Extending accelerated molecular dynamics methods to larger and more complex systems

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Many important materials processes take place on time scales that vastly exceed the nanoseconds accessible to molecular dynamics simulation. Typically, this long-time dynamical evolution is characterized by a succession of thermally activated infrequent events involving defects in the material. In the accelerated molecular dynamics (AMD) methodology, known characteristics of infrequent-event systems are exploited to make reactive events take place more frequently, in a dynamically correct way. For certain processes, this approach has been remarkably successful, offering a view of complex dynamical evolution on time scales of microseconds, milliseconds, and sometimes beyond. Examples include metallic surface diffusion and growth, radiation damage annealing processes, and carbon nanotube dynamics. In this talk, I will discuss some recent advances that are extending the range of applicability of the AMD methods to larger and more complex systems. For example, we now understand that the parallel replica dynamics method can give exact state to state dynamics even when the state definitions are not necessarily Markovian, provided the dephasing stage is continued long enough [1]. This provides a clean framework for treating more complex systems where it is difficult to define simple, deep states, or where it is desirable to lump many basins together into one superstate. We have also formulated a new, local version of the hyperdynamics method that gives constant boost as the system size is increased, in contrast to standard hyperdynamics, for which the boost decays towards unity as the system size is increased. We have also been exploring the limiting boost that is possible with hyperdynamics when the bias potential can be designed in an ideal way.

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Bifurcation diagram and thermally assisted magnetization reversals in spin−torque driven nanomagnets

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Driving nanomagnets by spin−polarized currents offers exciting prospects in magnetoelectronics, but the response of the magnets to such currents remains poorly understood because they apply a nonconservative force, called spin−transfer torque (STT), on the system. Here we investigate the effect of STT in various parameter regime. In the low−damping regime, we show that an averaged equation describing the diffusion of energy on a graph captures the dynamics of these magnets. From this equation, we can obtain the bifurcation diagram of the magnets, including the critical currents to induce stable precessional states and magnetization switching, as well as the mean times of thermally assisted magnetization reversal in situations where the standard reaction rate theory of Kramers is no longer valid. These results match experimental observations and give a theoretical basis for a Neel−Brown−type formula with an effective energy barrier for the reversal times. For higher values of the damping, results from large deviation theory can be used to identify the pathway and rate of switching of these magnets when the amplitude of the thermal noise is small.

Sampling saddle points on the free energy surface

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Efficient exploration of the free energy surface (FES) of a complex system at finite temperatures, though often desirable, remains a difficult task. Such explorations involve searching for metastable states and bot- tlenecks for transitions between these metastable states in the space of coarse grained variables. In this talk, I will present an algorithm for finding saddle points on the free energy surface "on-the-fly" without having to find the free energy function itself. This is done by using the general strategy of the heterogeneous multi- scale method, applying a macro-scale solver, here the gentlest ascent dynamics algorithm, with the needed force and Hessian values computed on-the-fly using a micro-scale model such as molecular dynamics. The algorithm is capable of dealing with problems involving many coarse-grained variables. The utility of the algorithm is illustrated by studying the saddle points associated with (a) the isomerization transition of the alanine dipeptide using two coarse-grained variables, specifically the Ramachandran dihedral angles, and (b) the beta-hairpin structure of the alanine decamer using twenty coarse-grained variables, specifically the full set of Ramachandran angle pairs associated with each residue.

Enhanced sampling of rare events in molecular dynamics simulations by nonlinear manifold learning

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Molecular Dynamics (MD) simulation is broadly used across various disciplines, including Materials Science, Chemistry and Biology. Continuous improvements of computational resources enable MD simulations of increasingly complex systems. However, the phenomenal challenge of bridging between the femtosecond time scale of molecular vibrations and millisecond-and-up time scale of conformational transitions remains, and calls for multiscale approaches for of complex molecular systems.

This time-scale disparity in systems with metastability makes it very difficult to obtain accurate sampling, and consequently hinders the connection between simulations and experiments. To overcome this issue, a number of enhanced sampling methods have been proposed, such as Metadynamics [1] and Adaptive Biasing Force [2]. However, the effectiveness of these methods relies on a good set of collective variables (CVs), which need to compactly characterize molecular conformations, capture metastability, and be differentiable. Identifying such a coarse model for the system is far from obvious for complex systems.

We present here a general method, Smooth and Nonlinear Data-driven Collective Variables (SandCV) [3], based on machine learning techniques to identify such CVs from available computational or experimental ensembles, and integrate them in enhanced sampling methods. SandCV is a versatile method and can be non-intrusively combined with the available molecular dynamics implementations. We show how this methodology can deal seamlessly with intrinsic manifolds of complex topology described with multiple parametrization patches.

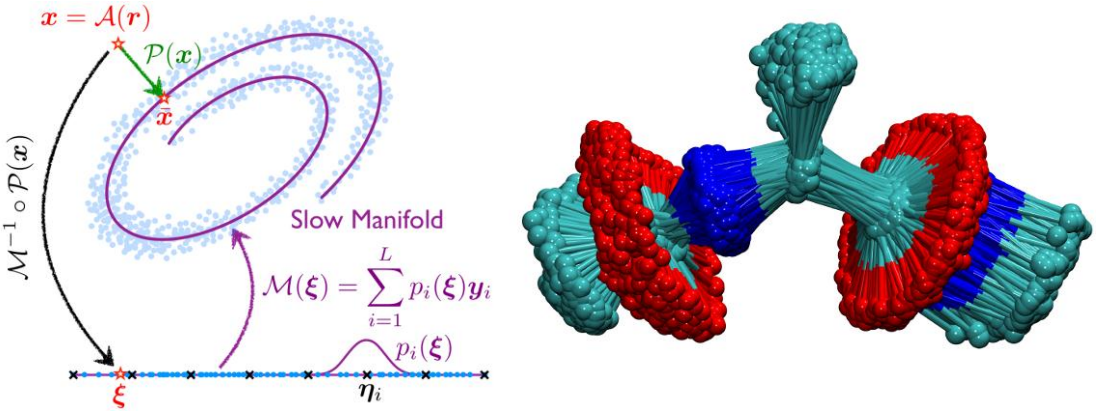

Figure 1: Left: schematic representation of SandCV algorithm. Right: High-dimensional configuration of alanine dipeptide as a benchmark molecule with two collective variables.

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Numerical methods for high-dimensional problems in computational materials science

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In this talk we present our recent research on numerical methods for high-dimensional problems with applications in the field of atomistic modeling and rational material design. In order to develop new materials with desired properties, theoretical support to predict the relationship between kinds of elements, structures and functions is necessary. Note that recent advances in high performance computing made it possible to perform large scale computational screenings and to obtain very accurate descriptions of complex quantum electronic interactions in the form of potential energy hypersurfaces. However, a global exploration (and approximation) of this surface is very complex due to its high dimensionality. Here, state-of-the-art methods from scientific computing open the possibility to circumvent this so-called curse of dimensionality. This way a large-scale computational screening, e.g. to generate new potential models, will become feasible. We will discuss the application of a hierarchy of state-of-the-art deterministic and stochastic methods known from the field of high dimensional problems for approximation, dimension reduction and global and local optimization. In particular, we will give details on sparse grid based techniques, which, under certain regularity conditions, allow to circumvent the curse of dimensionality at least to some extent. Furthermore, we will discuss the application of high-dimensional approximation and optimization techniques to automatically and efficiently generate appropriate potential models.

A Path Factorization Approach to Multiscale Stochastic Simulations

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A common issue limiting the efficiency of stochastic simulation algorithms is the numerical slowing-down that results from the occurrence of metastable states. Here, an exact algorithm based on a path factorization of the linearized propagator is proposed and applied to anomalous diffusion and to the precipitation of impurities via vacancy diffusion, two model systems in which metastability is important. Paths escaping from the metastable basins can be directly generated owing to the algorithm, which results in a dramatic acceleration of the simulations.

Overcoming Temporal and Spatial Multiscale Challenges in Materials Modeling and Computing

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Predictive computational models for materials must include the interactions of multiple spatial and temporal scales. These scales can be classified as microscopic, mesoscopic, and macroscopic for simplicity, but there are typically structures at many scales with varying degrees of separation. The promise of technologies based on engineered high performance materials make materials modeling an increasing focus of research and development.

Temporal and spatial multiscale challenges appear because the material response of crystalline solids is characterized by the nucleation, dynamics, and pattern formation of defects such as point defects (vacancies, interstitials, impurities), line defects (dislocations), and surface defects (grain boundaries, surfaces, cracks, etc.). To overcome these challenges and reach mesoscopic scales, we will present new mathematical foundations and computational algorithms for spatial coarse-graining (atomistic-to-continuum coupling methods) and temporal coarse-graining (accelerated dynamics methods), as well as methods combining spatial and temporal coarse-graining.