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Advancements in multi scale modeling: Adaptive resolution simulations and related issues

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Abstract. Adaptive resolution methods are becoming increasingly important in the study of complex systems by multi scale modeling. In this paper we present a brief overview of the method and highlight some questions that in our opinion are relevant for the future development of the method, and more in general of the field of multiscale modeling.

Research in complex systems requires the use of simulation methods that provide high-quality resolution on a variety of lengthscales. These properties develop on such a wide range of lengthscales that they cannot be covered by simple brute-force atomistic simulations. It is then convenient to physically partition the ensemble of particles simulated into sub-ensembles that are described with different resolution. By this scheme, the so-called Adaptive Resolution Simulation (AdResS) method, it is possible to considerably extend the range of physical processes that can be simulated [1,2]. Information collected locally at the atomistic scale is used to inform the computation of large-scale processes which is described at lower resolution. The computational gain is obtained by the use of the large-scale low-resolution descriptions, which speed up simulations. The low resolution coarse-grained regions often act as a bath to the atomistic local simulation when the volume of the low resolution region is much larger than the volume of the high resolution region. In this way, the high resolution region represents a grand canonical ensemble, while the whole simulation box represents a canonical ensemble.

1 Optimizing accuracy in AdResS simulations

From the computational side, an important goal is to be able to design AdResS formalisms that exchange information rapidly and reliably between the different levels of resolution while maintaining accuracy in the structural, thermodynamical, and possibly dynamical properties. Maintaining consistency between the resolution levels is not trivial, and in the present state of development of the method, the degree of consistency of the AdReS simulations depends both on the type of AdResS approach

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that is chosen and on the compatibility of the different models adopted in the regions that have different resolution.

Typically a simulation box is composed of two regions that have different granularity and a hybrid region where thermodynamic, dynamic, and molecular descriptions are slowly transitioned from one resolution to the other by means of a switching function. A Langevin thermostat is applied in the hybrid region to compensate the energy change due to the insertion or deletion of the degrees of freedom during the transition of a molecule between the two resolution regions.

In principle AdResS simulations should allow for the change of resolution "on the fly" as the molecules diffuse between regions with different granularity. At any given time, a molecule can be situated at the interface between two different resolution regions: in that case part of the molecule can be defined with a level of precision that is different from another part of the same molecule. This structural dis-homogeneity affects the dynamics and thermodynamics of the molecule in an important way. It is a time-dependent property as it evolves in time, depending on the molecular diffusion.

2 Physical quantities that are, or are not, conserved in AdResS: Balancing forces vs. balancing potentials

Adaptive resolution methods build on the choice of the model selected to bridge between the different resolution regions, and on the simulation models used in those regions. Depending on this choice specific physical properties are consistent with an equivalent fully atomistic simulation and others are not. In general, physical quantities cannot be conserved when the whole system is considered, but they can be conserved in the local atomistic region.

In general the goal of AdResS is to predict properties that are consistent in the high resolution region, while the low-resolution region acts as a "bath". In developing an AdResS method it is important to understand how to obtain a correct exchange of information between levels, to know which properties are conserved and which are not conserved across the different resolutions, and how the transferability of the AdResS description depends on the choice of the coarse-graining method. Some of these points are discussed in this volume.

Adaptive resolution approaches are of two broadly defined types where the first is concerned with deriving directly an equation for the balance of the forces in the system that contains two different levels of resolutions [1,2]; the second instead balances an effective Hamiltonian. The effective Hamiltonian is a free energy of the system, and depends on all the thermodynamic and molecular parameters that characterize the system simulated [3]. In the AdResS method the hybrid region is forced to reproduce uniform density across the simulation box, two and three-body distribution functions, the pressure, and the chemical potential of the atomistic region by adding the needed balancing forces as extra contributions. The extra balancing forces are calculated via an iterative procedure until convergence is obtained in each related structural and thermodynamic quantity. The inclusion of these forces that are space dependent implies that the total force is not conservative and so it is impossible to derive from the forces a Hamiltonian that conserves the energy. If an interpolation scheme is used for the Hamiltonian the force is not formally consistent with the balance of forces that ensures conservation of structure and thermodynamics across the entire simulation box [4].

In this volume, the paper by Wang and Agarwal explains in detail the inconsistencies between force-balanced and energy-balanced AdResS methods [4,5]. In the hybrid region, the conserved quantity is slowly switched on as the system includes an increasing number of degrees of freedom. This switch function depends on the degrees

of freedom and on the position of the molecule as it is transitioning between spatial regions that have different resolution. As a consequence, balancing the force does not conserve the energy and balancing the Hamiltonian does not conserve the forces. By selecting the formalism adopted in the adaptive resolution simulation, one is forced to assume non conservation of the related physical quantities that are commonly conserved in the more traditional molecular dynamic simulations where the resolution is fixed.

3 AdResS: Combining quantum with classical simulations

As pointed out by Wang and Agarwal [5], AdResS is intended to be a very general method with a flexible "container-like" formal structure. AdResS bridges simulations between representations that have different levels of resolution: this can be intended in a very general way as combining, for example path integral and atomistic models, or atomistic and coarse-grained models, or even coarse-grained models and a continuum approach.The theory is not limited to two regions and the hybrid interface, but it can be seen as describing the transition between multiple regions. An example is presented in the paper by Delgado-Buscalioni, Sablić, and Praprotnik, where the authors propose an AdResS method for molecular dynamic simulations with open boundaries [6]. In one realization of the open boundary molecular dynamics the atomistic subsystem is surrounded by the coarse-grained representation, which is still contained in a volume of fixed size, which is embedded in a continuum bath. By this scheme large molecules can be inserted in the atomistic region, as the surrounding coarse-grained region is open to the continuum exterior.

The success of an AdResS formalism often relies on the quality of the force field used in the simulations. It is important to use a reliable atomistic force field. Given the many years and the considerable effort spent in optimizing force fields for atomistic computer simulations by comparing results with experimental data, it is well-known that force fields are still lacking precision and consistency. Because present force fields predict a variety of most-probable configurational states, many problems that depend on the search of the most stable configuration achieve, slightly, different results depending on the force field utilized in the simulation [7]. With the rapid advancements in computer power, which allows routine access to microsecond timescale in atomistic simulations, the importance of developing highly reliable force fields consistent with experiments is becoming increasingly more evident.

In their paper, Bonella and Ciccotti present a broad discussion of the current approaches that bridge quantum to classical dynamics [8]. The authors identify a number of key issues that need to be addressed to progress the field of simulations at the interface between quantum and classical descriptions. Mixed quantum classical methods are roughly divided in two groups. The first is formed by numerically efficient but poorly justified ad hoc methods like Surface Hopping. The second contains rigorous but computationally expensive methods such as Liouville and LAND-Map. The first group can be used for larger systems than the rigorous theory, which is computationally expensive. Research at the interface between classical and quantum resolution is focusing on both finding a more rigorously justified form of ad hoc methods and in speeding up rigorous methods by adopting some level of approximation. Recent research is developing tools that can assess the accuracy of the methods when mixed approaches are used. These tools allow one to formally bridge between more phenomenological and more rigorous theories opening new avenues for the AdResS method at the interface with quantum mechanics. For a first attempt in this direction see the recent paper by Poma and Delle Site [9].

4 Physical quantities that are, or are not, conserved in AdResS: The choice of the coarse-grained model

The accuracy of the AdResS method depends also on the coarse-grained model that is used in the low-resolution region. For polymer liquids, coarse-grained models based on integral equation theories in the canonical ensemble are known to conserve excess free energy, structure, compressibility, and equation of state if properly solved [10,11]. However they do not conserve the internal energy nor the entropy [11]. They also do not conserve the dynamics, which has to be reconstructed *a posteriori* [12]. Dissipative particle dynamics, on the other hand, has been designed to conserve hydrodynamics by preserving momentum conservation, but it does not ensure consistency in the structural and thermodynamic properties [13]. Pressure is typically inconsistent in coarse-grained systems whose potential is derived using Iterative Boltzman Inversion methods or Force Matching, which are designed to reproduce the structure (pair distribution function) or the derivative of the potential of mean-force, which is the logarithm of the pair distribution function (force matching), but not the thermodynamics or the dynamics [14,15]. In practice, models that reproduce numerically the pair distribution function preserve the compressibility, but cannot ensure that either pressure or the equation of state are preserved. Numerical consistency of the pair distribution function and higher order distribution functions can still lead to inconsistency in the equation of state.

In general, however, the inconsistency in structural, thermodynamic, or dynamic properties inside an AdResS system is minimized if the level of coarse-graining is very limited [16]. This explain the success of AdResS when combined to the Martini model, for example, where the coarse-graining is limited to the grouping of four atomic sites [17]. While the error in the IBI method and in the Martini coarse-grained model is small because of the contained extent of coarse-graining, and the discrepancy with the full atomistic simulation can be easily corrected by adding effective forces in the hybrid region, the computational gain is also limited.

5 Conclusion and outlook

Adaptive resolution methods are useful and powerful computational schemes to simulate efficiently complex systems across multiple lengthscales, while maintaining different levels of resolution at the same time in the simulation box. One can envision in the long run, the existence of an adaptive resolution method that informs itself during the simulation on the temporary need of enhancing resolution locally on a specific spatial region of the simulation to collect the information needed to perform subsequent steps of the low resolution trajectories. The AdResS method should be capable of making decision about the need of collecting information, to act upon its own decision, and to evaluate when enough information is collected at a given resolution to revert to a different resolution. In a way, we could call this a machine-learning process [18].

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