ESCAPING FROM AN ULTRACOLD INFERNO: THE ULTRACOLD KRB DIMER REACTION

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EXECUTIVE SUMMARY

Ultracold chemistry, the study of reactions at temperatures below one millikelvin, offers an unprecedented opportunity for control over the outcomes of chemical reactions [1], and the ultracold molecules formed in these reactions have applications ranging from quantum computers [2] to investigations of fundamental constants of nature [3]. In this project, computational simulations of product formation in the ultracold potassium–rubidium (KRb) dimer reaction ([K2Rb2]* -> K2 + Rb2) were performed. Although ultracold reactions would typically be studied with quantum mechanics, computational simulation of time-dependent quantum mechanics is currently beyond reach for ultracold systems. Therefore, a combination of semiclassical and quantum mechanics have been employed on Blue Waters to bring simulation of ultracold chemical reactions within reach.

RESEARCH CHALLENGE

Several properties of the ultracold KRb dimer reaction make it computationally intractable with state-of-the-art quantum techniques. Cold systems entail simulation of long lengthscales, which require larger memory allocations and more computational operations. Furthermore, although the incoming and outgoing molecules in the ultracold KRb dimer reaction are cold, the reaction passes through a hot intermediate phase (Fig. 1) that entails simulation of short lengthscales. Simulation of both long and short length scales demands multiscale and multiphysics techniques. These issues are compounded by the chaotic behavior of the molecules shown in Fig. 2, which is expensive to simulate. These concerns must be addressed in order to gain an understanding of ultracold reactions, which is vital to theorists and experimentalists in ultracold chemistry as well as physicists and chemists developing ultracold technologies.

METHODS & CODES

To bring simulation of the ultracold KRb dimer reaction within reach, we make use of the fact that the reaction behaves like an ultracold inferno [4]. Although quantum mechanics is required to study the cold products, semiclassical mechanics can be used to study the extremely hot intermediate complex. Therefore, we combined quantum and semiclassical techniques to make investigation of the reaction possible. Blue Waters is then used to accelerate this research. To study the hot intermediate complex, Monte Carlo integration was performed with up to quintillions (1,000,000,000,000,000,000s) of sampling points on Blue Waters. Parallelized random matrix and R-matrix theory code was then used on Blue Waters to predict the final distribution and rate of formation of the cold products.

RESULTS & IMPACT

Preliminary results help confirm that parallelized semiclassical mechanics can be used to calculate the number of configurations efficiently in place of costly quantum mechanics. Analysis of the results of the ultracold KRb dimer reaction will directly inform ongoing experiments on the reaction, which will provide a deeper understanding of how chemical reactions occur at ultracold temperatures. This study is at the frontier of the field as it seeks to make simulation of reactions possible that are computationally intractable with existing time-dependent quantum

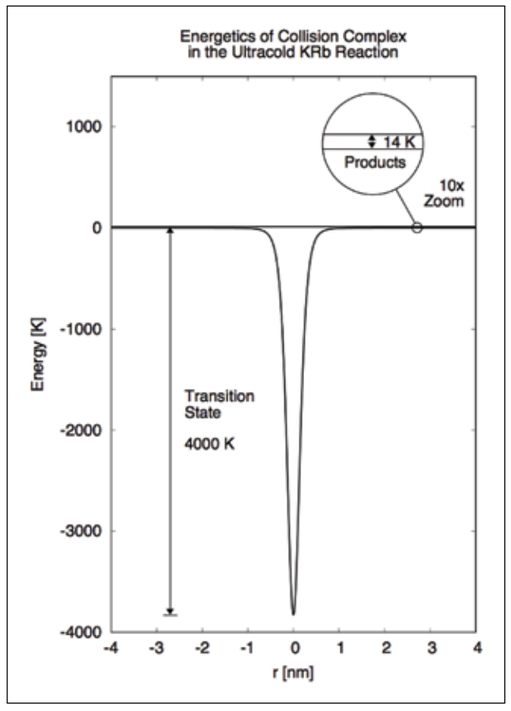

Figure 1: The ultracold KRb dimer reaction can be thought of as an ultracold inferno. In the reaction, a hot four-atom complex (4,000°K) breaks apart into two cold molecules (14°K). Magnification (10x) is shown to scale.

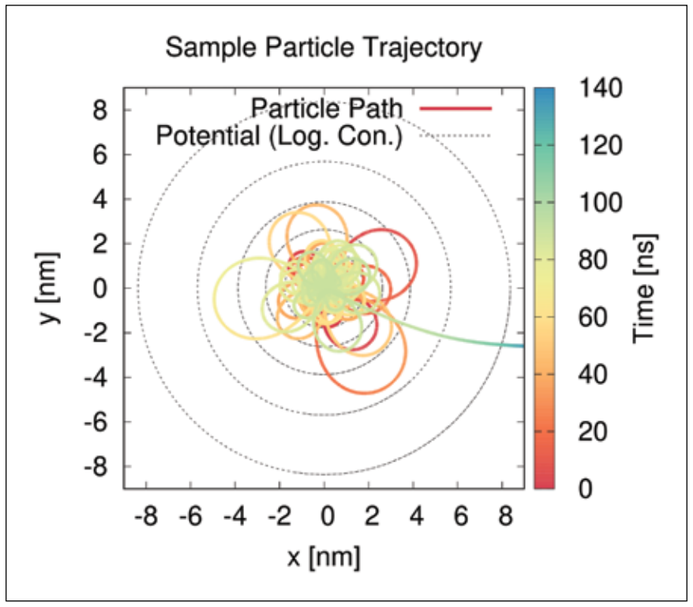

Figure 2: Computational simulation of the chaotic path of the molecules (multicolor line).

mechanical techniques. These techniques can be applied more generally to other chemical systems to help push the limits of reactions that can be studied in the field of computational chemistry. In collaboration with the results of the experimental Kang–Kuen Ni group, these computational results will help expedite development of key technologies such as quantum computers.

WHY BLUE WATERS

The Monte Carlo integration and statistical R-matrix theory codes are computationally intensive. The Blue Waters supercomputer can significantly reduce the amount of time it takes to run these calculations and provide an opportunity to perform many tests simultaneously. This speed and flexibility make calculation of statistical R-matrix theory codes practical. In addition, the Blue Waters support staff facilitate acceleration of the program speed and the program development. The hope is that Blue Waters resources will help show that it is possible to simulate ultracold chemical reactions efficiently with a combination of semiclassical and quantum mechanics.

Micheline Soley, a fifth-year doctoral candidate in chemical physics at Harvard University, works under the direction of Eric J. Heller and expects to graduate in May 2020.