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GUEST EDITORIAL

Computational and Theoretical Chemistry

Modern research in the chemical sciences seeks not only to make useful molecules and materials but to understand, design, and control their properties. Theory is at the very center of this effort, providing the framework for an atomic and molecular level description of chemical structure and reactivity that forms the basis for interpreting experimental data and provides guidance toward new experimental directions.

Great strides have been made in the development of accurate theories of atomic and molecular behavior for increasingly complex processes, in bulk states of matter as well as at interfaces. Molecular-level theory is being used to describe combustion, atmospheric chemistry, and enzymatic action. Corresponding contributions will be crucial for developing fossil fuel alternatives, for fully understanding global warming and ozone depletion, and for uncovering the molecular basis of life processes. The broad array of functional chemical structures that exist in natural materials and that are desirable in synthetic systems is widely appreciated. Progress is being made by theoretical chemists toward characterizing quantitatively the forces driving nanoscale assembly of chemical building blocks and the mechanisms by which spontaneous assembly can occur.

The calculation of static molecular structure and properties is an essential beginning, but the time evolution of molecular behavior must be understood as well. The quantitative theoretical characterization of the dynamics of chemical processes and the mechanisms behind these dynamics lies at the heart of our understanding of such fundamental chemistry as that of catalysis, where much progress is being made. The detailed description from theory of the complex chemical processes driving the sequence of events in the molecular machines of biology and the design of those targeted by modern nanoscience is a reasonable goal.

The expectation that an in-depth understanding of such complex systems is on the horizon is supported by recent history. At the outset of the 21st century, theoretical and computational chemistry has arrived at a position of central importance not only for theorists but also in the laboratories of most experimentalists and in many disciplines. These disciplines include not only chemistry but also biochemistry, chemical engineering, molecular biology, biomedical engineering, geophysics, and materials science. The prevalence of molecular calculations via quantum chemistry and the models of molecular mechanics as guidance and support for experimental research is a result of the maturation of concepts, methods, and algorithms developed over many decades within theoretical chemistry. Theoretical chemists have adapted their tools for use in industry and by experimentalists. It is then interesting to ask what new tools and deeper insights one might expect to be routinely accessible to researchers in the not too distant future. In this issue, a set of Accounts by researchers at the frontiers of theoretical chemistry are provided, which give insight into that future and reveal some of the underpinnings now being developed that will form the basis for new approaches. These Accounts include examples from analytic theory and computational studies in the areas of algorithm development and of new methods in electronic structure and in statistical mechanics.

Finally, applications that integrate state-of-the-art approaches from different areas of theory are included. The contributions describe research focused on advancing first principles quantum approaches to molecular structure and to dynamics, elements absent from most molecular modeling techniques in routine use. They address the integration of quantum electronic structure with modern methods from statistical mechanics to decipher chemical mechanisms in areas such as atmospheric chemistry and

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biochemistry. The development of algorithms to bring ever larger systems into the realm of feasible simulation is described, and analytical theory is applied to understanding potential new phenomena in nanoscale quantum devices. The systematic understanding of the interactions underlying biological self-assembly is also explored.

These Accounts certainly do not encompass all current efforts in the field nor all of the hurdles that remain but are intended to provide a perspective. Many molecular and material phenomena are characterized by multiple length and time scales. Molecules vibrate in far less than a picosecond, while chemical and biochemical processes often occur in greater than milliseconds. The length scales over which defects in solids affect material behavior range from nanometers to those visible to the unaided eye. Ultimately, multiscale theories integrating contributions from many sub-areas of theoretical chemistry should be able to describe long time, macroscopic behavior, starting purely from quantum mechanics at the atomic level. It is our belief that the outcome of such integration of theories will be a deeper understanding of the behavior of molecules, materials, and fluids. Finally, we hope that such deeper insight will allow us to address an exciting challenge to all theoretical chemists: to solve the inverse problem of property-driven synthesis—the design from first principles of molecules or materials possessing a prescribed target set of desired chemical and physical properties. We are confident that theoretical chemistry will continue to progress to this ultimate goal.

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Guest Editors

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