Center for Nanophase Materials Sciences

Next Generation Force Fields for Nanoscience

September 15-16, 2010

The goal of this workshop is to gather a number of the experts and participants in the field of materials force field development to discuss current approaches and to design a road map for improvement and implementation for nanoscience. Traditional empirical-based force fields have been very valuable tools for developing a structural understanding of a variety of systems, ranging from metals to oxides to biomolecules and other soft materials. Their added scientific value extends across many different fields. These force fields have generally been based on numerous approximations and derived from different types of experimental data, making them very suitable for that particular domain. But nanoscience has pushed the limits of the type of problems we can continue to address with the current state-of-the-art force fields, in particular due to the preponderance of interfacial interactions where electronic polarizability becomes especially important. Additionally, simulations of catalytic processes and other types of chemical reactions must take charge transfer into account. Even forces such as weak dispersion cannot be fully accounted for without having some form of polarizablility included.

It is clear that a new generation of force fields is necessary that will be able to keep up with the pace of problems that nanoscience and nanotechnology presents us. These force-fields will have to describe charge transfer in a robust yet efficient way, that will allow us to describe the dynamics and structure of a large variety of nanoscale systems and processes.

Many approaches exist to account for bond-breaking and bond-making events and for including polarizability (ReaxFF, OMOEBA, SIBFA, PFF, GEM, DRF90, AIREBO). The overall objective of this workshop is to work with experts in the field to discuss those approaches and to design a road map for improvement and for efficient implementation. A workshop report will be developed to solidify the discussions and to serve as a template for a journal review article on the topic.

Speakers:

Adri van Duin, Pennsylvania State University
David Sherrill, Georgia Tech
Donald Brenner, North Carolina State University
Susan Sinnott, University of Florida
Ilja Siepmann, University of Minnesota
Aidan Thompson, Sandia National Laboratory
Steven J. Stuart, Clemson University
Pratul Agarwal, Oak Ridge National Laboratory

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Title, Presenter, and Abstract in Order of Appearance

"Linking Protein Flexibility to Enzyme Catalysis"
Pratul Agarwal, Oak Ridge National Laboratory

Enzymes are phenomenal catalysts, as they can provide rate-enhancement by as much as 17 orders of magnitude. The role of enzyme structure in mechanism has been understood for some time; recently protein flexibility (internal protein motions) have also been linked to biophysical mechanism of catalysis. In this talk, evidence from a series of computational studies for the role of protein structure and dynamics in the enzyme mechanism will be discussed. Particular emphasis will be on identifying and characterizing the slow conformational fluctuations (microsecond-millisecond time-scales). The need for better methods, longer time-scales and emerging computer hardware (such as graphical processing units, GPUs) will be discussed.

"Fixing Inter-Atomic Potentials with Multiscale Modeling: Some Results and Speculation"
Don Brenner, North Carolina State University

Many-body interatomic potentials such as those derived within the EAM, (AI)REBO, ReaxFF, and COMB formalisms, together with the various flavors of charge equilibration schemes, have evolved to the point that a relatively wide range of materials and processes can be modeled with predictable accuracy. However, there remain properties such as the room temperature thermal conductivity of metals that rely on electronic degrees of freedom that in general cannot be appropriately captured with inter-potentials that neglect explicit electrons in exchange for computational efficiency. Presented in this talk will be some results and speculation as to how multi-scale modeling can "fix" inter-atomic potentials. In particular, our scheme using ad hoc feedback between atomic simulations and numerical solutions to thermal and electronic transport equations will be discussed, as will some thoughts on other types of atomic-continuum coupling that can enhance inter-atomic potentials.

"The ReaxFF Reactive Force Field Method: Concepts, Development, and Applications"
Adri van Duin, Pennsylvania State University

The ReaxFF method provides a highly transferable simulation method for atomistic scale simulations on chemical reactions at the nanosecond and nanometer scale. It combines concepts of bond-order based potentials with a polarizable charge distribution. Since its initial development for hydrocarbons in 2001, we have found that this concept is highly transferable, leading to applications to elements all across the periodic table, including all first row elements, metals, ceramics and ionic materials. For all these elements and associated materials we have demonstrated that ReaxFF can accurately reproduce quantum mechanics-based structures, reaction energies and reaction barriers, enabling the method to predict reaction kinetics in complicated, multi-material environments at a relatively modest computational expense.

In this presentation I will describe the current concepts of the ReaxFF method, the current status of the various ReaxFF codes, including parallel implementations. I will also

discuss current limitations and plans to resolve these.

Finally, I will present and overview of recent applications to a wide range of different materials, including combustion, catalysis, aqueous phase chemistry and material failure.

"Classical Molecular Dynamics Simulations of Hetergenous Interfacial Systems with COMB Potentials"
Susan Sinnott, University of Florida

Many high-performance devices consist of multilayers of materials that form heterogeneous interfaces with significant changes in bonding as one crosses from one side of the interface to the other. Traditionally, computational studies of these interfacial systems have relied on first-principles electronic structure approaches because of the difficulty in describing the changes in bonding environment with empirical approaches. While effective, such methods are limited to structures that are too small to model many aspects of the interface. Here, reactive, empirical, charge optimized many-body (COMB) potentials for use in classical molecular dynamics simulations of heterogeneous systems are reviewed. The COMB potentials have been parameterized for several systems, including Si, SiO_2, Hf, HfO_2, Ti, TiO_2, Cu, Cu_2O, Al, and Al_2O_3, and efforts are currently underway to add to this list. Several examples of the application of COMB potentials to physical problems will be discussed, and its predictions compared to experimental data and the results of first-principles methods. This work is supported by the National Science Foundation (DMR-0426870) and by the Department of Energy (DE-FG02-07ER46446).

"Simulating Micro-heterogenous Fluids and Soft Nanosystems: Needs for Empirical Force Fields and Fast Electronic Structure Methods"
Ilja Siepmann, University of Minnesota

The accuracy of particle-based simulations depends solely on the quality of the force field used to describe the inter- and intramolecular interactions for the chemical system of interest. This talk will highlight some of the successes and failures of utilizing empirical force fields or fast electronic structure methods for the simulation of micro-heterogeneous fluids and of nano-droplets. In addition, the needs for reliable experimental data on nano systems will be discussed.

"Quantum Benchmarks for the Assessment of Force Fields for Noncovalent Interactions, and Improved Polarizable Force Fields"
David Sherrill, Georgia Tech

Quantum chemical methods have recently made it possible to obtain highly accurate (within ~1 kJ/mol) benchmark interaction energies for van der Waals dimers with up to about 30 atoms. By using density fitting and Cholesky decomposition techniques, we have also implemented an extremely efficient method for decomposing interaction energies into their electrostatic, dispersion, induction, and exchange-repulsion components which is nearly as accurate as the most accurate benchmark methods for interaction energies and which scales to well over 100 atoms. Using such benchmark results and decomposition analysis, we will report results of recent collaborative efforts with ORNL and with the University of Florida to assess the ability of standard force fields to model pi-stacking and other

noncovalent interactions. We will also use these benchmark results to parameterize and test extensions to polarizable force fields which use a more general expression for the short-range repulsion term.

"Large Scale Molecular Dynamics Simulation Using Reactive Interatomic Potentials in LAMMPS"
Aidan Thompson, Sandia National Laboratories

This talk will be an overview of recent developments involving the implementation and use of reactive interatomic potentials in the LAMMPS parallel molecular dynamics package, with particular attention to the ReaxFF potential. ReaxFF was recently used to perform molecular dynamics simulations of PETN on systems of up to 500,000 atoms for about 100 picoseconds. Shock-loading was achieved by colliding a relaxed crystal against a reflective wall at two different velocities: 3 km/s and 4 km/s.

Particle velocity, density, stress, temperature and chemical composition are extracted as a function of time and position. The higher impact velocity results in a much narrower initial reaction zone and a faster transition to a steady shockwave propagation velocity. The weaker impact appears to approach the same the final propagation velocity, but on timescales that are an order-of-magnitude longer.

Start	End	Time	Topic	Speaker
Next Generation Force Fields for Nanoscience			**DAY ONE**	
Wednesday, Sept. 15				
9:00 a.m.	9:15 a.m.	15 minutes	Welcome/Overview	Bobby Sumpter, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory
9:15 a.m.	10:00 a.m.	45 minutes	TBD	Pratul Agarwal, Computer Science Research Group, Oak Ridge National Laboratory
10:00 a.m.	10:45 a.m.	45 minutes	Fixing Inter-Atomic Potentials with Multiscale Modeling: Some Results and Speculation	Don Brenner, Materials Sciene and Engineering, North Carolina State University
10:45 a.m.	11:00 a.m.	15 minutes	Break	
11:00 a.m.	11:45 a.m.	45 minutes	The ReaxFF Reactive Force Field Method: Concepts, Development, and Applications	Adri van Duin, Mechanical and Nuclear Engineering, Pennsylvania State University
11:45 a.m.	1:00 p.m.	1 hour, 15 minutes	Working Lunch, discussions/summary on morning topics	
1:00 p.m.	1:45 p.m.	45 minutes	Classical Molecular Dynamics Simulations of Hetergenous Interfacial Systems with COMB Potentials	Susan Sinnot, Materials Science and Engineering, University of Florida
1:45 p.m.	2:30 p.m.	45 minutes	Simulating Micro-heterogenous Fluids and Soft Nanosystems: Needs for Empirical Force Fields and Fast Electronic Structure Methods	Ilja Siepmann, Chemistry, University of Minnesota
2:30 p.m.	2:45 p.m	15 minutes	Afternoon Break	
2:45 p.m.	3:30 p.m.	45 minutes	Quantum Benchmarks for the Assessment of Force Fields for Noncovalent Interactions, and Improved Polarizable Force Fields	David Sherrill, Chemistry and Biochemistry, Georgia Tech
3:30 p.m.	4:15 p.m.	45 minutes	Recent Developments in Bond-Order Potentials	Steve Stuart, Physical Chemistry, Clemson University
4:15 p.m.	5:00 p.m.	45 minutes	Large Scale Molecular Dynamics Simulation Using Reactive Interatomic Potentials in LAMMPS	Aidan Thompson, Multiscale Dynamic Materials Modeling, Sandia National Laboraties
5:00 p.m.	5:45 p.m.		Break/Discussion, End Day	

Start	End	Time	
Next Generation Force Fields for Nanoscience			**DAY TWO**
Thursday, Sept. 16			
8:30 a.m.	8:35 a.m.	5 minutes	Welcome, Day Two
8:35 a.m.	9:15 a.m.	40 minutes	Summary/Discussion of Day One
9:15 a.m.	10:45 a.m.	1 hour, 30 minutes	Panel: Road Map Development Plans and Article Preparation
10:45 a.m.	11:00 a.m.	15 minutes	Break
11:00 a.m.	12:00 p.m.	1 hour	Continuation of Road Map
12:00 p.m.	12:45 p.m.	45 minutes	Working Lunch; round table discussion on collaborative opportunities
12:45 p.m.	1:45 p.m.	1 hour	Final Discussion of Roadmap and Summary Article Preparation
1:30 p.m.			End; Guests Depart for Airport