Wave Packet Based Statistical Approach to Complex-Forming Reactions

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Abstract

Combustion represents a key chemical process in energy consumption in modern societies and a clear and comprehensive understanding of the elemental reactions in combustion is of great importance to a number of challenging areas such as engine efficiency and environmental protection. In this award, we proposed to develop new theoretical tools to understand elemental chemical processes in combustion environments. With the support of this DOE grant, we have made significant advances in developing new and more efficient and accurate algorithms to characterize reaction dynamics.

This is an on-going grant starting in 08/01/2005, and ending in 08/14/2016. The aims of this grant were to develop new and more efficient algorithms to study dynamics of chemical reactions in gas environments such as combustion. To provide an accurate characterization of reaction dynamics, one needs to develop global potential energy surfaces that cover the reaction pathway and to carry out dynamical calculations on the potential energy surfaces. For the former, the most reliable approach is to determine the potential energy surface from ab initio calculations of the electronic energy at the relevant configuration space. This is not a trivial task, particularly in high dimensionality. In the latter, one either perform classical trajectory calculations or quantum reactive scattering calculations. while the former is computationally inexpensive, the latter provide the benchmark, especially when quantum effects, such as tunneling and resonances, are important. The calculated scattering attributes, including the integral and differential cross sections as well as the rate coefficients, can be directly compared with experimental measurements. With the DOE grant, we have made substantial contributions in both directions and significant advanced our knowledge in this field. These achievements have benefited from extensive collaborations with both experimentalists and theoreticians working in this field.

In this period, we have published 167 publications in chemical reaction dynamics, which greatly advanced our understanding of this field. The major achievements are listed below with one or more review articles associated with each item:

1. Development of a statistical model for complex-forming reactions using a wave packet method.[1]
2. Development of a wave packet based exact method for state-to-state quantum scattering for atom-diatom reactions.[1]
3. Development of the sudden vector projection model to explain mode specificity and bond selectivity in reactions.[2, 3]
4. Development of a high-fidelity neural network based method for fitting reactive and non-reactive potential energy surfaces with permutation invariance.[4]
5. Development of a wave packet based exact methods for state-to-state quantum scattering for tetra-atomic reactions.[5]
6. Application of the quantum scattering techniques to many reactions.[6]
7. Systematic investigation of detailed reaction dynamics of four-atom systems.[7]
8. Application of the ring-polymer molecular dynamics method for computing rate coefficients of gas phase reactions.[8]
9. Development accurate quantum methods for studying photodissociation dynamics, particularly those with nonadiabatic transitions.[9]

The details of research activities have been given in annual reports over the years

In summary, the support from DOE enabled us to develop new theoretical methods that significantly expanded our ability to study chemical reactions in the gas phase. The new capabilities greatly advanced our knowledge in the field of reaction dynamics.

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