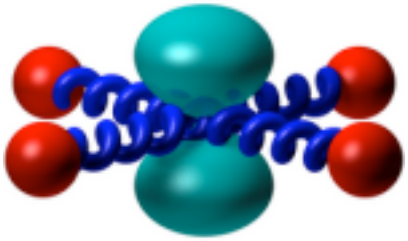
Hybrid Atomistic Methods for Materials and Biological Systems



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1 Workshop Details

1.1 Details

Timing
Number of days : 4
Start : 2006-07-10
end : 2006-07-13

Location of the activity
CECAM
46 allé e d'Italie
69007 Lyon
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1.2 Description

No description provided

2 Requested Support

CECAM

CECAM

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4 Presentation List

QM/MM simulations of proton transfer reactions and excited states properties in biological systems

Marcus Elstner
University Braunschweig, Germany

Abstract
In the last years, we have implemented the approximate Density Functional Theory Method SCC-DFTB into combined quantum mechanical molecular mechanical (QM/MM) algorithms, using several Molecular Mechanics force fields. The link atom approach is used and various of its variants have been tested so far. To include larger parts of the environment, we use a coupling of the QM/MM method to an implicit solvent treatment based on PB (GSBP). In some cases, protein polarization may be of importance and first results for the inclusion of polarization in the MM part are shown. Deficiencies of common DFT-GGA functional make the use of higher (level post-HF) approaches desirable in some cases, which leads to QM/QM/MM ONIOM type procedures. Reactions are described either with methods for global reaction path optimisation (NEB or CPR) or umbrella sampling techniques.

Enzymatic catalysis - simulation versus experiment.

Monika Fuxreiter
Institute of Enzymology, Hungary

Abstract
Enzymes can accelerate chemical events by more than 10 orders of magnitude as compared to the corresponding uncatalyzed reaction and computing the exact magnitude of the rate enhancement is an extremely challenging task. Upon targeting the experimental kinetic data all theoretical approaches face the problem of properly defining the catalytic effect that is also a key point for interpreting the results. In most cases the simulated system is limited by computer performance, rather than a rationally designed representative model is employed. Possible sources of the catalytic power will be discussed in relation to the model selection. The Empirical Valence Bond (EVB) method in combination with free energy perturbation and umbrella sampling (FEP/US) has been demonstrated to be a robust technique to handle enzymatic reactions. Using phosphoryl-transfer enzymes as illustrative examples various factors affecting the activation barrier of an enzymatic reaction will be reviewed out of which the long-range electrostatics and proper sampling are of prime importance.

Vibrational Dynamics of Impurities in Covalent Materials

Estreicher Stefan
Texas Tech University, United States

Abstract
Optical tools such as infrared absorption spectroscopy, Raman, or photoluminescence are commonly used to provide microscopic information about impurities in covalent materials such as semiconductors. This has prompted many theorists to calculate specific local vibrational modes

using first-principles techniques. However, a wealth of useful information can be obtained from the entire dynamical matrix of the system. In this talk, I will discuss the calculation of local and pseudo-local vibrational modes, vibrational entropies and free energies, vibrational lifetimes and decay channels. A simple procedure will be presented to prepare a supercell in thermal equilibrium and perform constant-temperature MD runs without thermalization or thermostat.

Extending the time scale in atomistic simulations by temperature accelerated dynamics

Francesco Montalenti
Università di Milano-Bicocca, Italy

Abstract
The development of very efficient and nicely scaling algorithms, together with the quickly increasing speed of modern computers, has made it possible to tackle several key problems in physics, chemistry, materials science, and biophysics by following an atomistic approach, i.e. by explicitly considering complex systems as made by individual atoms. While this allows one to perform static calculations characterized by a high degree of reliability, dynamics remains a problem. Depending on the level of the theoretical approach, molecular dynamics (MD) simulations can reach time scales ranging from, say, several picoseconds (ab initio) to several nanoseconds (empirical potentials), while a wide variety of fundamental experiments occurs on human time scales (seconds or longer!). In the last few years several possible ways for overcoming this problem were proposed. Here, I shall describe a promising method, called temperature-accelerated dynamics (TAD) (for a review on accelerated molecular dynamics techniques see [1]). The method, based on the harmonic approximation to the Transition State Theory, allows one to simulate rare-events based evolution at low temperatures at the typical experimental time scale. No a priori knowledge of the microscopic processes is required. After describing the key steps needed for deriving the method, I shall show some successful applications to thin-film growth phenomena [2] and to radiation-damage in MgO [3]. In the above examples, the importance of the time-scale extension and of the unbiased dynamics plays a key role in determining the simulation results. Finally, the main limitations of the method and the commonly encountered problems will be critically discussed.

References

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Extended defects formation, stability and mobility in bulk and nanostructured covalent materials

Laurent Pizzagalli
CNRS, France

Abstract
Dislocations are extended defects present in almost all materials, whose behavior (formation, stability, mobility) has a huge influence on the mechanical properties. Dislocations may also modify electronic properties of devices in heterostructures. Historically, dislocations have been essentially investigated within elasticity framework, or with classical potentials, although first principles methods are increasingly used. The main reason lies in the large computational cells that are required to deal with the long range strain field of dislocations. Here, we will present the results of several first principles studies of problems related to dislocation in covalent materials: the formation from surface defects, the stability and mobility of dislocations in bulk systems, and misfit dislocations at interface. Possible use of multiscale modelling methods will be discussed for each case.

Ehrenfest dynamics simulation of electronic damping in radiation damage
Daniel Mason
Imperial College, United Kingdom
Abstract
When a high energy neutron is fired into a fusion material, an initial billiard ball collision phase produces a displacement cascade of self-interstitial atoms and vacancies. After this a period of recombination of self-interstitial atoms and vacancies occurs, lasting

5 Poster List

Excited state properties of retinal compounds from OM2/MRCI/CHARMM
Paul Strodel
Accelrys Inc., United Kingdom
Abstract
The simulation of the photodynamics as the first steps in the reaction cycles of rhodopsin proteins continues to be a challenge for theory. Given the non-adiabatic nature of the transition process back from the electronic excited state to the ground state, a QM treatment of the chromophore, retinal, is indispensable, whereas it seems justified to describe the protein environment at the MM level. The choice of a proper QM method proves difficult for various reasons: (i) the size of chromophore system; (2) the availability the non-adiabatic coupling vector as a QM property; (3) the charge-transfer nature of the S0 to S1 transition. (4) correct description of the QM response to geometry changes and MM point charge environments. Taking the shift in the retinal absorption energy from vacuum into bacteriorhodopsin (bR) as a benchmark, we sketch that widely used QM methods have problems already at this level [Wanko2005]. This prompted us to use the valence-shell OM2 Hamiltonian[Weber2000] in combination with GUGACI[Koslowski2003] as QM method. We briefly show that in combination with the CHARMM forcefield, this method is able to explain the spectral shift between bR and sensorhodopsin. As a further test, we investigate the behaviour of OM2/GUGACI in terms of the S0/S1 potential energy surfaces along the reaction coordiante, taking a model chromophore for which ab-initio[Vreven1997] and DFT results[Wanko2004] are available. Here, the S1 minimum energy path leads into the proper conical intersection region associated with bond isomerization. Finally, we show that the new method desribes well the proper isomer-

ization reaction in S0/S1 propagations of small model compounds, as indicated by CASSCF calculations[Weingart2002]. Given these positive verifications of the new method, we are now in the position to proceed to more realistic systems.

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6 Program

Day 1: July 10 2006

Session : 1 Monday morning
09:25 to 09:30 : Welcome

09:30 to 10:10 : Presentation
Opening Talk
Ursula Roethlisberger

10:10 to 10:45 : Presentation
Embedding in covalent materials
Igor Abarenkov

10:45 to 11:20 : Presentation
Polaron and exciton localization: hard thing to prove
Alex Shluger

11:20 to 11:50 : Coffee Break

11:50 to 12:25 : Presentation
Atomistic features of selective CDK2 inhibition
Lucio Colombi Ciacchi

12:25 to 13:00 : Presentation
QM/MM simulations of proton transfer reactions and excited states properties in biological systems
Marcus Elstner

13:00 to 14:30 : Lunch Break

Session : 2 Monday afternoon
14:30 to 15:05 : Presentation
A combination of linear scaling and QM/MM techniques for the study of electron transfer reactions in biological systems and solar cells.
Joost VandeVondele

15:05 to 15:40 : Presentation
Hybrid coarse-grained/fully atomistic molecular dynamics simulations
Bernd ensing Ensing

15:40 to 16:15 : Presentation
Multiscale coupling in carbon materials
Michael Moseler

Day 2: July 11 2006

Session : 1 Tuesday morning
09:30 to 10:10 : Presentation
Linking Scales in Materials Simulation
J. Woods Halley

10:10 to 10:45 : Presentation
Boundary conditions and self-similarity
Ben Hourahine

10:45 to 11:20 : Presentation
Embedding quantum-mechanics in an interatomic potential using local energies
Noam Bernstein

11:20 to 11:50 : Coffee Break

11:50 to 12:10 : Presentation
Multiscale Hybrid Simulations of Brittle Fracture
James Kermode

12:10 to 12:45 : Presentation
LOTF QM/MM simulations applied to fracture in silicon
Tristan Albaret

12:45 to 14:30 : Lunch Break

Session : 2 Tuesday afternoon
14:30 to 15:05 : Presentation
Raman spectra of inhibitor-protein complexes
Simone Raugei

15:05 to 15:40 : Presentation
A hamiltonian electrostatic coupling scheme for QM/MM Car-Parrinello Simulations
Alessandro Laio

15:40 to 16:00 : Coffee Break

16:00 to 16:40 : Presentation
Beyond Hybrid Atomistic Modelling
Mike Payne

Day 3: July 12 2006

Session : 1 Wednesday morning
09:30 to 10:10 : Presentation
A modular approach to QM/MM calculations - ChemShell and the QUASI project
Paul Sherwood

10:10 to 10:45 : Presentation
The Growth of Copper Clusters over ZnO: the Interplay between Planar and Polyhedral Clusters
Sam French

10:45 to 11:20 : Presentation
First-principles simulations of conjugated materials
Matteo Tommasini

11:20 to 11:50 : Coffee Break

11:50 to 12:25 : Presentation
Extending the time scale in atomistic simulations by temperature accelerated dynamics
Francesco Montalenti

12:25 to 13:00 : Presentation
Ehrenfest dynamics simulation of electronic damping in radiation damage
Daniel Mason

13:00 to 14:30 : Lunch Break

Session : 2 Wednesday afternoon
14:30 to 15:05 : Presentation
Vibrational Dynamics of Impurities in Covalent Materials
Estreicher Stefan

15:05 to 15:40 : Presentation
ATP-Protein interaction: from the atomistic structure to statistical mechanics models
Pietro Ballone

15:40 to 16:00 : Coffee Break

16:00 to 16:40 : Presentation
The versatility of tight-binding
Mike Finnis

Day 4: July 13 2006

Session : 1 Thursday morning

09:30 to 10:05 : Presentation
Hybrid quantum schemes exploiting localisation
Lev Kantorovich

10:05 to 10:30 : Presentation
Extended defects formation, stability and mobility in bulk and nanostructured covalent materials
Laurent Pizzagalli

10:30 to 10:50 : Presentation
The LOTF scheme for biochemical applications
Steven Winfield

10:50 to 11:10 : Coffee Break

11:10 to 11:45 : Presentation
Enzymatic catalysis - simulation versus experiment.
Monika Fuxreiter

11:45 to 12:00 : Closing word

7 Organizer's report

7.1 Conclusions.

C0. Hybrid vs. multi-scale approaches. The way forward for some systems or for some length scales within a system may be hierarchical multi-scale modelling rather than simultaneous coupling of models into hybrid schemes.

C1. The space embedding problem. The workshop has highlighted how dimensionality plays a key role in selecting the appropriate embedding method. Methods appropriate for studying large organic molecules, because of their quasi-one-dimensional character may be sometimes less efficient, or inappropriate, for bulk solid materials, and vice versa.

C2. The MD time problem. Ever more refined/powerful accelerated dynamics schemes such as TAD have a clear potential for wide cross-disciplinary applications.

C3. Method development. The methods currently developed for materials have not yet achieved the same level of maturity of the most accomplished state-of-art biological applications.

C4. There is value to be gained in referring to energy landscapes, and in general, in trying to separate the overall system evolution/dynamics from local thermodynamics (e.g., by coarse-graining the variables set).

7.2 Recommendations.

R0 (cf. C0 above). Define where hybrid models are really necessary. A set of models (single-model computer programs) can often be used in a multi-scale hierarchical way, by just importing the relevant results from a level to the next in some suitable compact parametric form, avoiding the coexistence of two approaches in the same (therefore, hybrid) calculation. Note that hybrid approaches will still be useful, if not necessary, to validate the whole procedure. The transferability of multi-scale models is clearly an area where further analysis is needed.

R1 (cf. C1 above). Range of applicability. Guidance should be made available (through working groups, data bases, . . .) to help selecting/tuning the embedding technique most appropriate for the physical system investigated. The degree of non-locality of the embedded QM problem plays a role here as much as the kind of long-range effect considered (e.g., electrostatic, elastic, . . .) .

R2 (cf. C2 above). The present schemes should be further tested and refined so that new users know which method is the best option for their target system. Diffusion of comprehensive literature on the time problem and coarse graining techniques should be encouraged

R3. Error control, for efficiency. It is no use having highly accurate QM treatments if the errors introduced by the coupling or the environment (e.g.. for enzyme catalysis) are much higher than the QM ones. Some estimates should be made of each. There is a need for developing an unified agreed quality control procedure for the simulations. Many new complex problems may be tackled if an integrated approach could be established (including e.g., both theory and experiment in a common database) among network-connected teams who share a quality control standard.

R4. Probing the unknown. Progress is needed towards the ability to enforce Quantum Mechanical accuracy at will during the simulations, e.g., in systems sub-regions when this suddenly becomes necessary, to have truly predictive theoretical tool. The space-time subsection of interest of the system trajectory should not have to be known a priori in a simulation.

8 Key references

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