Generalizations of the Parallel Replica Dynamics method for long-timescale atomistic simulations

Predictive Simulation and Design of Materials by Quasicontinuum and Accelerated Dynamics Methods/Developing a Petascale Simulation Framework for Stress Corrosion Cracking

Danny Perez
Los Alamos National Laboratory
Los Alamos, NM, 87545

Abstract

One of the most common tools used to investigate the atomic-scale behavior of materials is to simply integrate the equations of motion of the system, i.e., to do molecular dynamics (MD). While this simple approach proves to be extremely powerful and predictive, its practical usefulness is often limited by the rather short timescales that can directly be simulated (about a microsecond or less). Because of this, many important but slow processes, e.g., the diffusion of defects and impurities or the initiation of plastic deformation in solids, are not amenable to direct simulation and have to be investigated using approximate methods. Over the last 15 years, so-called accelerated MD (AMD) methods were proposed to mitigate this limitation and allow for direct, high fidelity, MD simulations over significantly extended timescales.

In this work, we turn our attention to one of the AMD methods, namely the Parallel Replica Dynamics (ParRep) method. The core idea of this technique is to use multiple processors to parallelize an MD simulation in the time domain, which allows for the simulation of the evolution of small systems, whose dynamics are composed of a succession of rare events, over very long timescales. In the original formulation of ParRep, an event would be defined as the passage from a given basin of attraction of the potential energy surface to one of its neighbors. Here, we formally show that ParRep can be generalized to arbitrary partitions of configuration space and demonstrate how the error in the long-time dynamics can be controlled to arbitrary accuracy. This generalization opens the door to sophisticated time-wise multi-scale schemes where the partition of configuration space can be defined or adapted in order to maximize efficiency.

By leveraging the same core concepts, we then show that ParRep can be further generalized to handle systems where some component of the dynamics does not occur through a sequence of rare events, but, e.g., in a diffusive fashion. An example of such a system would be the diffusion and reaction of corrosive agents in a solution in contact with a solid substrate. While the evolution of the solid atoms and the corrosive reaction are indeed rare events, transport through the solution is not. This generalization significantly extends the range of applicability of ParRep and establishes it as the method of choice to accurately simulate the evolution of complex multi-timescale systems over long timescales.