Brainstorm meeting on the development of next generation force fields

12-14 September 2011

Organisers

- Jean-Philippe PIQUEMAL (UPMC-University Paris 6, France)
- Kenneth D. JORDAN (University of Pittsburgh, USA)

Introduction and motivation

Simulations of complex materials and chemical and biological systems require the use of model potentials (force fields). As a consequence, the results of the simulations are "only as good as the force field employed." In principle, high-level electronic structure calculations on appropriate model systems can provide the information needed for generating accurate force fields. As long time scale first principle simulations are still computationally intractable, new “polarizable” classical models grounded on quantum mechanics are needed.

State-of-the-art

Nowadays, modern molecular modelling techniques propose numerous potential applications, from materialsciences to protein structure prediction and drug design. Indeed, classical Molecular dynamics (MD) is now able to provide useful information to experimentalist as simulations are getting closer and closer to relevant biological timescales. Nevertheless, if MD is now able to produce microsecond trajectories, one should ask about the possible improvements of such simulations. At this point, two directions can be taken. The first consists in increasing the speed of MD softwares by coupling improved sampling methodology to massively parallel computers, having as goal to reach the second timescale. However, if this strategy will probably offer some interesting insights about physical or biophysical process, there is no doubt that the question of the accuracy of the used empirical energy functions, the socalled force fields, should be raised. Indeed, current simulations are mainly aimed to compute free energies and despite success, actual data are already sufficient to demonstrate that current molecular mechanics (MM) potentials have serious shortcomings. [1] This can be easily understood when considering that free energy required an accurate evaluation of both enthalpic and entropic contributions. If entropy can be recovered trough sampling efforts, enthalpy needs to be approximate from their quantum mechanical expression. In a way, classical MD can be simply seen as an approximate quantum Born-Oppenheimer MD approach treating the atomic nuclei as classical particles subject to interatomic forces. Presently, these latter remain obtained from empirical potentials far to reproduce first principles results. Therefore, MD should not be able to quantitatively describe vast numbers of systems dominated by difficult weak interactions such as H-bonds networks, metalloproteins and metal clusters, highly charges systems etc..., where Chemistry and electron correlation/relativity dominate. For these systems, the right tools are required. In this context, new methods such Anisotropic Polarizable Molecular Mechanics (APMM) procedures [2-9], the so-called polarizable force fields have been developed (see references [2] for a recent special issue published in J. Chem. Theory.

Comput.). These approaches share the common characteristic of including a more evolved representation of the electrostatic contribution to the interaction energy compared to the usual point charge approximation, allowing a closer reproduction of the anisotropic features of the ab initio Coulomb contribution. Meanwhile, new improved semi-empirical approaches [10-11] are emerging together with fast fragment based approximate quantum chemical techniques [12-14].

References

[1] T. Yoda, Y. Sugita, Y. Okamoto, J. Chem. Phys., 2004, 307, 269.
[2] W. L. Jorgensen, J. Chem. Theory Comput., 2007, 3, 1877
[3] A. J. Stone, Science 2008, 321, 787.
[4] J. W. Ponder, C. Wu, P. Ren, Vi. S. Pande, J. D. Chodera, M. J. Schnieders, I. Haque, D. L. Mobley, D. S. Lambrecht, R. A. DiStasio, M. Head-Gordon, G. N. I. Clark, M. E. Johnson, T. Head-Gordon, J. Phys. Chem. B, 2010, 114, 2549.
[5] N. Gresh, G. A. Cisneros, T. A. Darden, J-P. Piquemal, J. Chem. Theory. Comput, 2007, 3, 1960.
[6] P. Soderhjelm, A. Ohrn, U. Ryde, G. Karlstrom, J. Chem. Phys. 2008, 128, 014102
[7] G. Lamoureux, A. D. MacKerell, B. Roux, J Chem Phys 2003, 119, 5185.
[8] C. D. Wick, S. S. Xantheas, S. S. J. Phys. Chem. B, 2009, 113, 4146.
[9] R. Kumar, F. F. Wang, G. Jenness, K. D. Jordan, J. Chem. Phys., 2010, 132, 014309.
[10] T. J. Giese. D. M. York, J. Chem. Phys., 2007, 127, 194101.
[11] Y. Yang, H. Yu, D. M. York, Q. Cui, M. Elstner, J. Phys. Chem. A, 2007, 111, 10861
[12] C. Steinmann, D. G. Fedorov, J. H. Jensen, J. Phys. Chem. A., 2010, DOI: 10.1021/jp101498m
[13] T. Nagata, D. Fedorov, K. Kitaura, M.S. Gordon, J. Chem. Phys., 2009, 131, 024101.
[14] J. Gao, A. Cembran, Y. Mo, J. Chem. Theo. Comput., 2010, DOI: 10.1021/ct100292g

Objectives

The scope of this informal discussion that will bring brings together researchers with expertise in force field development and those with expertise in electronic structure method, is to allow discussion on the possible ways to improve the accuracy and efficiency of modern classical simulations of complex systems. Two main objectives:

- Identify the most crucial scientific issues one has to face in the research related to the "atomistic classical simulation problem" and in particular those that computational advanced polarisable approaches could help to solve, and
- Clarify which theoretical and computational methods one should develop and/or use to tackle specific problems, and which resources are needed to accomplish the tasks.

Therefore, the topic will focus on:

1) long time scale polarizable simulations of biomolecular systems, including those involving metals, and systems relevant for energy applications, including CO2 capture and sequestration.
2) High performance classical molecular dynamics: towards multi-millisecond polarizable simulations, parallel computers, GPUs and other dedicated processors.

We selected a small group of experts from academia, government and industry. The discussions throughout the meeting will facilitate new collaborations, leading to important advances.

Concerning the organization, the goal is to host around 15 invited talks of 40 minutes. There will also be about five short, ~15 minute contributed talks. This will leave ample time for discussions.

Another goal of this meeting is to encourage the participation of students and postdocs who will be able to present results in poster sessions.

Participant List

Jordan Kenneth D (jordan@pitt.edu)
University of Pittsburgh,USA

Plquemal Jean-Philip (jpp@lct.jussieu.fr)
UPMC - Université Paris 6, France

Borgis Daniel (daniel.borgis@ens.fr)
Ecole Normale Supérieur, Paris, France

Chipot Chris
Université de Nancy, France

Cisneros G. Andrés (andres@chem.wayne.edu)
Wayne State University, Detroit, USA

Clavaguera Carinne (carine.clavaguera@dcmr.polytechnique.fr)
Ecole Polytechnique, Palaiseau, France,

De Coursy Benoit
UMPC, France

Dehez François
Université de Nancy, France

Dognon jean-Pierre (jean-pierre.dognon@cea.fr)
CEA Saclay, France

Gresh Nohad (nohad.gresh@univ-paris5.fr)
Université Paris Descartes, France

Ingrosso Francesca
Université de Nancy, France

Jensen Jan (jhjensen@kemi.ku.dk)
University of Copenhaguen

Le Forestier Claude
Université de Montpellier, France

Marjolin Aude
CEA/UPMC, France

Martelli Fausto
Université d'Evry, France

Masella Michel (Michel.MASELLA@cea.fr)
CEA Saclay, France

Salanne Mathieu
UPMC - Université Paris 6, France

Söderhjelm Par (paer.soederhjelm@phys.chem.ethz.ch)
ETH Zurich

Vuilleumier Rodolphe (rodolphe.vuilleumier@ens.fr)
Ecole Normale Supérieur, Paris, France