Methods of Computational Chemistry: challenges and new developments

(part of the ancillary programme at the 7th EuCheMS Congress on Chemistry)

ACC Liverpool (UK)

29 August 2018, 14:00-17:30

Programme

14:00-14:05   Péter Szalay (Chair, Division of Computational and Theoretical Chemistry)
Welcome

Session 1. Chair: Tanja van Mourik

14:05-14:35   Daniel Crawford
The Molecular Sciences Software Institute

14:35-15:05   Leticia Gonzalez
The SHARC approach for non-adiabatic dynamics

15:05-15:35   Frank Neese
Recent developments in low order scaling wavefunction based ab initio methods and their application to material science problems

***** Break *****

Session 2. Chair: Hans-Peter Lüthi

16:00-16:30   Christof Hättig
Recent developments for the RI-CC2 and RI-ADC(2) programs in TURBOMOLE

16:30-17:00   Chiara Cappelli
The Virtual Multifrequency Spectrometer (VMS): status and perspectives

17:00-17:30   Erik Lindahl
Molecular Simulation with GROMACS - Petascale & Beyond

Abstracts

The Molecular Sciences Software Institute

T. Daniel Crawford

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The Molecular Sciences Software Institute (MolSSI) is a nexus for science, education, and cooperation serving the worldwide community of computational molecular scientists - a broad field including of biomolecular simulation, quantum chemistry, and materials science. The Institute focuses on the software infrastructure, education, standards, and best-practices that are needed to enable the molecular science community to open new windows on the next generation of scientific Grand Challenges, ranging from the simulation of intrinsically disordered proteins associated with a range of diseases to the design of new catalysts vital to the global chemical industry and climate change. The MolSSI is working to enable the computational molecular sciences community to work together to leverage its diverse capabilities that will reduce or eliminate the gulf that currently delays by years the practical realization of theoretical innovations. Ultimately, the Institute will enable computational scientists to tackle problems that are orders of magnitude larger and more complex than those currently within our grasp. This lecture will provide an overview of the Institute's activities, goals, and vision.

The SHARC approach for non-adiabatic dynamics

Leticia González

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We present the newest version of the Surface Hopping including ARbitrary Couplings (SHARC) method for excited-state nonadiabatic dynamics simulations [1]. SHARC is a generalization of the popular surface hopping method, allowing simulation of the full dimensional dynamics of molecules including any type of coupling terms beyond nonadiabatic couplings [2]. Examples of these arbitrary couplings include spin–orbit couplings or dipole moment–laser field couplings, such that SHARC can describe ultrafast internal conversion, intersystem crossing, and radiative processes. The key step of the SHARC approach consists of a diagonalization of the Hamiltonian including these couplings, such that the nuclear dynamics is carried out on potential energy surfaces including the effects of the couplings [3]—this is critical in any applications considering, for example, transition metal complexes or strong laser fields . Few examples will showcase the ability of SHARC to reveal deactivation pathways in molecular systems.

[1] S. Mai, P. Marquetand, L. González, *Nonadiabatic dynamics: The SHARC approach,*Wiley Interdiscip. Rev. Comput. Mol. Sci. (in press), DOI: 10.1002/wcms.1370, (2018)

[2] M. Richter, P. Marquetand, J. González-Vázquez, I. Sola, L. González, *SHARC – ab initio molecular dynamics with surface hopping in the adiabatic representation including arbitrary couplings*, J. Chem. Theory Comput. 7, 1253-1258, (2011)

[3] S. Mai, P. Marquetand, L. González, *A general method to describe intersystem crossing dynamics in trajectory surface hopping,* Int. J. Quant. Chem. 115, 1215-1231, (2015)

Recent developments in low order scaling wavefunction based ab initio methods and their application to material science problems

Frank Neese

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In the past 5-10 years, there has been rapid progress in the development of low-order scaling highly correlated ab initio methods. Based on the concept of „domain based local pair natural orbitals" (DLPNO) 99.9% of the canonical coupled cluster energy can be recovered. This opens the door for large scale applications to molecules and large cluster models of solids and surfaces. The methodology has been extended to cover open shells, excited states, molecular properties and multi-reference wavefunctions. The talk will provide a short introduction to the elementary principles of the method and illustrate the opportunities of the methodology with recent examples.

Recent developments for the RI-CC2 and RI-ADC(2) programs in TURBOMOLE

Christof Hättig

Ruhr-Universität Bochum (Germany)

The approximate coupled cluster singles and doubles model CC2 and the algebraic diagrammatic construction through second order, ADC(2), provide for single excitation dominated transitions for closed-shell molecules an accuracy close to that of the CCSD but at much lower computational costs. This made CC2 and ADC(2) for many applications attractive alternative to time-dependent density functional theory, TDDFT.

In this talk I will present an overview of the latest developments for the RI-CC2 and RI-ADC(2) programs in the TURBOMOLE package. This includes extension for the calculation of spin-orbit induced transitions moments to triplet states for phosphorescence lifetimes and circularly polarized phosphorescence and an implementations with reduced scalings of the computational costs with the systems size using pair natural orbitals.

Another recent development is the combination of the CC2 and ADC(2) methods with the continuum solvation model COSMO and an atomistic polarizable embedding (PE). For COSMO-ADC(2) and PE-ADC(2) we implemented very recently analytic gradients for excited states to investigate structures and the relaxation dynamics of the excited states in solvents and environments like host-guest complexes.

The Virtual Multifrequency Spectrometer (VMS): status and perspectives

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The impressive advances of computer power, effective and user friendly software and graphical interfaces are leading to the development of a new generation of virtual tools able to deal effectively with the complex systems and phenomena of current interest in the study of molecular systems. Going from collections of numbers for oversimplified models toward vis-a-vis comparison between in silico and in vitro outcomes for real systems together with 3D renderings and natural interfaces should finally overcome the residual diffidence of experimentalists for computer simulations. Among those virtual instruments, we will be concerned here with the multifrequency spectrometer (VMS) our group is developing in the last few years [1], which allows vis-a-vis comparison of experimental spectra with their simulated counterparts, and interpretation of the results in terms of the interplay among different well defined effects [2,3]. The main building blocks of this tool are first-principle and semiempirical [4] models based on the density functional theory for the proper treatment of stereo-electronic effects, polarizable atomistic [5] and continuum [6] models to deal with environmental effects, perturbative treatments for describing vibrational averaging effects by nuclear motions beyond the harmonic level [7], and stochastic approaches to deal with long-time contributions [8]. The most recent developments of the tool concern NMR [9] and microwave [10] spectroscopy together with inclusion of large amplitude motions in the simulation of vibrational and vibronic spectra [11].

VMS is accompanied with powerful 3D pre- and post-processing graphical tools, named VMS Draw, which include the following features highly craved by scientists and technologists working on both academia and industry:

- Advanced graphical user interface (GUI) which offers the unprecedented possibility of overlapping and comparing computed and experimental spectra.

- An all-in-one solution for visualizing and analysing several types of calculated spectra in the whole range of the electromagnetic spectrum.

In this presentation, the status of VMS and the ongoing efforts toward increasing its accuracy and range of applications are discussed.

[1] V. Barone, WIRES 6, 86-110 (2016).
[2] V. Barone Computational Strategies for Spectroscopy. John Wiley & Sons Inc.: Hoboken, New Jersey, 2012.
[3] V. Barone, A. Baiardi, M. Biczysko, J. Bloino, C. Cappelli, F. Lipparini, Phys.Chem.Chem.Phys. 14, 404 (2012).
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[9] D. Presti, A. Pedone, D. Licari, V. Barone, J. Chem. Theory Comput. 13, 2215 (2017).
[10] D. Licari, N. Tasinato, L. Spada, C. Puzzarini, V. Barone, J. Chem. Theory Comput. 13, 4382 (2017).
[11] A. Baiardi, J. Bloino, V. Barone, J. Chem. Theory Comput. 13, 2804 (2017).

Molecular Simulation with GROMACS - Petascale & Beyond

Erik Lindahl

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Molecular Dynamics simulation is increasingly becoming a standardized technique used also by traditional experimental labs. This is in large part due to better hardware and software performance that make it possible to model realistic systems on biologically relevant timescales, but another important reason is the huge amount of work that (largely silently) has gone into making simulations more predictable, improving force fields, and not least development of techniques to provide standard error estimates for predicted properties. In this talk, I will present some of the recent and current efforts to develop highly efficient and scalable molecular simulations in the GROMACS code for a wide range of hardware including GPUs and other accelerators. I will discuss how we all need to adapt not just our programs, but also our scientific problem formulations, to make them possible to solve on the next-generation computational resources currently in development - an hopefully provide some nice tricks on the way. Finally, I will present a number of automatic or semi-automatic ensemble-based techniques that make it possible to scale very efficiently to large resources even for quite small problems, and how we are increasingly trying to use simulations to generate data that can directly be compared to experimental measurements instead of merely generating trajectories.