

Multiscale physics, from quantum mechanical simulations of atoms and molecules to molecular dynamics

What is presented here is a large project which involves several Master theses projects. The various projects are listed below, opening up for several possibilities.

The aim of a program on multiscale physics is to develop a first principle approach to systems of relevance for a variety of fields, from materials science to nano-technology and biological systems and even atomic nuclei and stars. Common to all these systems is that they entail a truly multiscale physics program that involves a proper understanding of the links between the various scales, starting from quantum-mechanical first principle studies of atoms, molecules and eventually other spatially confined systems to Density functional theories and finally microscopically derived potentials to be used in molecular dynamics calculations.

The computations required for accurate modeling and simulation of large-scale systems with atomistic resolution involve a hierarchy of levels of theory: quantum mechanics (QM) to determine the electronic states; force fields to average the electronics states and to obtain atom based forces (FF), molecular dynamics (MD) based on such an FF; mesoscale or coarse grain descriptions that average or homogenize atomic motions; and finally continuum level descriptions. By basing computations on first principles QM it is possible to overcome the lack of experimental data to carry out accurate predictions with atomistic resolution, which would otherwise be impossible. Furthermore, QM provides the fundamental information required to describe quantum effects, electronically excited states, as well as reaction paths and barrier heights involved in chemical reactions processes. However, the practical scale for accurate QM today is $\sim 1,000$ atoms per molecule or periodic cell (a length scale of a few nanometers) whereas the length scale for modeling supramolecular systems in biology may be in the tens of nanometers, while elucidating the interfacial effects between grains in composite materials may require hundreds of nanometers, and modeling turbulent fluid flows or shock-induced instabilities in multilayered materials may require micrometers. Thus, simulations of engineered materials and systems may require millions to billions of atoms, rendering QM methods impractical. Nonetheless, QM methods are essential for accurately describing atomic-level composition, structure and energy states of materials, considering the influence of electronic degrees of freedom. By incorporating time-dependent information, the dynamics of a system under varying conditions may be explored from QM-derived forces, albeit within a limited timescale (~ 1 ps). The prominent challenge for theory and computation involves efficiently bridging, from QM first-principles, into larger length scales with predominantly heterogeneous spatial and density distributions, and longer timescales of simulation enough to connect into engineering-level design variables while retaining the appropriate accuracy and certainty. Equally challenging remains the inverse

top-down engineering design problem, by which macroscopic material/process properties would be tunable from optimizing its atomic-level composition and structure. The aim of this large project is to develop breakthrough methods to staple and extend hierarchically over existing to develop the necessary tools to enable continuous lateral (multi-paradigm) and hierarchical (multiscale) couplings, between the different theories and models as a function of their length- and timescale range a strategy often referred to as First-Principles-Based Multiscale-Multiparadigm Simulation. The enclosed figure here displays some of these ideas. The ultimate goal is a reversible bottom-up, top-down

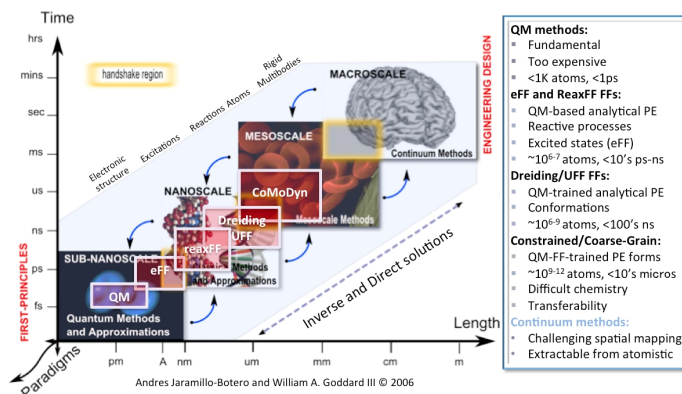


Figure 1: Example of multiscale hierarchy.

approach, based on first principles QM, to characterize properties of materials and processes at a hierarchy of length and timescales. This will improve our ability to design, analyze, and interpret experimental results, perform model-based prediction of phenomena, and to control precisely the multi-scale nature of material systems for multiple applications.

To achieve these goals, several theses projects are shortly defined below. These projects open also up for several fruitful collaborations between the involved MSc students. The group in Computational Physics has long-standing experience in defining projects where several students may find a common a ground, either from a formalism point of view or (and possibly and as well) phenomenological point of view.

There are several possible projects. The systems we have in mind are mainly molecules like SiO_2 , H_2O , CaCO_3 and other more complicated molecules. To model the interaction between such molecules and eventually derive microscopic interactions can be done via several MSc projects. The following projects can be defined and extended upon:

- Start with existing codes and thesis work by previous Master of Science students at the computational physics group. Here we think in particular of using existing Hartree-Fock codes for atoms and molecules and use these

to develop potential surfaces and thereby extract parametrized potentials for molecular dynamics calculations. One possible path is to start with a gas of hydrogen and compare the parametrized potentials from Hartree-Fock theory with existing models, as done in the thesis of Sverre-Arne Dragly <https://www.duo.uio.no/handle/10852/41843>.

- Quantum mechanical correlations are however treated only approximately in Hartree-Fock theory. Monte Carlo methods offer a path to compute exactly ground state properties of correlated quantum mechanical systems. Here one can use existing (developed at the Computational Physics group) or develop a variational and diffusion Monte Carlo code that computes properties such as binding energies, root mean square radii, charge distributions and local potentials. This topic can easily form the basis for one or two MSc thesis projects. These first principle approaches can be used to develop potential surfaces, which in turn can be used to parametrize potentials for molecular dynamics studies.
- If more complex atoms and molecules are involved, one may consider freezing the inner degrees of freedom using for example many-body perturbation theory to derive an effective interaction for electrons outside a chosen core. Alternatively one can use existing coupled cluster codes (also part of the computational quantum mechanics codes developed at the computational physics group) to derive potential surfaces and thereby parametrize potentials to use in molecular dynamics calculations.
- The quantum mechanical calculations can in turn be used to define better density functionals for the above atoms. Density functional theory can in turn be used to study for example lattices of atoms and molecules with say periodic boundary conditions.
- These effective potentials can in turn be compared with existing models such as the so-called ReaxFF potential. The implementation and studies of this potential for studies of say SiO_2 compounds could form the basis for one or two MSc thesis projects.