



Algorithms of GPU-enabled reactive force field (ReaxFF) molecular dynamics

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ABSTRACT

Reactive force field (ReaxFF), a recent and novel bond order potential, allows for reactive molecular dynamics (ReaxFF MD) simulations for modeling larger and more complex molecular systems involving chemical reactions when compared with computation intensive quantum mechanical methods. However, ReaxFF MD can be approximately 10–50 times slower than classical MD due to its explicit modeling of bond forming and breaking, the dynamic charge equilibration at each time-step, and its one order smaller time-step than the classical MD, all of which pose significant computational challenges in simulation capability to reach spatio-temporal scales of nanometers and nanoseconds. The very recent advances of graphics processing unit (GPU) provide not only highly favorable performance for GPU enabled MD programs compared with CPU implementations but also an opportunity to manage with the computing power and memory demanding nature imposed on computer hardware by ReaxFF MD. In this paper, we present the algorithms of GMD-Reax, the first GPU enabled ReaxFF MD program with significantly improved performance surpassing CPU implementations on desktop workstations. The performance of GMD-Reax has been benchmarked on a PC equipped with a NVIDIA C2050 GPU for coal pyrolysis simulation systems with atoms ranging from 1378 to 27,283. GMD-Reax achieved speedups as high as 12 times faster than Duin et al.'s FORTRAN codes in Lammps on 8 CPU cores and 6 times faster than the Lammps' C codes based on PuReMD in terms of the simulation time per time-step averaged over 100 steps. GMD-Reax could be used as a new and efficient computational tool for exploiting very complex molecular reactions via ReaxFF MD simulation on desktop workstations.

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1. Introduction

Molecular dynamics based on classical mechanics (classical MD or MD) is a basic and popular molecular modeling method that is widely applied in biochemistry, biophysics and material science. But classical MD only describes physical elastic collision between atoms with static bonds and fixed partial charges, which are not feasible for chemical reactive systems. Density functional theory (DFT) is a quantum mechanical (QM) modeling method used in physics and chemistry to investigate the electronic structure of many body systems with high accuracy when involved in chemical bonding [1]. But DFT is extremely computationally intensive and is

usually applied to systems containing tens or hundreds of atoms and picoseconds simulation timeframes [2].

Motivated by restrictions of the two approaches mentioned above for simulation of molecular systems involving chemical bonding, a number of recent efforts have been in progress for bridging the gap between quantum chemistry and classical molecular dynamics. Among these efforts, reactive force field (ReaxFF) developed by van Duin et al. [3] opened up a new pathway for studying molecular systems with chemical reactions. ReaxFF molecular dynamics (ReaxFF MD) presents the movement rule of chemical systems based on the bond order (BO) concept [4,5], which has been proven to be a smooth transition from non-bonded to single-, double- and triple-bonded system with all connectivity-dependent interactions to be the bond order dependent to ensure that the energy contribution disappears upon bond dissociation [6]. Since the bond orders in ReaxFF are updated in each time-step iteration, the connectivity of the system can continuously change with time [7]. ReaxFF MD allows the simulation of larger molecular reactive systems with acceptable accuracy when validated with DFT [3,6] and demonstrates its potential in simulating very complex systems on large scale supercomputers for up to millions of atoms with nanosecond time scale to predict reasonable reaction mechanism [5].

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Supercomputers are often beyond the reach of research groups and high performance computational tools on a single desktop workstation would be more useful for individual researcher in exploiting simulation applications. However, according to the benchmark of Lammmps [7] from Sandia National Laboratory, ReaxFF is the most CPU time and memory demanding potential among the 20 force fields when compared with LJ liquid potential. ReaxFF could take about 8.9 s for a single time-step (0.1 fs) for a system of PETN ([3-nitrooxy-2,2-bis(nitrooxymethyl)propyl] nitrate) crystal with 32,840 atoms in a simulation on a desktop computer using ReaxFF C code in Lammmps, i.e., such a calculation for 1 ns would cost more than 1030 days, that is hardly acceptable for simulation practitioners. The hardware limitation could be still a barrier for applying ReaxFF MD simulation on desktop computers, particularly for simulation of large molecular systems.

With the computing peak performance and memory bandwidth far exceeding CPU, the graphics processing unit (GPU) has shown its great potential more recently in accelerating MD simulation by bringing teraflops computational capability to desktop workstations, which may bring transformation of many computations from clusters to desktops [8,9]. Although there may be feature limitations, the benchmarks using state-of-the-art GPU enabled MD programs such as Amber [10], NAMD [11], Gromacs [12] have shown speedups of several folds, even one to two orders of magnitude, over the well optimized CPU implementations on a single PC. The continuing advances of GPU could provide not only an alternative for parallel but also a new opportunity to manage the computing power and memory demanding nature imposed on computer hardware by ReaxFF, especially for effective simulation on desktop workstations.

In this paper, we present the novel design and algorithms of our GMD-Reax, the first GPU enabled ReaxFF MD implementation to our best knowledge, both for increasing simulation system size and acceleration of ReaxFF, aiming at a computational tool on desktop workstations. It works on a PC equipped with a single C2050 GPU from NVIDIA with Fermi architecture [13]. GMD-Reax has been tuned to take advantage of GPU as much as possible. In addition, every effort was made to develop a general architecture in the code so that it can be applicable with the upgrade of GPU.

In this paper, the ReaxFF force field and its paralleled algorithm implementation on CPU clusters are overviewed in Section 2, which are closely related in dealing with the computing challenge of ReaxFF by paralleled strategy. Recent developments in GPU technology and Compute Unified Device Architecture (CUDA) used for GPU programming will be briefly reviewed in Section 3. The original algorithms of ReaxFF and its GPU based algorithms in our GMD-Reax are outlined in Section 4, and then performance benchmarks of GMD-Reax are analyzed accordingly in Section 5. We conclude the paper with brief discussion to further research topics in last section.

2. ReaxFF MD overview and related work

ReaxFF is an empirical reactive force field that was first reported for hydrocarbons [3] and then expanded in recent years by Duin and co-workers, for simulation of reactive molecular systems involving a range of elements such as carbon, hydrogen, oxygen, nitrogen and so on. ReaxFF MD has been successfully applied to a number of various complicated system simulations with reactions for the process of combustion and catalysis. ReaxFF has been employed in the study of several oxides such as Si/SiO₂ [14], V_xO_y [15], ZnO [16] and Al/Al₂O₃ [17] oxide interfaces for investigating catalytic mechanism or initial reaction path. ReaxFF has also been utilized to explore properties of new materials, namely single-walled carbon nanotubes (SWNT) [18] and Lithium Battery Electrolytes [19].

In addition, Mayernick [20] combined ReaxFF and Monte Carlo (MC) to perform simulated annealing to sample structural configurations of flat yttria-stabilized zirconia (YSZ) and obtained considerable results. More recently, ReaxFF was used in the combustion simulation of Illinois No. 6 coal char for a system containing over 30,000 atoms to explore the structural evolution of char structural and chemical mechanics by Castro-Marciano et al. [21] that demonstrated the ability of ReaxFF in dealing with large reactive molecular systems. These applications and the fact that the ReaxFF program has been distributed to over 150 research groups worldwide [22] as well have shown the capability and great potential of ReaxFF to handle complex chemistry and chemical diversity of larger molecular systems with chemical reaction to a wide range of materials and processes.

The ability of ReaxFF to describe bond formation and charge transfer for molecular systems in condensed phases comes from its special interatomic potential, in which the bonded interactions are based on distance dependent bond order functions that are adjusted to compensate for atomic over/under-coordination, and the atom charges are computed using electronegativity equalization. As stated in Eq. (1), the total potential energy in ReaxFF is the sum of the following energy terms [8]:

$$E_{\text{ReaxFF}}(r_{ij}, r_{ijk}, r_{ijkl}, q_i, \text{BO}_{ij}) = E_{\text{bond}} + E_{\text{lp}} + E_{\text{over}} + E_{\text{under}} + E_{\text{val}} \\ + E_{\text{pen}} + E_{\text{coa}} + E_{\text{tors}} + E_{\text{conj}} + E_{\text{Hbond}} \\ + E_{\text{vdWaaals}} + E_{\text{Coulomb}} \quad (1)$$

where E_{vdWaaals} and E_{Coulomb} are the energy of non-bonded interactions, namely the van der Waals and Coulomb interactions. In addition to E_{Hbond} , the energy of hydrogen bond, the rest of the energy terms are the bond order dependent and account for the bonded interactions that consists of E_{bond} , the energy of bond, E_{lp} for energy of lone-pairs, E_{over} of over-coordination, E_{under} of under-coordination, E_{val} of valence angle, E_{pen} of energy penalty for handling atoms with two double bonds, E_{coa} of coalition (three-body conjugation), E_{conj} of conjugated bonds (four-body conjugation), E_{tors} of torsion angles.

To avoid discontinuities on the potential energy surface, much more complex mathematical formulations for energy terms in Eq. (1) other than that in most of the classical MD methods are being used in ReaxFF [16] which significantly increase the computational complexity. Moreover, the time-step magnitude of ReaxFF MD is one order smaller than classical molecular dynamics (0.1 fs vs. 1 fs), resulting in its one order longer in computation time than that for classical MD. Unlike the fixed partial charges on atoms in classical MD, atomic partial charges in ReaxFF MD are dynamically approximated in a charge equilibration procedure by minimizing the Coulomb energy using Electron Equilibration Method (EEM) when the atomic coordinates are updated at each time-step [23]. Charge equilibration is quite computational expensive, although the procedure can be transformed into solving large sparse equations and the rank of the matrix is the total number of atoms. Therefore, ReaxFF MD can be 10–50 times slower than classical MD [22] and it becomes critical to improve its computing performance, especially when ReaxFF MD is used in simulation of complex chemical reactive systems.

As shown in Fig. 1, ReaxFF molecular dynamics treats atom interactions and their evolving with time in the same way as in classical MD. There are few dependencies and coupling between interacting atoms. The loop over atoms is basically data parallel that fits well with the nature of GPU and could take advantage of the great computing power of it. Meanwhile the memory demanding introduced by the bond order dependency and other complex calculations in ReaxFF can also be tackled with GPU's high memory bandwidth.

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