ESDW7: Quantum MD

Location: CECAM-IRL, University College Dublin
Webpage: https://www.cecam.org/workshop-0-1407.html
Dates: July 17, 2017 to July 28, 2017
Organizers: Graham A. Worth (University College London, United Kingdom), Donal MacKernan (University College Dublin, Ireland), Sara Bonella (EPFL, Switzerland)

1 State of the art

Quantum dynamics simulations solve the time-dependent evolution equation, for the nuclei and, in non-adiabatic cases, the electrons, of a molecular system and can follow its fundamental behaviour including, in principle, all quantum effects. These are often crucial for simulating fundamental reactivity, e.g. after a molecular collision, or the absorption of a photon, and are required for the understanding of many state-of-the-art experiments. This understanding is also becoming important for emerging technologies that need optimal properties of materials, for example in photo-activated technology molecules need to be engineered such that the energy flow into destructive pathways is suppressed. Other obvious uses are in technologies that rely directly on quantum properties, such as quantum computing.

Despite considerable advances in algorithms over the last couple of decades, quantum dynamics simulations still require large computational resources and suffer from poor scaling with respect to system size. Accurate calculations are restricted to treating only a few atoms. The field is also relatively fragmented, with few standard benchmarks and most work done as single codes within a research group. This complicates assessing the usefulness of approximate methods and comparing systematically their performance. The situation can be contrasted to the more mature field of quantum chemistry, in which the electronic problem is solved for static nuclei, where people work within the framework of a few large packages (Gaussian, Molpro, Q-Chem etc) with recognised benchmarks.

The field is however starting to change, with greater collaboration among the leading players, spurred also by a recent series of thematic workshops and schools held in the CECAM network. E-CAM is contributing to this transition via actions aiming at systematic development of community software and dedicated state-of-the-art workshops.

2 Training provided

Training included lectures on scientific and HPC subjects (see also program).

The scientific topics included basic presentations of state-of-the-art performances of quantum dynamical algorithms and examples of their applications. Sparse grid based methods, to be used as a reference for current and new approximate methods and for exact solution of low dimensional problems, were also presented. A dedicated lecture was reserved to survey the current situation in potential parametrization at the empirical level and via machine learning. The problem of matching quantum dynamical methods with accurate first principle evaluation of the energy, forces, and non-adiabatic couplings was also discussed with reference both to current methods' performance and the characteristics of available, scalable, electronic structure packages.

A topic of specific interest for the participants was the description and implementation of mixed quantum-classical methods. Two lectures and several discussion sessions were devoted to this and two main methods were considered: Wigner-Liouville dynamics and the exact factorization approach. These methods were selected also in view of specific modules to be developed at the workshop.

The software development training was provided by the E-CAM Software Manager (Alan O'Cais) and Software Developer (Liang Liang), in collaboration with experts from the Irish Supercomputing Center ICHEC. The topics covered by the E-CAM experts included all the main tools deployed for software creation and development (the structure and operation of the GitLab repository and EasyBuild), performance analysis (Scalasca) and benchmarking (JUBE). Dr. O'Cais and Liang also provided continued assistance to the software development teams at the workshop with particular focus to identifying effective parallelisation strategies for the quantum dynamics E-CAM codes PaPIM and Quantics. The training provided by the ICHEC experts focused instead on a four-part tutorial on OpenMP parallelization, which included hands on sessions.

The main scientific and strategic outputs of the meeting were:

- Mixed quantum-classical methods have consolidated their position as a well-established subtopic in quantum dynamics. The implementation of community level actions to ensure that this transition adopts fair evaluation criteria of the different approaches and is accompanied by a more systematic development of reliable, efficient, and sustainable software packages is timely. The EINFRA centres, and E-CAM in particular with its dedicated work package on quantum dynamics, can play a crucial role in this phase.
- Although some important technical difficulties remain to enable large scale calculations, recent progress and expert use of computational resources should rapidly favour progress. In particular:
- Most mixed quantum-classical methods are based on the simultaneous propagation of multiple (hundreds of thousands of) independent trajectories each contributing to the final result via a weighted average computed at the end of the run or at fixed time intervals during the dynamics. This structure is ideally suited for massive parallelization and can exploit advanced High Throughput Schemes very effectively. The current developments in this area in E-CAM provide a unique opportunity for collaboration and implementation of new computational strategies and should be fully exploited.
- First principles calculations of the electronic structure will also require advanced computational strategies and the optimal exploitation of HPC architectures. The added cost of the electronic structure evaluation, with its specific parallelization requirements, in fact, will probably lead to implementations combining the relatively trivial massive parallelization on the ensemble of trajectories with multi-nodal evaluation of the forces along each trajectory. Hybrid architecture exploitation (employing accelerators or GPUs for the electronic structure) should be explored.

3 List of software development projects

The following software development projects were pursued:

1. Exact methods:

1.1. Implementation of iterative schemes for eigensolvers, Lanczos and Davidson (leader G. Worth)

1.2. Implementation of sparse (Smolyak) grids for exact dynamics via transformation between grid and basis representation (leader D. Lavergnat)

2. Trajectory based methods:

2.1. Path Sampling in the Wigner-Liouville dynamics, implementing new sampling methods aimed at mitigating the sign problem and optimizing convergence (leader D. MacKernan)

2.2. Developments of the PaPIM E-CAM code (leader S. Bonella):

2.2.1. OpenPM parallelization in the initial condition sampling modules

2.2.2. New sampling schemes for initial condition based on Langevin dynamics

2.2.3. Inclusion of the quantum thermal bath method in the suite of available integration schemes in the code

2.2.4. Development of an interface (prototype with CP2K) for effective interfacing with first principle electronic structure codes

2.3. Developments of the Quantics E-CAM code (leader G. Worth):

2.3.1. Interface with non-adiabatic surface hopping software package

2.3.2. Interface with Q-Chem package for first principle electronic structure

3. Exact factorization methods (leader F. Agostini)

3.1. Integration of the method in the CPMD package for first principle electronic structure

3.2. Implementation of the algorithm for evolution of nuclear equations of motion.

Modules of industrial interest. After discussions with our partner, IBM, the following modules are being developed to assist design of superconducting Qubits:

1. LocConQubit: a toolbox of subroutines and functionalities that implements Local Control Theory for the construction of control pulses for tuning universal quantum gates.
2. OpenQubit: patch to the LocConQubit suite to include effects of the environment in the simulation of the quantum gate.

The modules above (at various stages of development at the writing of this report) will be integrated in the E-CAM Quantum Dynamics modules, deliverables D3.3, D3.4. All modules are stored on our E-CAM software repository (see under Quantum Dynamics Modules).

4 Future plans

A second face to face meeting took place March 21-23 2018 at University College Dublin. The meeting had two main objectives: (1) to ensure that the merging of modules in the E-CAM repository planned to fulfil the requirements of Deliverable D3.3 had been successfully completed and/or to resolve any pending issues in that connection; (2) discuss continuation of work and start planning calendar for new modules and their delivery also in view of the third ESDW in quantum dynamics, to be held at Maison de la Simulation, Paris in June 2018.

Both tasks were accomplished with no significant difficulties.