Multiscale Detection of Temporal Features in Molecular Dynamics Simulations: Applications to Acceleration Methods

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ABSTRACT

Extending the time scales accessible through direct molecular dynamics simulations is an area of active research. New acceleration techniques promise to achieve experimentally relevant time scales. A common ingredient in many of these acceleration schemes is the clear identification of the current state of a system in the presence of strong thermal noise. Furthermore, the detection of transitions, and precise determination of transition times, also presents challenges.

We exploit a wavelet-based technique for detecting temporal features in molecular dynamic simulations, including the detection of meta-stable structures and transition events. The properties of a wavelet basis allow the detection of features over multiple time scales. The details of our implementation allow the detection of temporal features to be computationally efficient and possible during an ongoing molecular dynamics simulation.

Keywords: molecular dynamics, infrequent events, multiresolution analysis, wavelet analysis

1 INTRODUCTION

Molecular dynamics (MD) simulations can study important physical processes such as the evolution of material defects. The advantage of MD methods is that no *a priori* assumptions need be made regarding the relevant mechanisms. Currently the total simulation time accessible with conventional MD simulations remains limited to less than a microsecond. Extending MD simulations to experimentally relevant time scales represents an ongoing challenge in computational physics, especially when the underlying dynamics is dominated by thermally activated, infrequent events. Significant progress towards extending the time scales of MD simulations has been made using novel acceleration techniques [1]–[3].

The *parallel-replica* method [2] uses multiple processors for statistically independent simulations of a system starting from the same initial state. A transition to a new state occurring on any one of the processors causes all simulations to be reset to this new state. With the appropriate accounting algorithm, the accumulated time over all processors may be used for an estimate of the transition time, and in this way, the accessible simulation time is boosted by the number of available processors.

The *temperature accelerated dynamics* (TAD) method [3] extends the simulation time by running the MD simulation at high temperature to accelerate the thermally induced transitions. These high-temperature transition times are then extrapolated to equivalent low temperature transition times using certain assumptions based on harmonic transition state theory. With the appropriate accounting algorithm, simulation time is effectively extended.

An underlying assumption of these acceleration techniques is transition state theory, where the dynamics are viewed as proceeding through an infrequent thermal activation from one potential energy basin to another. A requirement in the implementation of these and other acceleration techniques is the ability to determine what state a system is in and to detect with precision when a transition has occurred.

These tasks can be difficult in even simple systems. The thermal noise, which must be present to induce a transition, makes the configuration of the system at any time step unrecognizable as a well-defined state or structure. A common method to resolve this problem is to periodically stop the MD simulation and relax the system. These periodic stop-and-quench operations can be costly, resulting in a trade-off: the less frequent the stop-and-quench operations, the less precision one has in identifying transition times. For optimum efficiency with these methods, one needs to impose an assumption about likely transition times. This stop-and-quench approach will be compromised by systems with multiple reaction mechanisms and substantially different transition times, as well as rapid, dynamically correlated events.

2 REAL-TIME MULTIRESOLUTION ANALYSIS

We propose to resolve the difficulties in identifying structures and in determining the state of the system, as well as detecting transition events, using the technique of real-time multiresolution analysis (RTMRA). A wavelet-based detection scheme is constructed to detect meta-stability and transitions in the atomic coordinates

of the system. Formally, the time series corresponding to each atomic coordinate, x(t), is expanded upon a wavelet basis,

$$x(t) = \sum_n \bar{x}_n \phi_n(t) + \sum_m \sum_n \tilde{x}_n^m \psi_n^m(t) \qquad (1)$$

where $\phi_n(t) = \phi(t-n)$ and $\psi_n^m(t) = 2^{m/2}\psi(2^m t - n)$ are the scaling functions and wavelets, respectively, from which the wavelet basis is constructed. Here, $\phi(t)$ and $\psi(t)$ are, respectively, the mother scaling function and wavelet used to generate a particular wavelet basis set. The wavelet basis constructed by Haar [4] is used in this work for its simplicity and strict relationship to local averages. Simply described, a scheme is constructed to examine the magnitude of wavelet expansion coefficients for indications of temporal features associated with local stability and transition.

The dilational symmetry of the basis set allows the detection of these temporal features to be performed over multiple time scales. No *a priori* assumption are required regarding the characteristic time scales involved in the overall dynamics.

In theory, if the magnitude of all wavelet expansion coefficients are found to be below a predetermined threshold, it may be concluded that (1) over the corresponding time interval, the system is meta-stable and stochastically sampling a well-defined potential energy basin, and (2) the average configuration of the system over this time interval will closely approximate the local minimum energy configuration that would be found through relaxation. Conversely, if the magnitude of any wavelet expansion coefficient is found to be above a certain predetermined threshold, it may be concluded that the system is in transition over this time interval, moving from one potential energy basin to another.

In Theory, the use of wavelet analysis to identify temporal features in molecular dynamics simulations is straightforward. However, to be efficient, great care must be exercised in the implementation.

Figure 1 shows schematically the integration of these RTMRA techniques with an existing MD code. The original MD code is essentially retrofit with these techniques by replacing the previous "write" statements, recording the atomic positions, with calls to the time-domain RTMRA algorithm. A critical component in this algorithm is the real-time wavelet transform (RTWT) used to incrementally generate the wavelet expansion coefficients. This RTWT is more efficient than the more conventional fast wavelet transform and is capable of treating streaming data with minimal overhead. The time-domain RTMRA manages the flow of wavelet expansion coefficients to the detection algorithms. This task is complicated by the varying rates at which the hierarchy of wavelet expansion coefficients are generated.

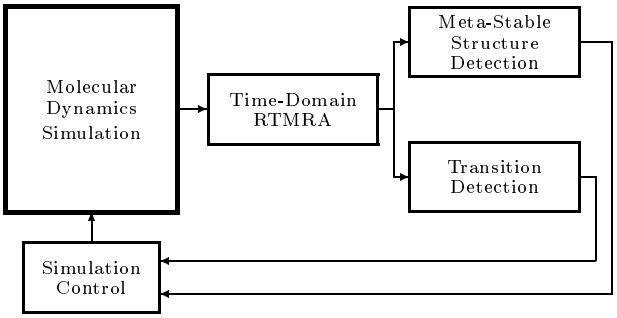

Figure 1: Integration of RTMRA techniques with an existing MD code for run-time temporal feature detection

REFERENCES

[1] A. F. Voter, Phys. Rev. Lett. **78**, 3908, 1997.
[2] A. F. Voter, Phys. Rev. B **57**, 13985, 1998.
[3] M. R. Sørensen and A. F. Voter, J. of Chem. Phys. **112**, 9599, 2000.
[4] A. Haar, Math. Ann. **69**, 331, 1910.

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