Quantum Simulations of Reactive Materials under Extreme Conditions

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Abstract: Understanding chemistry under high pressures and temperatures in the laboratory can require detailed atomistic and kinetic knowledge about reactants and products under these conditions. However, in many cases insufficient data exists for the equation of state and chemical reactivity of these materials under the conditions attained during experiments. Molecular dynamics (MD) simulations using quantum theories such as Kohn-Sham Density Functional Theory (DFT) hold promise as an independent route to determining simple chemical pictures of ionized intermediates and reaction mechanisms, and can help identify atomic-scale properties that determine observed macroscopic kinetics. DFT-MD simulations, though, require immense computational effort per simulation time step and consequently are usually limited to picosecond time scales and nanometer system sizes. In contrast, many chemical events occur over nanosecond time scales or longer, and experiments can probe micron length scales or beyond. In this regard, we have developed a family of semi-empirical quantum simulation methods and classical reactive MD force fields, all of which yield several orders of magnitude increase in computational efficiency while retaining most of the accuracy of quantum calculations. Our methods thus allow for direct simulation of many high-pressure experiments with quantum mechanical accuracy, where chemical events can equilibrate on timescales orders of magnitude longer than can be achieved with standard DFT. Here, we present several different applications of our models, including the pyrolysis of hydrocarbon polymers, the formation of carbyne fibers from expanding liquid carbon droplets, and the shock synthesis of life building molecules in impacting astrophysical ices. Our approaches provide a straightforward way to determine highly accurate, computationally efficient models for materials over a broad range of conditions, where physical and chemical properties can be difficult to interrogate directly and there is historically a significant reliance on quantum simulations for interpretation and validation of experimental results.

Biography: Nir Goldman received a B.S. in Chemistry from Yale University in 1997 and a Ph.D. in Physical Chemistry from the University of California, Berkeley, in 2003. He then joined Lawrence Livermore National Laboratory (LLNL) as a post-doctoral researcher, where he was promoted to the position of staff scientist in 2006. His current research interests involve the development of novel approaches to quantum mechanical simulations of chemical reactivity within condensed matter, including materials under extremely high pressures and temperatures and the astrobiological synthesis of life-building compounds under extreme thermodynamic conditions.