Computational and Theoretical Chemistry

Portfolio Description

The Computational and Theoretical Chemistry (CTC) program supports development, improvement, and integration of new and existing theoretical and massively parallel computational or data-driven strategies for the accurate and efficient prediction or simulation of processes and mechanisms relevant to the Basic Energy Sciences (BES) mission. Focus is on non-empirical next-generation simulation of complex processes that require simultaneous computational implementation, testing, and development of new theories and algorithms. Efforts should provide fundamental solutions to problems associated with efficient conversion to clean, sustainable, renewable, novel or highly efficient energy use. Efforts must, directly or as part of multi-scale simulation methods, improve the ability to simulate processes at the molecular- and nanoscales. This includes computational and theoretical tools that enhance analysis of spectroscopic measurements or efforts aimed at enhancing accuracy, precision, applicability, scalability, or the fundamental basis of all variants of quantum-mechanical simulation methods. Developments of spatial and temporal multi-scale/multistage methodologies that allow for time-dependent simulations of resonant, non-resonant, and dissipative processes as well as rare events are encouraged. Developments of capabilities for simulation of light-matter interactions, bond breaking, conversion of light to chemical energy or electricity, and the ability to model and control externally driven electronic, magnetic, and spin-dependent transport processes in laboratory or natural/solvated environments are encouraged.

Scientific Challenges

Research in Computation and Theoretical Chemistry is fundamental to meeting the grand challenges for basic energy sciences, as identified in the recent report on this topic from the Basic Energy Sciences Advisory Committee, *Challenges at the Frontiers of Matter and Energy: Transformative Opportunities for Discovery Science* (https://science.energy.gov/bes/community-resources/reports/). Specific opportunities include:

- Improve efficiency for quantum-mechanical based simulations of chemical and molecular processes that impact the BES mission. Such improvements are achieved through multi-scale coupling, melding of chemical, physical and mathematical methods, or improved parallelization.
- Develop new theoretical time-domain and frequency-domain simulation tools for computing structural, transport, and optical properties of nanoscale systems in polarizable environments.
- Develop methods to computationally determine how to externally control both resonant and non-resonant energy, charge, spin and matter transfer processes in chemical and molecular systems with low-energy sources of radiation or applied fields, small thermal swings, and/or relatively minor changes in the external environment.

Projected Evolution

Topics of interest include practical predictive methods for (1) excited-state or collective phenomena in complex molecular systems, (2) nontraditional or novel basis sets, meshes or representation of quantum-mechanical degrees of freedom, and (3) simulation and coupling of all interactions/scales that depend upon electronic, vibrational and atomistic structure to improve descriptions of dissipative interactions due to weak and strong interactions among matter, radiation, fields, environment, and solvents.

Methods for, or applications to, systems that do not explicitly consider rearrangements of quantum-mechanical degrees of freedom are not supported.

Significant Accomplishments

This activity has played a major role in the development of quantum chemistry methodologies for accurate predictions of chemical properties. These developments have led to theories and computer codes for the calculation of thermodynamic properties and chemical reaction rates in the gas phase as well as the properties of complex molecular systems in the condensed phase. Accomplishments include the development and application of new approaches to density functional and traditional wave-function-based methods for predicting energetic processes involving ground- and excited-electronic states. These developments allow BES researchers to predict excited states in large light-harvesting complexes, address thermal and electronic transport through molecules, quantify dynamics associated with multiple carrier generation, investigate conversion of visible light into chemical energy and address plasmon-driven chemical reactions. They have also led to new approaches for non-destructive spectroscopic evaluation and interrogation of chemical conversion and separation systems and for unprecedented approaches to probing potential energy surfaces in mesoscale systems such as metal-organic frameworks.

Unique Aspects

The CTC activity is unique in its long term support of a number of fundamental chemical science areas, and in its integration of capabilities from research universities and DOE national laboratories, enabling long-term progress in difficult scientific areas as well as effective coupling to DOE missions. The CTC program is fully integrated with other BES research activities, contributing principally to the Gas Phase Chemical Physics and Condensed Phase and Interfacial Molecular Science programs, but also providing significant support to efforts spanning BES chemistry, biochemistry and geochemistry research. A unique component of this program is its support for extremely complex research that requires simultaneous development of theoretical and massively parallel computational implementation.

Mission Relevance

The CTC activity aims to advance the chemical physics goals described above and also advance mission areas across BES. For example, supported activities advance next-generation solar energy, sunlight-to-fuels, and energy storage concepts.

Relationship to Other Programs

Research under this activity complements research supported across BES and coordinates and leverages efforts with other agencies and facilities. The CTC program co-funds efforts with the Office of Advanced Scientific Computing Research (ASCR) where appropriate for the BES and ASCR missions, and has supported and participated in efforts with the technical community and other agencies to foster advanced approaches for the design of materials and chemistry. These efforts have included workshops on BES-relevant Scientific Discovery through Advanced Computing, workshops on materials and chemistry by design, and workshops aimed at understanding the increasing role of computational chemistry in industry.