Multiscale Design of Advanced Materials based on Hybrid *Ab Initio* and Quasicontinuum Methods

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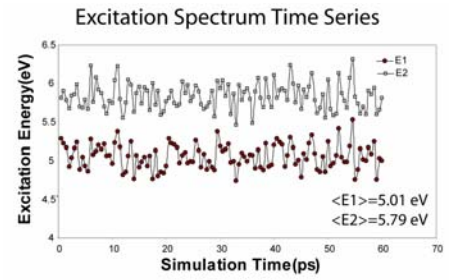
A multiscale method based on hybrid* ab initio *and the quasicontinuum method is being developed on a rigorous mathematical, physical, and chemical foundation. Rather than being based on classical interatomic potentials, this method will utilize quantum mechanics-based potentials capable of realistically describing the complex chemical bonding required to meet the design needs of advanced materials. In the past year several accomplishments were made on the project, including the development of an adaptive multilevel finite element first principles solver, extensions of the quasicontinuum method to complex crystals and objective structure, and the development of a highly accurate approach for excited states in large molecular systems at finite temperature.

This project unites researchers from mathematics, chemistry, computer science, and engineering for the development of new multiscale methods for the design of materials. Our approach is highly interdisciplinary, but it has two unifying themes: first, we utilize modern mathematical ideas about change-of-scale and state-of-the-art numerical analysis to develop computational methods and codes to solve real multiscale problems of DOE interest; and, second, we take very seriously the need for quantum mechanics-based atomistic forces, and base our methods on fast solvers of chemically accurate methods.

It is widely appreciated that to use computational methods to the design of materials encompassing a wide assortment of elements from the Periodic Table, highly efficient methods based as closely as possible on accurate quantum mechanics are needed. One of the centerpieces of this project is the development of an O(N) *ab initio* molecular dynamics method. During the last year we have developed an adaptive multilevel finite element first principles solver using the FeTK finite element libraries (www.fetk.org). The matrix



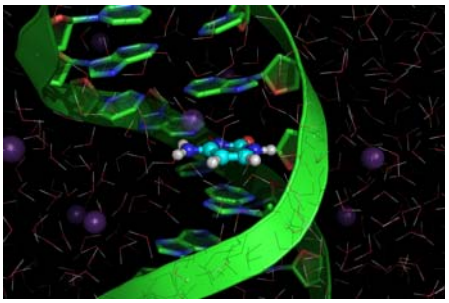


Figure 1. Dynamics of the excitation spectrum of cytosine in the native DNA environment using a hybrid coupled cluster (CR-EOMCCSD(T)) and molecular mechanics

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representations of the discrete Hamiltonian operator in the finite element basis are always O(N) sparse due to the local support nature of finite element basis functions. This development also makes use of completely unstructured simplex meshes that have the advantage of giving resolution of the near singular features around atomic nuclei using minimal computational resources. In the next year of the project, we will parallelize the method. However, with the advent of platforms like BlueGene/L, where hiding communication latency is essential, this is a difficult task, especially for software such as FeTK that use “unstructured meshes”. To facilitate this task an actor-based model for parallelism called Thyme will be used. This run-time data-driven substrate has been shown to enable the benefits of latency tolerance without having to manage the low-level details.

One of the fundamental theoretical obstacles in using adaptive methods in multiscale modeling continues to be the lack of a rigorous convergence theory for most adaptive methods. A step toward building more results along these lines was taken this year by Prof. M. Holst and colleagues L. Chen and J. Xu, where they rigorously establish both convergence and optimality of a general class of adaptive methods for mixed finite element approximations of elliptic systems.

Major extensions and capabilities are being added to the QC method of Tadmor (www.qcmethod.com). At the moment, work has focused on extending the QC to three-dimensional and complex lattices and more generally to objective structures using the very general rules-of-construction discovered by Watson and Crick and Caspar and Klug. We have shown that the whole of crystal physics can be essentially generalized using these structures. The resulting structures called "Objective Structures,” include many of the most important structures studied in science today: carbon nanotubes, the capsids and tails of many viruses, the cilia of some bacteria, DNA octahedra, buckyballs, actin and some microtubules, and certain severely bent and twisted beams. We have recently developed a methodology for computing such structures.

The ultimate goal of an atomistic-to-continuum numerical method is the capability to compute mesoscale or macroscale properties by a nearly minimum number of degrees of freedom. In crystals under moderate loads, the structure is elastic except for the cores of various singularities (dislocations, grain boundaries). Numerical indicators are being developed to detect emerging singularities in the elastic region and to track their complex evolution. More reliable and efficient thermostats for finite temperature are also being developed.

Finally, in the previous year several studies of using multiscale methods were carried out. Of particular note is a recent study performed by Dr. Marat Valiev In collaboration with Dr. Karol Kowalski (PNNL). In this work they designed a multiscale dynamical approach that combines coupled-cluster theory with molecular dynamics simulations and used it to study the dynamics of the excitation spectrum of cytosine in the native DNA environment (Figure 1).

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