BULLETIN OF THE POLISH ACADEMY OF SCIENCES
TECHNICAL SCIENCES
Vol. 53, No. 4, 2005

Discussion of recent developments in hybrid atomistic-continuum methods for multiscale hydrodynamics

N.G. HADJICONSTANTINOU*

Mechanical Engineering Department, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

Abstract. We discuss recent progress in hybrid atomistic-continuum methods with particular emphasis on developments in boundary condition imposition in molecular simulations, an essential ingredient of hybrid methods. Both Dirichlet (state variable) and flux boundary conditions are discussed. We also briefly review various coupling approaches and discuss the effects of compressibility and molecular fluctuations on the choice of coupling method. Common elements between hybrid methods and related multiscale simulation approaches are also briefly discussed.

Key words: hybrid atomistic-continuum method, multiscale.

1. Introduction

By limiting the molecular description to regions where it is needed, hybrid methods allow the simulation of complex phenomena which require modeling at the molecular scale without the prohibitive cost of a fully molecular calculation. In this paper we discuss recent progress in this rapidly expanding field. The discussion builds upon a recent review [1] and covers the two major challenges in hybrid simulation, namely the choice of coupling method and the imposition of boundary conditions on the molecular simulation, with particular emphasis on the latter.

Here we limit ourselves to the discussion of hydrodynamics applications. Consequently, the term continuum description will be understood to refer to the Navier-Stokes set of equations. As will be discussed below, and in complete analogy with continuum numerical solution methods, compressibility has significant bearing on the chosen approach [2]. In this paper coupling of both compressible and incompressible Navier-Stokes descriptions to a molecular description will be discussed; it will thus be assumed that the appropriate description is chosen depending on the extent to which compressibility effects are important. On the molecular side, we limit ourselves to a classical molecular dynamics treatment [3] where particles move according to Newton's equation of motion

$$m_i \ddot{\mathbf{r}}_i = \mathbf{F}_i = -\sum_{j \neq i} \nabla U(r_{ij}) \tag{1}$$

where m_i, $\mathbf{r}_i$ and $\mathbf{F}_i$ are the mass, position vector and force on particle i, the latter assumed to be captured by simple pair potentials $U(r_{ij})$ (such as a Lennard-Jones), where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$. Although more complex potentials may be used, these do not affect the current discussion significantly. We will also discuss dilute gases for which molecular dynamics methods become inefficient compared to Boltzmann descriptions [4]; in this paper the "molecular description" for dilute gases will be provided by a stochastic particle method for solving the Boltzmann equation known as the direct simulation Monte Carlo [4,5].

The coupling method is the numerical strategy for exchange of information between the different physical descriptions/subdomains (molecular and continuum) in a way which ensures a seamless coupled solution. For example, the chosen coupling technique may require imposing equality of the flow velocity in the two descriptions at the matching interface or across an overlap region [6,7].

One would expect that in choosing a coupling method, the best approach would be one that makes use of the existing numerical methods literature; such an approach allows the use of already developed, robust matching schemes (e.g. domain decomposition [8]) with the added benefit that these will be immediately compatible with at least one of the subdomain formulations (the continuum problem).

This information exchange required by the coupling technique typically requires the imposition of data from one subdomain in the form of boundary conditions/constraints on the "solvers" of the other subdomain and vice versa. In the example given above, the coupling recipe of matching solutions by imposing equality of flow velocity, requires the ability to impose velocity boundary conditions on each subdomain; this may be a time-dependent (explicit coupling in time, e.g. [7]) or a time-independent (time-independent coupling, e.g. [6]) formulation.

Information transfer from the molecular to the continuum subdomain is usually a well-defined process since, in analogy to the process of extracting macroscopic fields from molecular simulations, it is typically achieved through coarse graining the molecular field to the continuum field resolution (physical and spatial) [3]. Moreover, imposition of generalized boundary constraints on the continuum domain can be conveniently formulated in terms of boundary conditions.

*e-mail: ngh@mit.edu

The reverse process is more problematic: information from the continuum subdomain is limited to hydrodynamic variables which correspond to the first few moments of the complete molecular distribution function [3] and is thus insufficient to completely specify the non-equilibrium molecular state. In other words, generating realizations of non-equilibrium molecular states that are consistent with desired hydrodynamic fields/constraints is very challenging; the constraints involved are usually of the integral type, but more importantly, the non-equilibrium molecular distribution function is in most cases unknown. The process just described, which we will refer to as imposition of boundary conditions on the molecular simulation, has, in our opinion, not been resolved in a completely satisfactory manner to date. Successful resolution of this problem will have broader impact in molecular hydrodynamics; imposing macroscopic constraints/boundary conditions on molecular simulations is not only important in hybrid method applications, but also a variety of other situations where problems of interest are formulated in hydrodynamic terms.

As pointed out in the previous review [1], boundary condition imposition on molecular simulations and the choice of coupling technique are not necessarily coupled. Although the latter determines the nature of information transfer between the suddomains, it does not necessarily constrain the way this is to be achieved. Returning to the example discussed above, the requirement that velocities are aqual at a matching interface or across an overlap region, does not constrain the way this is to be implemented on either subdomain. In fact, it is preferable to formulate the problem in such a way that these two functions are completely decoupled. This affords the most flexibilty in customizing the hybrid methodology to the physics of the problem at hand, a recommended approach for any numerical solution. As will be explained below, the choice of coupling technique is not unique and from a number of studies to date it appears that a variety of coupling techniques can be used to achieve a globally consistent solution, provided that numerical choices which respect the flow physics are made.

2. Recent developments

Before focusing on boundary condition imposition in molecular simulations we will briefly touch upon a number of other considerations, such as the role of molecular fluctuations and the choice of coupling method. Although the choice of coupling method is very important, our discussion will be brief since this topic was thoroughly discussed in [1].

2.1. Molecular fluctuations: physics or noise? The existence of a molecular subdomain in a hybrid calculation means that molecular fluctuations are an integral part of the calculation. Although molecular fluctuations sometimes lead to interesting physical phenomena [9], they are also the source of significant statistical uncertainty which makes low-noise molecular simulations very expensive. Additionally, in a recent paper, Tysanner and Garcia [10] show that molecular fluctuations in flows out of equilibrium may cause numerical artifacts if care is not taken in correctly defining hydrodynamic quantities in terms of molecular data.

In [11], analytical results for the relative statistical uncertainty as a function of samples for typical hydrodynamic quantities in low speed flows (where statistical noise presents a problem) were obtained. The relative statistical uncertainty of hydrodynamic quantity Q, E_Q, is defined as the one standard deviation in the uncertainty in the measurement of Q, divided by the magnitude of Q. These results were obtained using equilibrium statistical mechanics; the assumption of equilibrium is very reasonable for low speed flows in which the deviation from equilibrium is small. One finding of this study for dilute gas flows is that the relative statistical error in hydrodynamic flux measurement, E_f, is related to the relative statistical error in the corresponding state property measurement, E_s, by $E_f \sim E_s/Kn$, where Kn is the Knudsen number (defined as the ratio of the molecular mean free path to the characteristic flow lengthscale). This implies that in low speed gas flows, using hydrodynamic fluxes to couple the Navier-Stokes and atomistic region is at a considerable disadvantage compared to coupling using state variables since the coupling will take place in regions where $Kn \ll 1$. This result, namely that the relative statistical uncertainty in fluxes is significantly larger than the relative statistical uncertainty in the corresponding state variables, was also verified for dense fluid flows in [12].

Molecular fluctuations may affect the accuracy, stability and efficiency of the hybrid solver. For example, molecular data, if insufficiently averaged, may cause numerical instability when imposed onto a continuum solver. Another example illustrating the effect of molecular fluctuations can be found in [13] where a fully adaptive mesh and algorithm refinement scheme was presented; this scheme automatically introduces the molecular description (dilute gas in this case) as the finest level of refinement based on continuum-description breakdown criteria. Numerical experiments using this scheme showed that statistical fluctuations were responsible for spurious growth of the molecular region, even at equilibrium: this occured because spontaneous fluctuations between adjacent computational cells resulted in spurious gradients that exceeded the breakdown criteria. This resulted in the development of bounds on the number of statistical samples required to avoid spurious growth of the molecular domain [13]. A recently developed variance reduction technique [14] for dilute gases may be used if molecular fluctuations need not be retained.

If hydrodynamic fluctuations are important to the problem studied, e.g. triggering an instability [9,15], they can be retained; Alexander et al. [16,17] have demonstrated that explicit time-dependent flux-based coupling formulations preserve the fluctuation spectrum of the molecular description throughout the molecular region while the fluctuations rapidly decay into the continuum region. They also demonstrated that correct fluctuation spectra can be obtained in the entire hybrid domain by using an appropriate fluctuating hydrodynamics formulation [16–18] in the continuum subdomain.

2.2. Coupling method. The selection of coupling method has been discussed at length before [1]. The main message of this discussion is that the choice of a coupling method needs to be based on the physics of the problem of interest and not on a preset notion that a particular coupling method is more appropriate than all others. In other words, no general hybrid method that can treat all hydrodynamic problems exists. In fact, although a number of approaches may be possible, similarly to continuum numerical methods, in order to develop a robust and efficient simulation method, it is important to allow the flow physics to dictate the appropriate formulation, while the numerical implementation is chosen to cater to the particular requirements of the former.

Both molecular fluctuations discussed above and a second important consideration, namely timescale decoupling originally discussed by Hadjiconstantinou in [6], are intimately linked to the flow physics (flow speed). Timescale decoupling is required because explicit integration at the molecular timestep to the global solution time (or steady state) is very computationally expensive, if not infeasible, if the Navier-Stokes subdomain is appropriately large (leading to long global evolution timescales). This is because the molecular timestep is significantly smaller (dense fluids–MD) or at best smaller (dilute gases–direct simulation Monte Carlo (DSMC) [4]) than the Courant-Friedrich-Lewy (CFL) stability timestep [2] at comparable discretization levels.

High speed (compressible) gas flows have characteristic timescales that scale with the compressible CFL timestep which is not very different from a DSMC timestep in a dilute gas simulation, while the high speeds mean that relative statistical uncertainties are smaller [11]. On the other hand, incompressible flows have characteristic timescales that are much longer than the CFL timestep [2] and thus explicit integration at the molecular timestep is more prohibitive, while low flow speeds mean that statistical noise is significant. It appears that a coupling procedure based on an explicit (in time) control-volume-type flux matching scheme is appropriate and perhaps more convenient for a compressible flow, but not an incompressible one. In fact, a number of hybrid methods [19–21] treating compressible gas flows using explicit time integration with a finite-volume-type coupling technique have been developed as a natural extension of already existing Navier-Stokes solution methods. Such approaches have reached a reasonable maturity level. Recent developments in compressible formulations include techniques which extend the adaptive mesh refinement (AMR) concept to mesh and algorithm refinement, by including the molecular description as the finest level of refinement [13,22].

In contrast, for incompressible flows, implicit methods are required that provide solutions without the need for explicit integration in time. One such implicit method for steady state problems has been proposed by the author and collaborators for liquids [6,23]; it is based on a domain decomposition approach known as the Schwarz alternating method [8,24]. The very general nature of this framework (coupling is achieved by an iterative exchange of boundary conditions across an overlap region) makes it very flexible: although first used for liquids, it has been already extended to gas flows [1] and was recently used to simulate flow through microfluidic filters [25] yielding significant computational savings. Recent work [26] has shown that if an iterative steady state formulation is used for the atomistic description (Boltzmann equation), the inner (Boltzmann) and outer (Schwarz) iterations may be interleaved, leading to further computational savings.

As pointed out in [1], the timescale limitations discussed above have not been apparent to date because in typical *test problems* published so far, the continuum and atomistic subdomains are of the same size, i.e. of molecular dimensions. It should be recalled, however, that hybrid methods are useful when the continuum subdomain is significantly larger than the molecular subdomain. The limitations arising from timescale considerations are apparent in a recent study which uses an explicit in time coupling method to treat an incompressible steady problem [7]; according to the authors, extending the study to larger systems is limited by the need to explicity integrate the atomistic domain to the long time required for the continuum domain to reach steady state.

2.3. Boundary condition imposition. Imposing general hydrodynamic boundary conditions on a molecular simulation domain has received some attention in recent years. As stated above, this is a challenging problem with potential applications beyond hybrid methods. Examples include approaches appearing in a number of areas in multiscale simulation which incorporate molecular information into continuum frameworks, e.g. the Equation-free framework of Kevrekidis [27].

In the interest of simplicity our discussion will refer to a one-dimensional formulation (see Fig. 1); higher-dimensional implementations directly follow. Let the molecular and continuum subdomains be denoted by Ω and C, respectively, and let their common boundary be denoted by $\partial\Gamma$. Except from the most trivial flows, the net mass flux across $\partial\Gamma$ will not be zero. This means that imposition of hydrodynamic boundary conditions also implies the supply of the correct flux of molecules (positive or negative) through $\partial\Gamma$. What makes this problem so challenging is the fact that molecules must arrive at $\partial\Gamma$ in the correct state as described by the non-equilibrium molecular distribution function consistent with the hydrodynamic state to be imposed. Although coupling between the molecular and continuum description, and thus exchange of boundary conditions, should only take place in a region were both are valid, the non-equilibrium distribution function will, in general, be unknown: even in the case of a simple fluid in the Navier-Stokes limit, the non-equilibrium distribution function is only known in limiting cases, e.g. a dilute gas [4].

The most convenient way to implement the required conditions is to introduce a particle reservoir R by extending Ω into C (see Fig. 1); in R, particle dynamics may be altered in such a way that the desired boundary conditions appear on $\partial\Gamma$ under the assumption that the influence of the perturbed dynamics decays sufficiently fast and does not propagate into Ω; in other words, one assumes that the relaxation distance for both the velocity distribution function and the fluid structure is small compared to the characteristic size of Ω. One of the rea-

sons reservoirs are popular is that they provide for an overlap between the continuum and molecular subdomains; this overlap provides more opportunities for constraining the molecular flow to the "underlying" continuum flow and lends itself naturally to approaches which use overlap regions as part of the coupling approach (e.g. [6,7,28,29]); note that in these cases, rather than further extending Ω into C, the reservoir usually is part of the overlap region (see Fig. 2).

Implementation details are typically problem dependent. For example, note that in the configuration of Fig. 1 the outer edge of the molecular dynamics domain is ∂R, whereas in the configuration of Fig. 2 the outer edge of the molecular dynamics domain is $\partial\Omega$; although these differences complicate the development of a general discussion, the actual scientific challenges and solutions associated with the two configurations are very similar. We will attempt to keep our discussion as general as possible: unless otherwise stated, we will be referring to the configuration of Fig. 1.

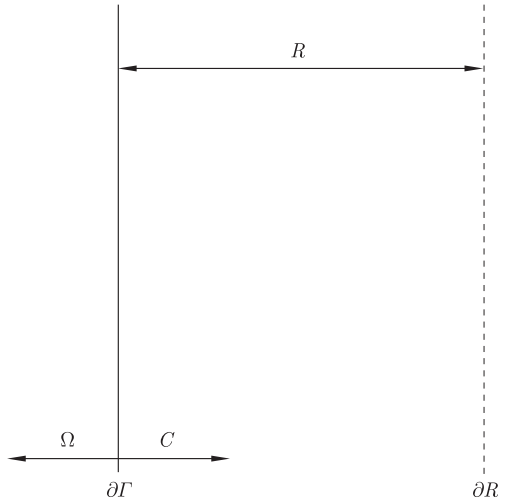

Fig. 1. One-dimensional illustration of boundary condition imposition using a reservoir R

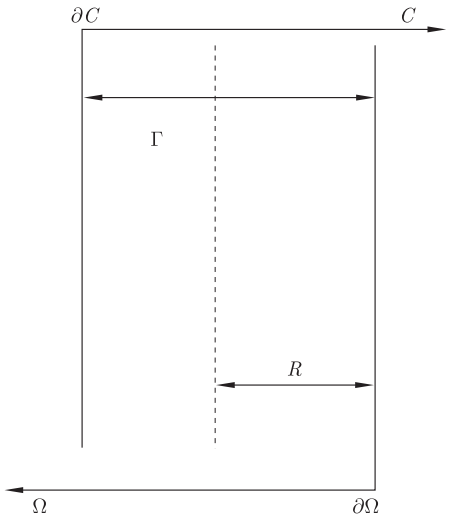

Fig. 2. One-dimensional illustration of boundary condition imposition using a reservoir R in the presence of an overlap region Γ

We will begin our discussion with dilute gases where the distribution function is known in the Navier-Stokes limit. We will then discuss dense fluids in which a number of theoretical questions remain open.

Dilute gases. In a dilute gas, the non-equilibrium distribution function in the Navier-Stokes limit has been characterized [4,30] and is known as the Chapman-Enskog distribution. It can be written as $nf(\mathcal{C})$ where n is the local number density,

$$f(\mathcal{C}) = f_0(\mathcal{C})[1 + \Phi(\mathcal{C})], \tag{2}$$

$\mathcal{C}$ is the molecular thermal velocity, $f_0(\mathcal{C})$ is the Maxwell-Boltzmann equilibrium distribution function, and $\Phi(\mathcal{C})$ is a small perturbation function parametrized by the local heat flux vector and stress tensor (the exact form of $\Phi(\mathcal{C})$ can be found in [4,30,32]). Use of this distribution to impose boundary conditions on molecular simulations of dilute gases results in a robust, accurate and theoretically elegant approach.

Typical implementations [22] require particle generation and initialization within R during every simulation timestep; these particles are drawn from the Chapman-Enskog distribution as parametrized by the local continuum solution to be imposed. Particles that move into Ω within the simulation timestep are added to the simulation whereas particles remaining in R are discarded. Particles leaving Ω to R are also discarded. Recent implementations [13,22,31] show that imposing a linearly interpolated form of the continuum field within the reservoir region provides sufficient accuracy. Generation of particles according to a linear density gradient can be achieved using a variety of methods [31], including acceptance-rejection schemes. Similarly, $f(\mathcal{C})$ can be generated using an acceptance-rejection scheme [32].

Dense fluid flows. Unfortunately, for dense fluids (even simple dense fluids) the non-equilibrium distribution function corresponding to Navier-Stokes flow has not been characterized. Due to strong molecular interactions in the dense fluid limit, describing the fluid state requires knowledge of both the molecular velocity distribution and fluid structure. Strong molecular interactions also make termination of the molecular dynamics domain significantly more challenging: the outer edge of the molecular region needs to be terminated in a way that does not have a large effect on the fluid state inside Ω.

Using a Maxwell-Boltzmann distribution to generate the velocities of molecules on $\partial\Gamma$ in non-equilibrium situations will in general lead to slip. To overcome this difficulty, Li et al. [33] used a Chapman-Enskog distribution to impose boundary conditions. They studied a simple Couette flow in which molecules crossing $\partial\Gamma$ to enter Ω acquire velocities drawn from a Chapman-Enskog distribution parametrized by the local values of the required velocity and stress boundary condition. Although this approach was only tested for a simple shear flow, it appears to give reasonable results (within molecular fluctuations).

Dirichlet boundary conditions on MD simulations can also be imposed through the method of constraint dynamics [28]. This approach has appeared in a number of methods featuring an overlap region [7,28,29]. In this approach, the required boundary condition (e.g., referring to Fig. 2, flow velocity on

$\partial\Omega$ or throughout R as specified by the continuum solution) is formulated as a constraint and introduced into the equations of motion of the particles in the constraint region. Although the particular formulation in [28] did not allow for flow across $\partial\Omega$, this feature can be integrated into this approach as shown by Nie et al. [7]; this generalization and the remaining elements of the approach by Nie et al. will be discussed below. In summary, although constraint dynamics methods that enforce the required boundary condition can be formulated, their effect on the local distribution function and thus fluid behavior is not well understood. In other words, it is not clear what the effect of the modified Newton equations obeyed by the constrained particles is on the flowfield in the remainder of Ω and the hybrid solution.

Although the choice of a time-dependent flux-based coupling motivated by a compressible formulation in the continuum subdomain is not recommended for dense fluids[1], a fair amount of work has been done in imposing flux boundary conditions on molecular simulations. Flekkoy et al. [35] achieve flux matching at $\partial\Gamma$ (referring to Fig. 1) by using external forces to control particle momentum in the reservoir. The magnitude of these forces is such that the total force on the fluid particles in the reservoir region is the one required by momentum conservation. By varying the spatial distribution of the normal component of this force in a way which prevents particle escape from ∂R, Flekkoy et al. use this force as a means of terminating the reservoir. More specifically, each particle in R is subject to a fraction of the total normal force as determined by a spatially dependent weighing factor which diverges as the particle approaches ∂R; this ensures that particles do not escape the reservoir region while particles introduced at ∂R move towards Ω. To remove the heat generated by these forces and keep the temperature constant a Langevin thermostat is used. Particles are introduced into R with velocities drawn from a Maxwell-Boltzmann distribution.

Delgado-Buscalioni and Coveney [36] extended this approach to impose both momentum and energy fluxes by using an Usher algorithm to insert particles in R with the desired specific energy, which is beneficial to imposing the desired heat flux; the Usher algorithm facilitates particle insertion at the desired location of the energy landscape while eliminating the risk of particle overlap in physical space (at some computational cost). Inserted particle velocities are again drawn from a Maxwell-Boltzmann distribution. In the presense of a heat flux the temperature is no longer constant in R; instead, the requisite temperature gradient is imposed by a small number of thermostats placed at stations in the direction of the gradient. Although this technique appeared to be successful at imposing flux boundary conditions with moderate error, recent work [37] reports stability problems and the appearance of discontinuities in state properties across $\partial\Gamma$. In response to this, a new method [37] which uses a minimum entropy production criterion to formulate the imposition of momentum and energy fluxes was proposed. This approach uses the fact that mass, momentum and energy added to the reservoir region will flow to Ω through $\partial\Gamma$ assuming that the associated inertias of R are small. The particles in R are acted upon by a force field designed to add the desired energy and momentum to them while R stays close to equilibrium. Although the equilibrium requirement limits the arbitrary choice in reservoir force, it is not clear that it is a hydrodynamically consistent choice for the state of R. Although it is well-known that the deviation from equilibrium under Navier-Stokes conditions is very small (this can be verified for a dilute gas by inserting typical values in $\Phi(\mathcal{C})$), an algorithm which promotes local equilibrium (tests in [37] show moderate deviation therefrom) cannot guarantee that the desired non-equilibrium molecular state is achieved in R; this, in turn, implies that the transport properties of the fluid may be inconsistent with Navier-Stokes behavior in R. In other words, even if the flux across $\partial\Gamma$ is correct (assuming zero inertia for R) consistent behavior of the state variables on $\partial\Gamma$ and in the neighborhood cannot be guaranteed. It is possible that the resulting discrepancy is small; this may be clarified when this approach is used in a hybrid method.

Although the non-unique choice of force fields and local Maxwell-Boltzmann distributions is not very theoretically pleasing, the use of forces in the reservoir region has been adopted by a number of researchers as a way of controlling particle motion (both domain termination and flow properties). For example, the recent work of Nie et al. [7] combines constraint dynamics to impose Dirichlet (state variable) boundary conditions with a force field which prevents particles from escaping the reservoir region.

The application of a normal force to control particles escaping Ω (referring to Fig. 2) has recently been put on a firmer theoretical footing by Werder et al. [29] who recognize that the origin of this force is the particles that would reside in a homogeneous and infinite extension of the molecular region[2]. In other words, the force on a particle close to $\partial\Omega$ receives two contributions: the first is the one explicitly calculated in the simulation, namely the force from all particles in the simulation within the potential cutoff; the second is the contribution of all particles beyond the molecular domain termination, $\partial\Omega$, which are within the potential cutoff. The key lies in the realization that the spatial distribution of this force can be re-created through knowledge of the fluid structure which is contained in the radial distribution function [3]. Although the fluid radial distribution function is not known away from equilibrium, one can obtain a good approximation to this average force by using the equilibrium radial distribution function. Simulation tests [29] show that using this approach results in a significantly improved description of the fluid state in Ω (close to $\partial\Omega$). Since Werder et al. do not prevent the particles from escaping from Ω but rather focus on providing the correct long-

[1] See [2] for a discussion of challenges resulting from application of compressible solution methods to essentially incompressible flowfields; see [1], as well as Section 2.2, for a discussion of timescale limitations arising from the above choice; see [1,29,34] for a discussion of the prohibitive statistical noise in flux quantities compared to corresponding state variables.

[2] Their approach also requires a very small reservoir region whose thickness is one averaging cell width.

range force field, specular reflection at $\partial\Omega$ is used to contain particles in Ω, while an Usher algorithm is used for introducing new particles in the case that there is a net mass inflow to Ω.

As stated above, the need to impose boundary conditions on molecular simulations does not only arise in hybrid methods. Recent developments in multiscale simulation have resulted in a number of simulation methods which incorporate molecular information in a number of ways. One example is the Equation-free framework of Kevrekidis et al. [27] which uses information obtained from microscopic (molecular) solvers over small periods of time and small spatial domains to integrate macroscopic equations over large domains and long times. Performing these simulations requires the ability to initialize and subject molecular systems to boundary conditions derived from macroscopic fields (for example from the previous macroscopic timestep). In the area of initialization, progress has been made [38] in cases where a separation of timescales exists between the microscopic dynamics and macroscopic dynamics of interest. In a similar fashion, in the case of spatially smooth solutions, it is shown in [39] that appropriately initialized reservoir regions will provide boundary conditions for sufficient time for the problem to be integrated forward in time for a special class of homogenization problems. Although the assumptions of smoothness at macroscopic scales makes these techniques applicable to different classes of problems, communication and idea exchange between the various multiscale simulation disciplines may prove to be very useful in the future.

2.4. Validation. Hybrid methods are typically validated by comparing their results with fully molecular simulations of the same problem. This limits validation problems to small physical domains, since a fully molecular solution needs to be feasible. As discussed in Section 2.2 this practice has masked the issue of timescale discrepancy between the molecular dynamics integration timestep and the timescale of the nominally large continuum subdomain. Typical comparisons are limited to hydrodynamic fields suitably (ensemble) averaged to reduce the statistical uncertainty to levels permitting meaningful comparisons; recent work on the role of fluctuations has extended comparisons to other statistical moments of these fields, such as the variance and spatial correlations [16,17].

Unfortunately, the numerical error associated with hybrid procedures is not well characterized, in part due to the statistical uncertainty associated with molecular fluctuations and the large computational cost associated with its removal [11] which makes convergence studies very challenging. Using a coupling method for which consistency can be shown and numerical error estimates for finite discretization can be derived, can help eliminate or bound the error resulting from the coupling procedure. Other sources of error include the approximation inherent in the boundary condition imposition methods discussed in Section 2.3 which remains largely uncharacterized, and the effect of molecular fluctuations on the continuum solver. A final source of error results from "incompatibilities" between the continuum and molecular models; these may arise from a number of factors, such as molecular effects present in the molecular description but neglected in the continuum model (e.g. shear thinning) or statistical uncertainty in the properties (e.g. transport coefficients) of the molecular fluid.

Incompatibility between the two models also arises in connection with the breakdown of the continuum description and the choice of location for the matching interface. Breakdown of the continuum description usually means that the discrepancy between the continuum and molecular model is expected to exceed some error tolerance. In this sense, even a conservative placement of the matching interface may still imply that matching is occuring in regions where some incompatibility between the two descriptions exists, especially in situations where the limits of the continuum description are not well characterized.

3. Final remarks

It appears that although from a theoretical point of view the imposition of boundary conditions on molecular simulations remains an open problem, significant progress has been made in developing schemes which exhibit small numerical error in imposing the desired boundary conditions. One exception to this observation is the dilute gas case where the Chapman-Enskog distribution provides a robust and accurate method for imposing boundary conditions. As in the case of continuum numerical schemes, in the presence of continuous progress, flexibility in adopting appropriate elements from previous approaches is a key step to the development of more sophisticated, next-generation hybrid methods. Work on adaptive algorithm refinement methods [13] has shown that these require robust criteria for Navier-Stokes or continuum description breakdown [4,13] that are both physically accurate but also insensitive to molecular fluctuations.

The importance of choosing the coupling method based on flow physics cannot be overstated. Solution of appropriately large, time-dependent problems is still not possible if the molecular domain needs to be explicitly integrated for the total time of interest. Iterative steady-state solution frameworks which decouple timescales have been successfully developed for low speed, incompressible flows and should be used when appropriate. For large-scale time-dependent flows, new frameworks are required which allow timescale decoupling; alternatively, the resolution may lie in the development of coarse grained molecular simulation methods. Progress in this direction has been made with dissipative particle dynamics methods [40]. In the case where separation of timescales exists between the atomistic region dynamics and the global evolution timescale, projective techniques [27] may be used to propagate the hybrid field forward in time, following brief explicit integration during which the two descriptions are coupled.

Acknowledgments. The author wishes to thank H.S. Wijesinghe, R. Hornung, A.L. Garcia, L. L. Baker, I.G. Kevrekidis, A.T. Patera and B.J. Alder for helpful comments and discussions.

REFERENCES

[1] H.S. Wijesinghe and N.G. Hadjiconstantinou, "Discussion of hybrid atomistic-continuum methods for multiscale hydrodynamics", *International Journal for Multiscale Computational Engineering* 3, 189–202 (2004).

[2] P. Wesseling, *Principles of Computational Fluid Dynamics*, Springer, 2001.

[3] M.P. Allen and D.J. Tildesley, *Computer Simulation of Liquids*, Oxford Science Publications, 1987.

[4] G.A. Bird, *Molecular Gas Dynamics and the Direct Simulation of Gas Flows*, Clarendon Press, Oxford, 1994.

[5] F.J. Alexander and A.L. Garcia, "The direct simulation Monte Carlo method", *Computers in Physics* 11, 588–593 (1997).

[6] N.G. Hadjiconstantinou, "Hybrid atomistic-continuum formulations and the moving contact-line problem", *Journal of Computational Physics* 154, 245–265, (1999).

[7] X. Nie, S. Chen and M.O. Robbins, "Hybrid continuum-atomisitc simulation of singular corner flow", *Physics of Fluids* 16, 3579–3591 (2004).

[8] P.L. Lions "On the Schwarz alternating aethod. I.", *First International Symposium on Domain Decomposition Methods for Partial Differential Equations*, eds. R. Glowinski, G. Golub, G. Meurant and J. Periaux, SIAM, Philadelphia, 1–42 (1988).

[9] M. Moseler and U. Landman, "Formation, stability, and breakup of nanojets", *Science* 289, 1165–1169 (2000).

[10] M.W. Tysanner and A.L. Garcia, "Non-equilibrium behaviour of equilibrium reservoirs in molecular simulations", *International Journal for Numerical Methods in Fluids* 48, 1337–1349 (2005).

[11] N.G. Hadjiconstantinou, A.L. Garcia, M.Z. Bazant, and G. He, "Statistical error in particle simulations of hydrodynamic phenomena," *Journal of Computational Physics* 187, 274–297 (2003).

[12] T.U. Werder, "Multiscale simulations of carbon nanotubes in aqueous environments", *Ph.D. Thesis*, Swiss Federal Institute of Technology, Zurich, 2005.

[13] H.S. Wijesinghe, R. Hornung, A.L. Garcia, and N.G. Hadjiconstantinou, "Three-dimensional hybrid continuum-atomistic simulations for multiscale hydrodynamics", *Journal of Fluids Engineering* 126, 768–777 (2004).

[14] L.L. Baker and N.G. Hadjiconstantinou, "Variance reduction for Monte Carlo solutions of the Boltzmann equation", *Physics of Fluids* 17, Art. no. 051703 (2005).

[15] K. Kadau, T.C. Germann, N.G. Hadjiconstantinou, P.S. Lomdahl, G. Dimonte, B.L. Holian, and B.J. Alder, "Nanohydrodynamics simulations: an atomistic view of the Rayleigh-Taylor instability", *Proc. of the National Academy of Sciences* 101, 5851–5855 (2004).

[16] F.J. Alexander, A.L. Garcia, and D. Tartakovsky, "Algorithm refinement for stochastic partial differential equations: I. Linear diffusion", *Journal of Computational Physics* 182, 47–66 (2002).

[17] F.J. Alexander, A.L. Garcia, and D. Tartakovsky, "Algorithm refinement for stochastic partial diffential equations: II. Correlated systems", *Journal of Computational Physics* 207, 769–781 (2005).

[18] L.D. Landau and E.M. Lifshitz, *Statistical Mechanics*, Part 2, Pergamon Press, Oxford, 1980.

[19] D.C. Wadsworth and D.A. Erwin, "One-dimensional hybrid continuum/particle simulation approach for rarefied hypersonic flows", *AIAA Paper* 90–1690 (1990).

[20] D.C. Wadsworth and D.A. Erwin, "Two-dimensional hybrid continuum/particle simulation approach for rarefied hypersonic flows", *AIAA Paper* 92–2975, 1992.

[21] R. Roveda, D.B. Goldstein, and P.L. Varghese, "Hybrid Euler/direct simulation Monte Carlo calculation of unsteady slit flow", *Journal of Spacecraft and Rockets* 37, 753–760 (2000).

[22] A.L. Garcia, J.B. Bell, W.Y Crutchfield, and B.J. Alder, "Adaptive mesh and algorithm refinement using direct simulation Monte Carlo", *Journal of Computational Physics* 54, 134–155 (1999).

[23] N.G. Hadjiconstantinou and A.T. Patera, "Heterogeneous atomistic-continuum representations for dense fluid systems", *International Journal of Modern Physics C* 8, 967–976 (1997).

[24] M. Fortin and R. Aboulaich, "Schwarz's decomposition method for incompressible flow problems", *First International Symposium on Domain Decomposition Methods for Partial Differential Equations*, eds. R. Glowinski, G. Golub, G. Meurant and J. Periaux, SIAM, Philadelphia, 333–349 (1988).

[25] O. Aktas and N.R. Aluru, "A combined continuum/DSMC technique for multiscale analysis of microfluidic filters", *Journal of Computational Physics* 178, 342–372 (2002).

[26] L.L. Baker and N.G. Hadjiconstantinou, "Implicit hybrid simulation framework for steady-state dilute gas flows", *International Journal for Multiscale Computational Engineering* 3, 49–58 (2005).

[27] I.G. Kevrekidis, C.W. Gear, J.M. Hyman, P.G. Kevrekidis, O. Runborg, and C. Theodoropoulos, "Equation-free, coarse-grained multiscale computation: enabling microscopic simulators to perform system-level analysis", *Communications in Mathematical Sciences* 1, 715–762 (2003).

[28] S.T. O'Connell and P.A. Thompson, "Molecular dynamics-continuum hybrid computations: a tool for studying complex fluid flows", *Physical Review* E 52, R5792–R5795 (1995).

[29] T.U. Werder, J.H. Walther and P. Koumoutsakos, "Hybrid atomistic-continuum method for the simulation of dense fluid flows", *Journal of Computational Physics* 205, 373–390 (2005).

[30] S. Chapman and T.G. Cowling, *The Mathematical Theory of Non-uniform Gases*, Cambridge University Press, 1970.

[31] H.S. Wijesinghe, "Hybrid atomistic-continuum formulations for gaseous flows", *Ph.D. Thesis*, Department of Aeronautics and Astronautics, Massachusetts Institute of Technology, 2003.

[32] A.L. Garcia and B.J. Alder, "Generation of the Chapman-Enskog distribution", *Journal of Computational Physics* 140, 66–70 (1998).

[33] J. Li, D. Liao, and S. Yip, "Nearly exact solution for coupled continuum/MD fluid simulation", *Journal of Computer-Aided Materials Design* 6, 95–102 (1999).

[34] R. Delgado–Buscalioni, E.G. Flekkoy, and P.V. Coveney, "Fluctuations and continuity in particle-continuum hybrid simulations of unsteady flows based on flux-exchange", *Europhysics Letters* 69, 959–965 (2005).

[35] E.G. Flekkoy, G. Wagner, and J. Feder, "Hybrid model for combined particle and continuum dynamics", *Europhysics Letters* 52, 271–276 (2000).

[36] R. Delgado–Buscalioni and P.V. Coveney, "Continuum–particle hybrid coupling for mass, momentum and energy transfers in unsteady fluid flow", *Physical Review* E 67, (article No) 046704 (2003).

[37] E.G. Flekkoy, R. Delgado–Buscalioni, and P.V. Coveney, "Flux boundary condtions in particle simulations", *Physical Review* E 72, (article No) 026703 (2005).

[38] C.W. Gear and I.G. Kevrekidis, "Constraint-defined manifolds: a legacy-code approach to low-dimensional computations", submitted to *Journal on Scientific Computing*; (can be found as physics/0312142 at arXiv.org).

[39] G. Samaey, D. Roose and I.G. Kevrekidis, "The gap-tooth scheme for homogenization problems", *Multiscale Modeling and Simulation* 4, 278–306 (2005).

[40] P. Espanol, "Hydrodynamics from dissipative particle dynamics", *Physical Review* E 52, 1734–1742 (1995).