum_n |\Delta \mathbf{v}_n|^2 = \sum_n |\mathbf{v}_n^{\text{out}} - \mathbf{v}_n^{\text{in}}|^2, \tag{2}$$

where $\Delta \mathbf{v}_n$ is the change in particle velocity of the n^{th} particle ($\mathbf{v}_n^{\text{in}}$ and $\mathbf{v}_n^{\text{out}}$ are the velocity before and after the transformation). This quantity characterized the artificial disturbance to particle dynamics which should be minimized as much as possible.

As an example let us consider a simple problem of 1D heat conduction, for which we wish to impose a boundary condition of high temperature T_h at $x = 0$ and low temperature T_l at $x = 1$. Intuitively one could imagine doing the following: when a particle crosses either boundary $x = 0$ or 1, give it a random velocity drawn from distribution (1) with parameters $\bar{\mathbf{v}} = 0$ and $T = T_h$ or T_l. However, this procedure does not work. When implemented in practice in the case of homogeneous heating ($T_h = T_l$), results show that the bulk temperature reaches a value of $T_h/2$.

The scheme fails because one is dealing with conditional probability. The speed distribution of atoms which cross the boundary is *different* from the speed distribution of atoms in the bulk. Instead it is weighted by the normal velocity,

$$dP = \frac{v \exp\left(-\frac{v^2}{2T}\right) dv}{\int_0^{+\infty} v \exp\left(-\frac{v^2}{2T}\right) dv} = \frac{v}{T} \exp\left(-\frac{v^2}{2T}\right) dv, \qquad 0 < v < +\infty. \tag{3}$$

$\langle v^2 \rangle$ from distribution (3) is $2T$, not T. Thus, if we sample the *boundary crossing atoms* using bulk distribution (1) with parameter $T = T_h = T_l$, the energy can only be balanced in a statistical sense when the bulk temperature reaches $T_h/2$.

A more subtle defect of this scheme is that, for whatever the incoming velocity v_{in} of the particle before hitting the boundary, a new velocity v_{out} is drawn from a given distribution, say $g(v)$, entirely *independent* of v_{in}. Thus if we evaluate the disturbance to particle dynamics using (2), it is always substantial. In the case of *homogeneous heating*, even when the correct distribution (3) is used in drawing v_{out}'s and the system has reached the desired temperature $T = T_h = T_l$, the scheme continues to disturb the particles by giving each boundary crossing atom a new v_{out}. On the other hand, if we just let $v_{\text{out}} = v_{\text{in}}$, i.e., do nothing, the system temperature stays at T_h! A more intelligent particle controller should automatically tune down its influence as the system approaches the desired state, a behavior we may call the coalescence property.

We now formulate the above ideas mathematically. Suppose $\{X_n\}$ conform to distribution function $f(X)$:

$$dP(\eta < X < \eta + d\eta) = f(\eta) d\eta, \tag{4}$$

and we would like the series to conform to a different distribution g. We propose to achieve this by replacing every X with another number Y which is distributed according to g. Thus the goal is to find a transformation $\mathcal{T}$,

$$\mathcal{T} : X_n \to Y_n, \tag{5}$$

with the requirement that if $\{X_n\}$ conforms to distribution $f(X)$, $\{Y_n\}$ will conform to $g(Y)$:

$$dP(\xi < Y < \xi + d\xi) = g(\xi) d\xi. \tag{6}$$

There are many possible $\mathcal{T}$'s. However our previous discussion shows that the following property is desirable: *if* $f \equiv g$, *then* $\mathcal{T}$ *gives* $Y_n \equiv X_n$. Generally, to incorporate the idea of *minimally disturbing the dynamics*, we adopt the reasonable criterion that

$$B[\mathcal{T}] = \langle (Y - X)^2 \rangle \tag{7}$$

be minimized among all possible $\mathcal{T}$'s.

We see that if $\mathcal{T}$ is randomly drawing Y from $g(Y)$ without referencing to X, it satisfies the basic requirement (6) but not the coalescence property. We call this $\mathcal{T}_1$ transformation. To incorporate coalescence one may consider $\mathcal{T}_2$,

$$\mathcal{T}_2 : \begin{cases} Y = X : & p \leq \frac{g(X)}{Kf(X)} \\ \text{draw } Y \text{ randomly from } g(Y) : & p > \frac{g(X)}{Kf(X)} \end{cases}$$

where p is a random number uniformly distributed over $[0, 1]$, and K is a constant (scheme fails if K does not exist) such that

$$Kf(X) \geq g(X) \ \text{ for } \ -\infty < X < +\infty. \tag{8}$$

But is $\mathcal{T}_2$ the best one?

We propose the following transformation, in the form of an implicit relation,

$$\mathcal{T}_3(X \to Y) : \int_{-\infty}^{X} f(\xi)d\xi = \int_{-\infty}^{Y} g(\xi)d\xi. \tag{9}$$

It can be checked that Y_n indeed conforms to distribution g if the incoming random variable X_n conforms to f, and it satisfies the coalescence property. We believe that $\mathcal{T}_3$ is the mathematically optimal transformation which minimizes (7). A tentative proof is given in [5]. This assertion should come as no surprise because it is the only one-to-one continuous mapping which satisfies (6) without extra randomness like in $\mathcal{T}_2$. Our experience shows that while $\mathcal{T}_3$ works rather well, it is an implicit algorithm and could be computationally demanding.

One may choose to operate $\mathcal{T}$ in a finite-volume region or just at a certain boundary, which are called bulk or boundary particle controllers respectively. The subtleties of boundary particle controllers are already mentioned in the first example. When the average disturbance to microscopic particle dynamics, defined by some reasonable measure, due to the operation of a family of particle controllers serving certain fixed purpose, is minimized, it is called the optimal particle controller (OPC). We believe that for one- or decoupled multi-dimensional distributions, $\mathcal{T}_3$ is OPC. The explicit formulas for general 3D $T(\mathbf{x})$, $\bar{\mathbf{v}}(\mathbf{x})$ boundary particle controller are given in [5], while $\rho(\mathbf{x})$ coupling involves particle insertion or removal which is more difficult to treat (see [4]). But explicit $\rho(\mathbf{x})$ boundary condition is rare. Also, those derivations ignore $f^{(2)}$, which cause certain problems, and we will derive more accurate versions by factoring $f^{(2)}$ into the distribution function. In all cases the general discussions about OPC hold.

EXTENDED BOUNDARY CONDITION

In last Section a method was proposed to control the field boundary condition of an MD simulation that results in 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 specified boundary, thus providing an *exact* theoretical 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.1, 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 field boundary conditions on the core boundary $\partial\mathcal{C}$. Due to molecular chaos in fluids, short wavelength

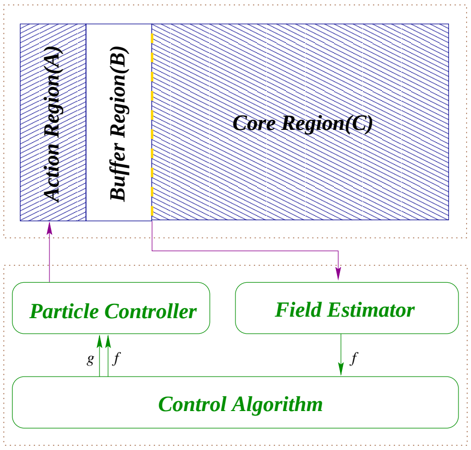

Figure 1: Schematic diagram of the Extended Boundary Condition (EBC), which incorporates a field estimator, a particle controller and a feedback control algorithm.

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. The action in $\mathcal{A}$ is in general a matter of choice, but here we will use the OPC developed in the last section 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.

Note that although $\mathcal{A}$ and $\mathcal{B}$ are discrete-particle regions governed by molecular dynamics as $\mathcal{C}$, they are not real physical domains that appear in the Schwarz domain decomposition. They are constructs existing in the MD simulation solely 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. It in turn can be used as a perfect benchmark for comparing the actual performances of $\mathcal{T}_1$ and $\mathcal{T}_3$, as we will see.

In applying the EBC, the buffer region $\mathcal{B}$ is between $\mathcal{A}$ and $\partial\mathcal{C}$. Actions are applied on $\mathcal{A}$ instead of $\partial\mathcal{C}$ in such a way that the desired field at $\partial\mathcal{C}$ is obtained. The question then becomes, given the desired flow velocity at $\partial\mathcal{C}$, v_C^*, how does one control the desired flow velocity at $\mathcal{A}$ (prescribed value to the OPC), v_A^*? Adopting a first-order feedback control algorithm, we may write,

$$\frac{dv_A^*(t)}{dt} = -\kappa(v_C(t) - v_C^*), \tag{10}$$

where the rate constant κ should be picked according to the approximate response time at $\partial\mathcal{C}$ due to an action on $\mathcal{A}$, making $v_C(t)$ converge to v_C^* as quickly as possible without oscillation.

Compared to 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 come out automatically on $\partial\mathcal{C}$. The drawback is that for general BC, multi-variable feedback control is needed, which is quite a challenging task.

COUETTE FLOW TEST

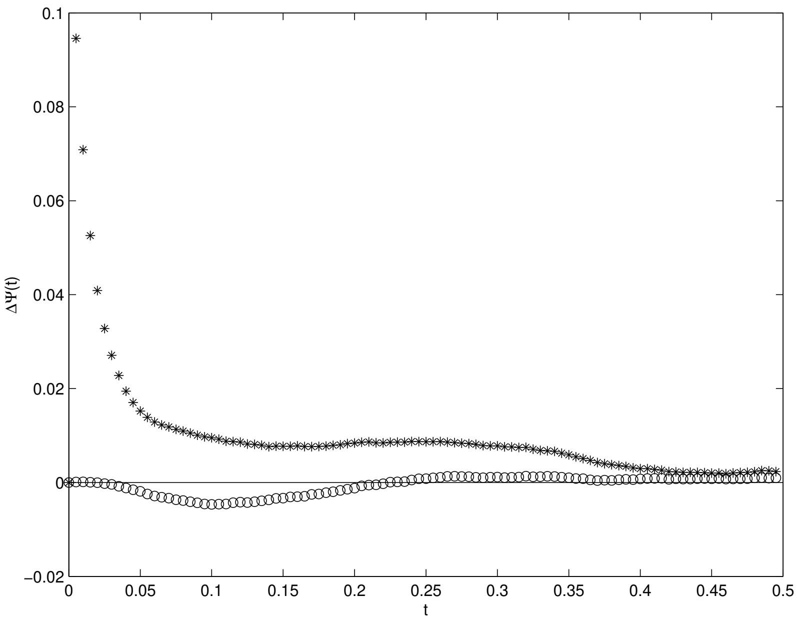

Figure 2: Comparison of the change to the normalized velocity autocorrelation function $\Psi(t)$ due to $\mathcal{T}_1$ and OPC. $\Psi_{\mathrm{EBC}}(t) - \Psi_{\mathcal{T}_1}(t)$ is plotted in stars and $\Psi_{\mathrm{EBC}}(t) - \Psi_{\mathrm{OPC}}(t)$ is plotted in circles.

In this Section we demonstrate how the above ideas are implemented for Couette flow simulation. The process is somewhat reversed: we first implement the direct BC's using $\mathcal{T}_1$ and OPC, obtain the actual flow velocity value at $\partial\mathcal{C}$, and then use EBC to reproduce that actual value at $\partial\mathcal{C}$ by acting on $\mathcal{A}$. We then in turn calculate a specially defined particle velocity auto-correlation function with EBC, and compare those of the direct approaches to it, to show that OPC is indeed much more effective than $\mathcal{T}_1$ in preserving the particle dynamics.

The velocity autocorrelation function is a well-used quantity which characterizes microscopic dynamics. We can define its localized and normalized version

$$\Psi(t) = \frac{\langle \mathbf{v}_i(0) \cdot \mathbf{v}_i(t) \rangle}{\langle \mathbf{v}_i(0)^2 \rangle}, \tag{11}$$

for any i^{th} particle that crosses $\partial\mathcal{C}$ and comes into $\mathcal{C}$ region at $t = 0$; its contribution to $\Psi(t)$ is terminated whenever the particle re-crosses $\partial\mathcal{C}$. Since the EBC acts on $\mathcal{A}$ instead of $\partial\mathcal{C}$, $\Psi_{\mathrm{EBC}}(t)$ should be the same as that of nature where no artificial disturbance can be discerned on $\partial\mathcal{C}$. On the other hand, it is not so if we directly act on $\partial\mathcal{C}$. However, $\Psi(t)$ of direct OPC should be better than that of direct $\mathcal{T}_1$ in comparison with $\Psi_{\mathrm{EBC}}(t)$. Fig. 2 shows the difference $\Psi_{\mathrm{EBC}}(t) - \Psi_{\mathcal{T}_1}(t)$ in stars and $\Psi_{\mathrm{EBC}}(t) - \Psi_{\mathrm{OPC}}(t)$ in circles. Notice there is a drastic loss of correlation in the first 0.05 ps, clearly attributable to the reaction of surrounding atoms when its direction is suddenly altered from its original motion. On the other hand, $\Psi_{\mathrm{OPC}}(t)$ is almost indistinguishable from $\Psi_{\mathrm{EBC}}(t)$.

DISCUSSIONS

We have developed a hybrid continuum-atomistic simulation scheme[4, 5] based on the concept of domain decomposition, in the same spirit as Hadjiconstantinou and Patera[3]. Such methods are useful for problems where much of the region of interest can be treated by continuum description, but a small critical, embedded part requires atomistic simulation. While Hadjiconstantinou and Patera implemented their method with attention to both the atomistic and continuum representations, we have focused our efforts on techniques for imposing continuum BC to atomistic simulations. In this work matching of the two different levels of representation, each with its own set of degrees of freedom, takes place in the overlap region through an implicit transformation which we have called the Optimal Particle Controller, with Eqn (1) serving as the bridge that depends on both particle and field variables. Relative to the conventional method of sampling from a desired distribution such as the $\mathcal{T}_1$ transformation, OPC has the advantage that it is least disturbing to the particle dynamics, and that disturbance is proportional to the rate of dissipation in the system instead of the absolute magnitude of the fields.

We have incorporated OPC along with the Thermodynamic Field Estimator and the Extended Boundary Condition into the framework of alternating Schwarz method and implemented it in a study of shear flow. We should regard the numerical results[5] as proof of the principles of the entire hybrid scheme. Since each one of these three techniques has its own novel features, further applications, including separate investigations of each technique, will be worthwhile to bring out their capabilities and limitations. We believe that relative to the other hybrid methods recently proposed [1, 3] our method should be the most gentle as long as treating the atomistic subdomain is concerned. For this reason the method should be most useful when one is interested in delicate or subtle molecular effects where minimizing local disturbance to particle dynamics is a significant concern.

Up to now we have only been concerned with steady state fluid problems. One may wonder if a continuum solver could be dynamically coupled with an MD solver using the above techniques in real time. Nobody has the exact solution yet. On the other hand, the timescale of an macroscopic dynamical event is usually much greater than that of its underlying microscopic mechanism, thus to the critical microscopic region at any given moment of the event, the outside world would seem to be in a steady state flow condition and could be modeled by our techniques.

The case for solids is different. Solids, unlike fluids, have long range order. The assumption of molecular chaos is invalid here, and a buffer zone cannot cover up the disturbed dynamics elsewhere, which is the underpinning of the Extended Boundary Condition. Thus, it is quite difficult to isolate a region in a crystalline material since the phonons have very long mean free paths, and their scattering and reflections determine important macroscopic properties such as the thermal conductivity. Although researchers have developed successful techniques to couple continuum with atomistic regions for static calculations, no one has yet claimed to successfully implement such a scheme to study finite temperature properties.

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