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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 compares the estimated current fields with the desired fields at the boundary and modifies the action 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 a mutually consistent solution for steady-state problems, as particles in the MD region of interest have no way to tell any difference from reality. Finally, we explain the importance of using a higher order single-particle distribution function, in light of the Chapman–Enskog development for shear flow.

Keywords: Buffer, Continuum, Feedback, Inference, Least disturbance, Molecular dynamics

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 the Alternating Schwarz method [4, 5] and treated the matching in terms of refining the boundary conditions imposed on each of the overlapping subdomains through an iterative process (see Fig. 1).

Our approach [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 results of an MD simulation, to be plugged into a continuum solver as the boundary condition (BC) and (b) to perform the inverse, i.e., make sure that particles in an MD simulation do correspond to

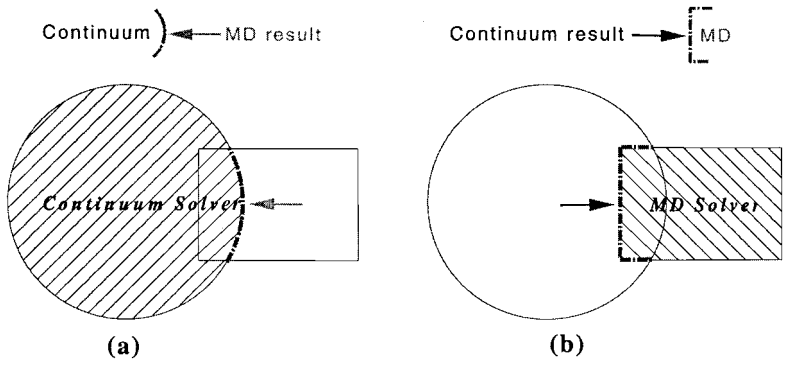

Figure 1. Alternating Schwarz formalism to get a joint solution in two subdomains handled by different solvers. The atomistic subdomain will be treated by MD simulation, while the other subdomain will be treated by an appropriate continuum solver. In the iteration, two kinds of boundary condition (BC)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

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 [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 an exact solution to the problem of (b). Combining (a) and (b) in the Alternating Schwarz formalism (Fig. 1), one could obtain exact unified solution for most steady-state fluid problems.

In Section 4, we explain the significance of adopting a more accurate single-particle distribution function, other than the lowest-order local Maxwellian distribution, that is especially critical when the direct OPC (stage one) method is used to impose the BC in (b). Referring to 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 individual 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 a many-body Newton's equation. The bridge linking the two, we

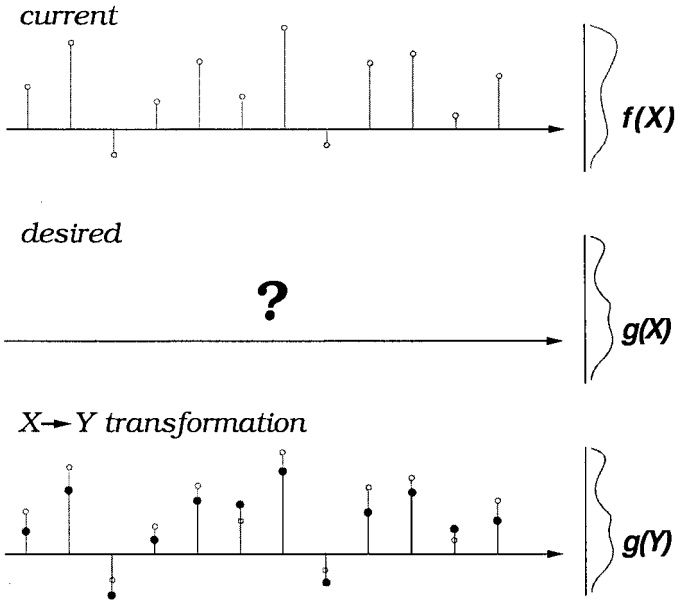

Figure 2. Given that an incoming sequence $\{X\}$ (circles) satisfies a current distribution $f(X)$, but one would like to change $\{X\}$ to $\{Y\}$ (dots) such that Y satisfies the desired distribution $g(Y)$, what $\mathcal{T} : X \to Y$ transformation should one use? Note that the average difference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

think [3, 6, 7, 9], should be the single-particle distribution function

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

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, Equation 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 Equation 1 and having small gradients, because otherwise it makes no sense to do coupling 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 physics in the Chapman–Enskog development [9], assuming Equation 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, 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

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

where $\mathbf{v}_n^{\text{bef}}$ and $\mathbf{v}_n^{\text{aft}}$ are the velocities of the n^{th} particle before and after the $\mathcal{T}$ transformation, on the constraint that $\mathbf{v}_n^{\text{aft}}$ 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 at a certain boundary such as $\partial\mathcal{C}$, which are then called bulk or boundary OPC, respectively. The subtlety of boundary OPC involving conditional probability was 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-dimensional 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 Equation 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 least disturbance to the particle dynamics in the sense of Equation 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 its 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 in 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 on $\mathcal{A}$–$\mathcal{C}$ separation. Only the hydrodynamic variables which correspond to the three collisional invariants would still have influence on $\partial\mathcal{C}$. The action in $\mathcal{A}$ is in general a matter of choice, but we use the boundary OPC introduced in Section 2 because it minimizes the artificial disturbance, therefore $\mathcal{B}$ 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 sends instructions to the particle controller.

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 and do not appear in the Schwarz domain decomposition diagram in Fig. 1. They are only numerical constructs in the MD sub-step of the iteration, to ensure that the physical region of interest, $\mathcal{C}$, has the correct field boundary

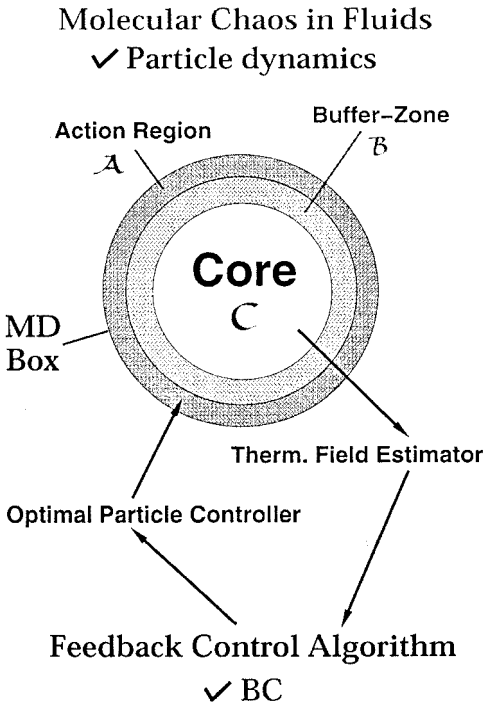

Figure 3. Schematic diagram of the extended boundary condition (EBC), which incorporates a field estimator, a particle controller and a feedback control algorithm.

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 to directly applying OPC on $\partial\mathcal{C}$ without a buffer zone, EBC has two advantages: (1) It generates practically no disturbance to the core region. (2) We do not need to worry about $f^{(2)}$ and 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.

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 be present naturally at $\partial\mathcal{C}$. But, if one directly applies OPC on $\partial\mathcal{C}$ as BC, one 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 5184 LJ6-12 atoms at a reduced temperature of 1.1,

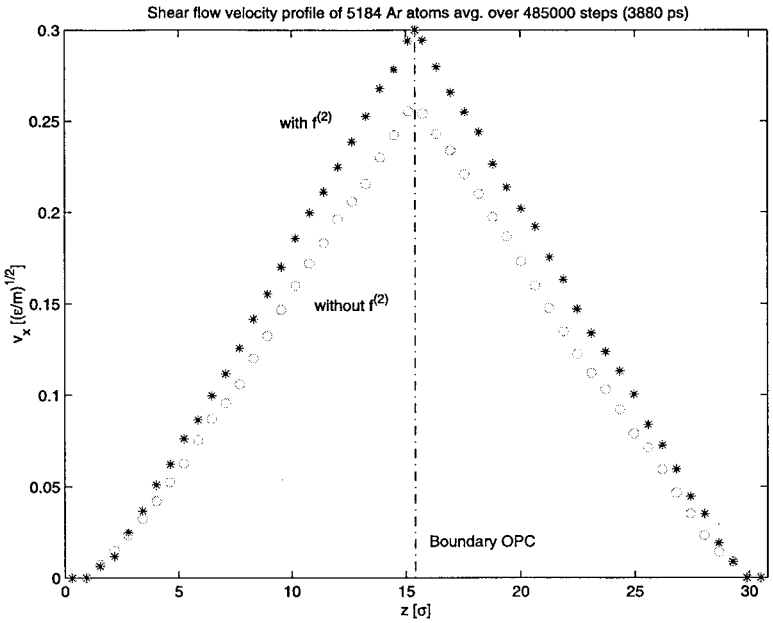

Figure 4. Result of applying direct boundary OPC with and without $f^{(2)}$ in a simple shear flow scenario.

with solid walls on both sides[1]. However, if one just uses $f^{(0)}$ to derive and implement the 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 differential area vanishes whenever the velocity distribution is isotropic. On the other hand, shear stress and $f^{(2)}$ do exist in the fluid bulk 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 expense of $\bar{V}_x$.

The common description on $f^{(2)}$ is the Chapman–Enskog development [9] in kinetic theory. 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 major contributor to fluxes (shear stress, heat current) in a liquid, whereas it is relatively less important 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 flux to $f^{(2)}$, the internal conversion between $f^{(2)}$ and $g(\mathbf{r})$ can be accomplished in a relatively 'peaceful' manner in the skin region near the boundary without causing great error to other properties, unlike the prior scenario where not enough flux is assigned to $f^{(2)}$, and the fluids have to pay for it by themselves.

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

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 an exact solution for a so-called quasi-Maxwell model [9], where $\phi^{(2)}$ turns out to be a summation 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 the 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 Equation 4 by $\exp(\phi^{(2)}_{shear})$, a common maneuver in perturbation expansion, to show that the only significant 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 $\{(v_x+v_z)/\sqrt{2}, v_y, (v_x-v_z)/\sqrt{2}\}$, as

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

in the exponent, with the 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 Equation 10 is that it is readily computable on the fly in a coupled continuum-MD simulation, without any extra parameters. And one can easily check that by including off-diagonal coupling in Equation 4, the stress carried by f is exactly $\tau_{xz}(\mathbf{x})$, which is necessary for achieving mechanical equilibrium. To include $\gamma(\mathbf{x})$ in the bulk OPC is very simple: f is still factorizable in the $\{(v_x+v_z)/\sqrt{2}, v_y, (v_x-v_z)/\sqrt{2}\}$ frame, so Equation 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 their spatial gradients, and then impose direct $f^{(0)} + f^{(2)}$ OPC using the Chapman–Enskog $f^{(2)}$ with those gradients. It achieves the same order of accuracy as initially using $f^{(0)} + f^{(2)}$ TFE.

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References

1. Koplik, J. and Banavar, J.R., Annu. Rev. Fluid Mech., 7 (1995) 257.
2. O'Connell, S.T. and Thompson, P.A., Phys. Rev., E 52 (1995) 5792.
3. Hadjiconstantinou, N. and Patera, A.T., Int. J. Modern Phys. C, 8 (1997) 967.
4. Lions, P.L., In First International Symposium on Domain Decomposition Methods for Partial Differential Equations, SIAM, 1988, pp. 1–42.
5. Smith, B.F., Bjorstad, P.E. and Gropp, W.D. Domain Decomposition: Parallel Multilevel Methods for Elliptic Partial Differential Equations, Cambridge University Press, Cambridge, 1996.
6. Li, J., Liao, D. and Yip, S., Phys. Rev. E, 57 (1998) 7259.
7. Li, J., Liao, D. and Yip, S., Mat. Res. Soc. Symp. Proc., 538 (1998) 473.
8. Li, J., Liao, D. and Yip, S., to be submitted.
9. Uhlenbeck, G.E. and Ford, G.W., Lectures in Statistical Mechanics, American Mathematical Society, Providence, RI 1963, p. 102.