Next generation quantum based molecular dynamics: challenges and perspectives
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State of the art:

Molecular dynamics (MD) simulation is a powerful computational tool that is widely used in materials science, chemistry and biology. MD simulations provide a general approach to understand and analyze a wide range of material systems and serve as a valuable complement to support and interpret experiments. MD simulations allow us to directly "see" and manipulate phenomena at the atomic scale and evaluate how properties can be modified to optimize specific designs.

The field of MD is currently dominated by classical simulation methods, where the interactions between atoms are described through classical force fields in which the complex electronic structure of molecules and solids is reduced to simple, parameterized, interactions between atoms that are fitted to experiments and/or electronic structure calculations. Unfortunately, classical MD simulations can be applied only to a limited set of materials, and are often not reliable outside the initial parameterization scope. This is especially problematic in situations when there is significant interatomic charge transfer between atoms, or there is covalent bond formation or breaking. In fact, for many important problems a meaningful parameterization may not even be possible. A quantum based MD therefore offers not only a more accurate and trustworthy replacement of classical MD, which is critically needed, but it also provides an essential path to modeling systems and phenomena that are clearly beyond the reach of existing, classical methods. Unfortunately, QMD simulations remain intractable for most systems because of their computational complexity.

The Workshop was successful in interconnecting different communities involved in quantum-based molecular dynamics modeling working on large-scale atomistic simulations of materials and devices, regardless the specific scientific subarea of interest. The workshop focused on most recent developments of computational and theoretical methods and techniques stimulating an intense dialogue between ab-initio, semi-empirical, empirical and classical method communities.

The program consisted of 30 invited talks of 40 minutes (35+5) each and a poster session presenting 28 contributed posters. In addition, social events were held (including a reception and conference dinner) to allow for informal exchanges. The invited talks were given by well-established scientists from different theoretical communities from all around the world, which acted as platform for interesting cross-/interdisciplinary discussions. The invited talks were followed by a poster session where the younger participants could show their scientific work and exchange of ideas with a broad knowledge in computational chemistry, solid state physics and computational materials science. The organization was very compact with the scientists accommodated in the same hotel fostering exchange and discussion between the participants also outside the meeting room.

The following emerged as key topics:

a) **Fast quantum based MD methods** (Stable, energy conserving algorithms that reduce the computational overhead of SCF optimization to a minimum and that can be combined with O(N) sparse matrix schemes that can be efficiently parallelized for the current generation of high performance computers.) A key problem with the size of the computational pre-factor that so far has limited many practical applications of O(N) based QMD simulations was addressed in several of the discussions as well as how stability can be achieved even in the limit of vanishing SCF convergence.

b) **Low pre-factor Order-N electronic structure algorithms** (Methods that have a computational complexity that scales only linearly with the system size, instead of the cubic scaling of traditional methods. In this respect a low pre-factor can be achieved by applying sparse matrix algebra and new parallelization strategies.) A number of O(N) approaches to efficient parallelism, both over shared and distributed memory platforms were presented as well as different algorithms to achieve linear scaling. Some of parallel linear scaling electronic structure implementations demonstrated rapid recent advances by possibly as much as an order of magnitude in run time performance compared to state-of-the-art less than a year ago. This topic was covered by representatives of five of the most important method development groups.

c) **Approximate DFT-methods** (Self-consistent charge density functional based tight-binding methods and semi-empirical schemes that can reduce the computational overhead compared to direct DFT methods by about 2 orders of magnitude for suitable systems.) Progress with the implementation of an extended Lagrangian Born-Oppenheimer formalism were presented that further reduces the computational overhead in approximate DFT methods by an order of magnitude. New approaches to combine approximate DFT based QMD schemes with linear scaling electronic structure theory were also presented.

d) **Quantum-derived many-body potentials** (Bond-order, Learn-on-the-fly and Neural Network potentials, machine learning for parametrization. These enable enhanced transferability over classical potentials.) New methods, for example, how neural networks can be trained to represent complex interatomic potential energy surfaces by including long range forces were presented, as well as recent progress with bond-order potentials.

e) QM/MM hybrid approaches (Multi-scale approaches where the effects and responses of the extended surroundings are modeled with more approximate but computationally faster classical force fields, while retaining quantum mechanical accuracy for the more complex core region(s).)

f) Accelerated MD for bridging time scales (Techniques to extend the effective time scale of MD simulations as well as methods to boost MD driven sampling of the phase space.)

g) Advanced integration schemes for MD (Multiple time step algorithms, parallelised integrators for dynamics, path-integral formulations of quantum nuclear motion)

h) Challenges in Computational Science / hybrid architectures (Formulation and design of algorithms for large-scale QMD simulations that take full advantage of advanced massively parallel hybrid architectures, and look towards the next generation of > 100 Petaflop parallel architectures.)

The invited talks and intense discussions contributed to define the state of the art at a computational level, regarding performance, scalability, parallelisation on different platforms and hence range of applicability of different quantum based MD techniques. Common targets for method developments between different communities have been discussed, focusing on hierarchical and domain multi-scale coupling techniques, interconnection between electronic structure and classical methods.

The Workshop Speakers and Participants agreed to have a similar meeting in 2 to 3 years from now to evaluate the rapid progress in this field.

Software infrastructure:

There are several open source and commercial software solutions for classical and quantum mechanical based MD simulations. The workshop recommended developing repositories of highly accurate and open quantum based data for validation of more approximate methods (such as DFTB and classical potentials). Vice versa parameter sets for the approximate methods could also be stored in such a repository.

The participants believe that the scientific exchange of researchers from different disciplines is key to make progress in the most challenging problems of theoretical materials and bio-systems modeling. This exchange can be achieved by interdisciplinary conferences and workshops. Moreover, it would be desirable to enable exchange not only at the level of researchers, but also at the level of PhD and Masters students.

The field of theoretical materials modeling relies strongly on the availability of state-of-the-art computer code and computing resources. Therefore, the continued funding for the development of modeling software is an important infrastructure requirement: Only when codes are freely available can new ideas be tested and developed by a broad community of researchers. Since new codes that address the most challenging materials and bio-systems applications will push the limits of current supercomputing resources, it is also important that funding of these resources remains significant in the future.

Funding:

The workshop identified research areas that might be funded by the EU 2020 program particularly through the EINFRA and COST programs, and workshop participants would support an application involving multi-scale applications of Quantum based Molecular Dynamics to either of these programs. Modeling of materials stability, interface design and bio-molecular interactions could form areas of particular focus.

Industrial and Societal interest:

Progress in the field of molecular dynamics is of great interest to Industry (representatives of Atotech Deutschland GmbH and the Fraunhofer Institute for Mechanics of Materials being present at the workshop for example). Classical force-field MD is ubiquitous in fields such as pharmaceutical design (e.g. Unilever, …) and more general bio-science (Procter & Gamble), targeted surfactants (e.g. BP, Schlumberger) and nano-fluidics (?). Other developing areas of industrial interest include high energy density materials (explosives) with coupling of mass and heat transfer to very fast chemical reactions; battery technologies and energy storage, specifically for applications to car manufacture; chemical corrosion driven crack propagation in engineering materials.

Further developments in these areas require advancement to the quantum MD level of modeling. Transferability demands and increased chemical complexity also require robust parameter free approaches (or methods that transfer smoothly to more approximate fast methods), but the challenges of long time scales and large relevant phase spaces also require advanced integration and sampling techniques. Molecular dynamics applications are currently substantial users of present high performance computing (featuring on the roadmaps of several national HPC programs), but migration to near term platforms that require efficient use of even more processors presents immanent new challenges. Robust, open, and most importantly validated, software and methods are essential to reliably drive future developments and applications in this field.