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 propogator 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 propogator 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 $\Delta t_f = \sqrt{m\sigma_1^2/\epsilon_1}$ and $\Delta t_s = \sqrt{M\sigma_2^2/\epsilon_2}$, where σ_i and ϵ_i are the Lennard-Jones parameters for component i. For il-

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Multiple Time Scales Jeremiah U. Brackbill,Bruce I. Cohen,2014-05-10 Multiple Time Scales presents various numerical methods for solving multiple time scale problems The selection first elaborates on considerations on solving problems with multiple scales problems with different time scales and nonlinear normal mode initialization of numerical weather prediction models Discussions focus on analysis of observations nonlinear analysis systems of ordinary differential equations and numerical methods for problems with multiple scales The text then examines the diffusion synthetic acceleration of transport iterations with application to a radiation hydrodynamics problem and implicit methods in combustion and chemical kinetics modeling The publication ponders on molecular dynamics and Monte Carlo simulations of rare events direct implicit plasma simulation orbit averaging and subcycling in particle simulation of plasmas and hybrid and collisional implicit plasma simulation models Topics include basic moment method electron subcycling gyroaveraged particle simulation and the electromagnetic direct implicit method The selection is a valuable reference for researchers interested in pursuing further research on the use of numerical methods in solving multiple time scale problems **Classical And Quantum Dynamics In Condensed Phase Simulations: Proceedings Of The International School Of Physics** Bruce J Berne,Giovanni Ciccotti,David F Coker,1998-06-17 The school held at Villa Marigola Lerici Italy in July 1997 was very much an educational experiment aimed not just at teaching a new generation of students the latest developments in computer simulation methods and theory but also at bringing together researchers from the condensed matter computer simulation community the biophysical chemistry community and the quantum dynamics community to confront the shared problem the development of methods to treat the dynamics of quantum condensed phase systems This volume collects the lectures delivered there Due to the focus of the school the contributions divide along natural lines into two broad groups 1 the most sophisticated forms of the art of computer simulation including biased phase space sampling schemes methods which address the multiplicity of time scales in condensed phase problems and static equilibrium methods for treating quantum systems 2 the contributions on quantum dynamics including methods for mixing quantum and classical dynamics in condensed phase simulations and methods capable of treating all degrees of freedom quantum mechanically

Computational Molecular Dynamics: Challenges, Methods, Ideas Peter Deuflhard,Jan Hermans,Benedict Leimkuhler,Alan E. Mark,Sebastian Reich,Robert D. Skeel,2012-12-06 On May 21 24 1997 the Second International Symposium on Algorithms for Macromolecular Modelling was held at the Konrad Zuse Zentrum in Berlin The event brought together computational scientists in fields like biochemistry biophysics physical chemistry or statistical physics and numerical analysts as well as computer scientists working on the advancement of algorithms for a total of over 120 participants from 19 countries In the course of the symposium the speakers agreed to produce a representative volume that combines survey articles and original papers all refereed to give an impression of the present state of the art of Molecular Dynamics The 29 articles of the book

reflect the main topics of the Berlin meeting which were i Conformational Dynamics ii Thermodynamic Modelling iii Advanced Time Stepping Algorithms iv Quantum Classical Simulations and Fast Force Field and v Fast Force Field Evaluation Computer Simulations in Condensed Matter: From Materials to Chemical Biology. Volume 1 Mauro Ferrario,Giovanni Ciccotti,Kurt Binder,2007-03-09 This comprehensive collection of lectures by leading experts in the field introduces and reviews all relevant computer simulation methods and their applications in condensed matter systems Volume 1 is an in depth introduction to a vast spectrum of computational techniques for statistical mechanical systems of condensed matter Volume 2 is a collection of state of the art surveys on numerical experiments carried out for a great number of systems **Reaction Dynamics in Clusters and Condensed Phases** Joshua Jortner,R.D. Levine,A. Pullman,2012-12-06 The Twenty Sixth Jerusalem Symposium reflected the high standards of these distinguished scientific meetings which convene once a year at the Israel Academy of Sciences and Humanities in Jerusalem to discuss a specific topic in the broad area of quantum chemistry and biochemistry The topic at this year s Jerusalem Symposium was reaction dynamics in clusters and condensed phases which constitutes a truly interdisciplinary subject of central interest in the areas of chemical dynamics kinetics photochemistry and condensed matter chemical physics The main theme of the Symposium was built around the exploration of the interrelationship between the dynamics in large finite clusters and in infinite bulk systems The main issues addressed microscopic and macroscopic sol vation phenomena cluster and bulk spectroscopy photodissociation and vibrational predissociation cage effects interphase dynamics reaction dynamics and energy transfer in clusters dense fluids liquids solids and biophysical systems The interdisciplinary nature of this research area was deliberated by intensive and extensive interactions between modern theory and advanced experimental methods This volume provides a record of the invited lectures at the Symposium **Liquid Crystals and their Computer Simulations** Claudio Zannoni,2022-07-28 A comprehensive introduction to liquid crystals and their computer simulations suitable for students researchers and industrial scientists Wspc Reference On Organic Electronics, The: Organic Semiconductors (In 2 Volumes) Seth R Marder,Jean-luc Bredas,2016-06-24 This 2 volume set provides the reader with a basic understanding of the foundational concepts pertaining to the design synthesis and applications of conjugated organic materials used as organic semiconductors in areas including organic photovoltaic devices light emitting diodes field effect transistors spintronics actuation bioelectronics thermoelectrics and nonlinear optics While there are many monographs in these various areas the emphasis here is both on the fundamental chemistry and physics concepts underlying the field of organic semiconductors and on how these concepts drive a broad range of applications This makes the volumes ideal introductory textbooks in the subject They will thus offer great value to both junior and senior scientists working in areas ranging from organic chemistry to condensed matter physics and materials science and engineering Number of Illustrations and Tables 168 b w illus 242 colour illus 13 tables **Multiscale Computational Methods in Chemistry and Physics** Achi Brandt,Jerzy Bernholc,Kurt Binder,2001 This book brings

together interdisciplinary contributions ranging from applied mathematics theoretical physics quantum chemistry and molecular biology all addressing various facets of the problem to connect the many different scales that one has to deal with in the computer simulation of many systems of interest in chemistry e g polymeric materials biological molecules clusters surface and interface structure Particular emphasis is on the multigrid technique and its applications ranging from electronic structure calculations to the statistical mechanics of polymers **Long Time Scale Computer Simulations of Proteins** Peter Eastman,2000 **Theoretical Biophysics Technical Report** ,1991 *Multiple-time-scale Order Reduction for Stochastic Kinetics and Molecular Simluation of Crystallization* Ethan Allen Sturman Mastny,2007 1953, □□□□□ □□□□ Dissertation Abstracts International ,2008 Bridging the Time Scales Peter. Nielaba,Michel Mareschal,Giovanni Ciccotti,2014-01-15 **Mathematical Reviews** ,2008 Proceedings of the ... ASME Design Engineering Technical Conferences ,2005 Monte Carlo and Molecular Dynamics of Condensed Matter Systems Kurt Binder,Giovanni Ciccotti,1996 **Bridging the Time Scales** Peter Nielaba,Michel Mareschal,Giovanni Ciccotti,2002-12-19 The behaviour of many complex materials extends over time and lengthscales well beyond those that can normally be described using standard molecular dynamics or Monte Carlo simulation techniques As progress is coming more through refined simulation methods than from increased computer power this volume is intended as both an introduction and a review of all relevant modern methods that will shape molecular simulation in the forthcoming decade Written as a set of tutorial reviews the book will be of use to specialists and nonspecialists alike *Physics Briefs* ,1993 **Pacific Symposium on Biocomputing** ,1996

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