Molecular dynamics algorithm for multiple time scales: Systems with disparate masses

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A frequently encountered problem in molecular dynamics is how to treat the long times that are required to simulate condensed systems consisting of mixtures of light and heavy particles. Standard methods require the choice of time step sufficiently small to guarantee stable solution for the low mass component with the consequence that these simulations require a very large number of central processing unit cycles to treat the relaxation of the heavier component. In this note, we present a new method that allows one to use a time step appropriate for the heavy particles. This method uses a similar idea to numerical analytical propagator algorithm, an algorithm we invented to treat high frequency oscillators interacting with low frequency baths and is based on a choice of a reference system for the light particle motions. The method is applied to the case of a liquid containing 864 Lennard-Jones spheres, 824 of these particles having a mass, $M = 100$ and 40 spheres picked at random have a mass $m = 1$. It is shown that molecular dynamics using the new algorithm runs seven to ten times faster than standard methods and this approach as well as suitable generalizations should be very useful for future simulations of quantum and classical condensed matter systems.

I. INTRODUCTION

Consider a system consisting of a mixture of light (mass $= m$) and heavy spheres (mass $= M$). In such systems, there is a disparity in the molecular dynamic time scales. If one wishes to simulate such systems using the standard integrators of molecular dynamics, then the maximum time step that can be used to integrate the equations of motion must be chosen to insure accurate integration of the low mass component with the consequence that a very small time step is needed. When a large disparity in time scales exists, a very large number of central processing unit (CPU) cycles will be required to allow the slow degrees of freedom to fluctuate enough to obtain converged time averages for the whole system.

In this paper, a method for accelerating the simulation of such systems is presented. This method, called RESPA (reference system propogator algorithm), is a variant of the numerical analytical propagator algorithm (NAPA), algorithm that we invented for treating the problem of high frequency oscillators coupled to low frequency oscillators.[1] The RESPA method is based on numerical solutions of the reference system equations. The gist of the method is to define a dynamical reference system for the fast motions and to derive equations of motion for the deviation $\delta(t)$ of the fast coordinates from the reference system coordinates. These deviations are coupled to the equations of motion of the slow coordinates. The fast dynamical system is integrated for n small time steps $n\delta t$ holding the slow coordinates fixed. The time dependence of the reference system is then fed into the coupled equations for $\delta(t)$ and the slow coordinates and the resulting equations are integrated for one large time step

$\Delta t = n\delta t$. The initial conditions for each large time step are then chosen so that this deviation $\delta(t)$ is zero with the consequence that the deviation is always kept small. The only approximation in this algorithm springs from the numerical integrator used to integrate the equations of motion of the reference system and the coupled equations. Otherwise, the method is self-correcting and exact. For simplicity, the reference system is taken to be the Hamiltonian of the original system with the slow coordinates held fixed at their values at the beginning of the time step.

Teleman and Jönsson[2] have proposed a multiple time-step (TJMTS) method in which the forces are separated into slow and fast components. This separation yields a set of coupled equations of motion for the slow and fast degrees of freedom. TJMTS uses a small step δt to advance the fast degrees of freedom n steps holding the slow variables fixed. The slow degrees of freedom are then updated using a time step $\Delta t = n\delta t$. This method does not correct for the errors incurred in the approximate factorization of the equations of motion, a fact which shows up in poor energy conservation. This is well illustrated when we compare the results of RESPA, TJMTS, and velocity Verlet[3] (using a small time step). Swindoll and Haile[4] have proposed a more accurate multiple time-step method than Teleman and Jönsson, but their method requires high-order spatial derivatives of the potential and is therefore more computationally intensive than RESPA.

For simplicity, we apply this new method to the simulation of a mixture of Lennard-Jones spheres consisting of 824 heavy spheres of mass $M = 100$ and 40 light spheres of mass $m = 1$. For Lennard-Jones (LJ) spheres, the two time scales are $\Delta t_f = \sqrt{m\sigma_i^2/\epsilon_i}$ and $\Delta t_s = \sqrt{M\sigma_i^2/\epsilon_i}$, where σ_i and ϵ_i are the Lennard-Jones parameters for component i. For il-

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Molecular Dynamics Algorithm For Multiple Time Scales

TD Snyder

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