Extended software development workshop in quantum dynamics

Location: CECAM-FR-MOSER Maison de la Simulation
Webpage: https://www.cecam.org/workshop-0-1641.html
Dates: June 18, 2018 to June 29, 2018
Organizers: Federica Agostini, Basile Curchod, Ari Paavo Seitsonen

1 State of the art

Simulating the behavior of a microscopic system requires the development of approximations, algorithms, and computer softwares that beat the exponential growth of the numerical cost with the number of degrees of freedom to solve quantum-mechanical equations of motion. Building collaborations between theoreticians and computer scientists is, therefore, a critical step towards an optimal exploitation of the computational power available at high-performance computing (HPC) facilities, aiming at the study of molecular systems with increasing complexity.

Theoretically, the two main classes of approaches to solving the quantum molecular dynamical problem are wavepacket propagation schemes and trajectory-driven methods. The difference between the two classes lies in the way the nuclear degrees of freedom are treated, either fully quantum mechanically or within the classical approximation. In the first case, basis-functions contraction techniques have to be introduced to represent the nuclear wavefunction as soon as the problem exceeds 5 or 6 dimensions. In the second case, the nuclear subsystem is approximated classically, or semiclassically. Although leading to a loss of some information, this approximation offers the opportunity to access much larger systems for longer time-scales. In relation to trajectory-driven techniques, a significant amount of work has been proposed to recover some quantum-mechanical features via appropriately sampling the initial conditions from the Wigner distribution.

On the computational side, a large part of the cost of a calculation is spent to evaluate electronic properties. Also, the nuclear dynamics part of a calculation becomes itself a very costly computational task in the case of wavepacket propagation methods. Thus, algorithms for molecular dynamics simulations are not only required to reproduce realistically the behavior of quantum systems in general cases but also to scale efficiently on parallelized HPC architectures.

2 Training provided

The ESDW was organized in two parts.

During the first week, oral presentations focused on the more theoretical aspects of quantum molecular dynamics simulations. The points mentioned in the state-of-the-art section have been discussed, based on didactic lectures of 45 minutes, followed by 30 minutes of discussions. Each lecture provided a general overview of the presented topic, ensuring their accessibility not only to the experts in the field but also to the master and PhD students attending the workshop. In the first lecture, the multi-configuration time-dependent Hartree (MCTDH) method was introduced to the participants. MCTDH is undoubtedly among the most

successful approaches to evolve (nuclear) wavefunctions fully quantum mechanically. Still, alternative strategies for quantum dynamics simulations are continuously developed, for instance by identifying procedures to optimize the “space” where the wavefunction information is computed. In this context, the power of replacing Cartesian grids with Smolyak grids, effectively reducing the computational cost of the calculation, has been discussed in a second lecture. Various examples of trajectory-driven approaches were presented, ranging from the simplest, yet very effective, trajectory surface hopping and Ehrenfest schemes, to the more involved but indeed more accurate, coupled-trajectory mixed quantum-classical (CT-MQC), and quantum-classical Liouville equation (QCLE). One also finds, at the interface between wavepacket and trajectory schemes, methods like Gaussian-MCTDH, variational multi-configuration Gaussian (vMCG), or multiple spawning, which exploit the support of trajectories to propagate (Gaussian) wavepackets and allow to recover some of the information lost with a purely classical treatment; these strategies were introduced as well. Another theme of the theory lectures was on the sampling of initial conditions for trajectory-driven techniques, more specifically on how such sampling can be used to account for quantum nuclear effects, such as zero-point energy and tunneling, that are clearly missed in “classically”-based nuclear dynamics. In this context, approaches to sampling the Wigner distribution and the quantum thermal bath method have been presented.

The second week was dedicated to hands-on training on parallel computing. For these exercises, the supercomputer Poincaré (la Maison de la Simulation, Saclay), containing server-grade GPU accelerators, has been employed. In particular, two sessions of 3 slots each (90 minutes per slot) have been proposed on OpenACC and ScaLAPACK. Both practicals have, thus, focused on practical training of the use of parallel-computing techniques for GPUs and the use of the linear algebra library in parallel programming.

During both weeks, the afternoon sessions were entirely dedicated to code development. In particular, the participants of the ESDW have been involved in the development or finalization of the modules selected as 2018 deliverables for the ECAM work package 3 on Quantum Dynamics. The computer scientists attending the meeting provided the developers with strong support in optimizing their codes.

3 List of software development projects

The modules that have been developed at the ESWD are CTMQC, ModelLib, QCLE (single path), QQ Interface for Quantics, SHZagreb for Quantics, and OMP for Quantics.

CTMQC and QCLE (single path) are modules designed to perform excited-state molecular dynamics simulations in electron-nuclear systems. Nuclear trajectories are evolved under the effects of quantum-mechanical electrons differently, though, as CTMQC is based on the approximate quantum-classical solution of the exact factorization, whereas QCLE focuses on solving the quantum-classical Liouville equation. Both modules are currently not designed for ab initio simulations, that is, electronic-structure information is not computed on the fly but has to be pre-computed.

ModelLib is a library of model Hamiltonians that can, for instance, be employed for calculations based on CTMQC. A subset of model potentials included includes the Henon-Heiles model, phenol potentials, Tully models, vibronic models, and linear H-bond model.

The additional modules developed during the ESDW10 are interfaces and functionalities that were included in the Quantics software. Quantics is the quantum dynamics package performing MCTDH, G-MCTDH, and vMCG calculations, based on either pre-computed

potential energy surfaces or on-the-fly computed electronic structure properties. Quantics (i) has been interfaced with QChem based on the module QQ Interface, (ii) allows to perform surface hopping calculations based on the module SHZagreb, and (iii) has been optimized for parallel calculations using OpenMP based on the module OMP Quantics.

The modules CTMQC, QCLE(single path), and OMP for Quantics have been designed to be fully compatible with HPC. For most modules, a merge request has been opened and deliverables documenting them will be submitted by the end of Summer 2018 for the ECAM work package on Quantum Dynamics.

4 Participant list

Organizers

Agostini, Federica
Université Paris-Sud, Laboratoire de Chimie Physique, France
Curchod, Basile
Durham University, United Kingdom
O'Cais, Alan
Jülich Supercomputing Centre, Germany
Seitsonen, Ari Paavo
Ecole Normale Supérieure, France

Bonella, Sara – CECAM, EPFL, Switzerland
Borgis, Daniel - Maison de la Simulation, France
Calvo, Florent - CECAM-FR-RA et CBP, France
Carof, Antoine - Ecole Normale Supérieure, France
Castagna, Jony - STFC Daresbury Laboratory, United Kingdom
Christopoulou, Georgia - University College London, United Kingdom
Coretti, Alessandro - Department of Mathematical Sciences, Politecnico di Torino, Italy
Finocchi, Fabio - Institut des NanoSciences de Paris (INSP), France
Gomez Rodriguez, Sandra - University of Vienna, Austria
Hasnaoui, Karim - University Paris-Sud, LCP, France
Hochlaf, Majdi - Universite Paris-Est Marne-La-Vallee, France
Huppert, Simon - Sorbonne Université, France
Kelly, Aaron - Dalhousie University, Canada
Lauvergnat, David - Université Paris-Sud, Laboratoire de Chimie Physique, France
MacKernan, Donal - University College Dublin, Ireland
Malis, Momir - CECAM, EPFL, Switzerland
Mangaud, Etienne - Laboratoire Collisions Agrégats Réactivité, France
PLE, Thomas - Sorbonne Université, France
Robin, Carmen - University Paris-Sud, LCP, France
Sanz Sanz, Cristina - Department of Applied Physical Chemistry, Autonoma University of Madrid, Spain
Schild, Axel - ETH Zürich, Laboratory for Physical Chemistry, Switzerland
Van Haeften, Alice - UCL, United Kingdom
Vuilleumier, Rodolphe - Ecole Normale Supérieure, Paris, France
Worth, Graham A. - University College London, United Kingdom