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A Tribute to Donald J. Kouri

Introduction

This Tribute relates to two periods in Don's life. The first, described by Michael Baer, covers the period starting in 1970 and ends in the middle of the eighties, and the second, written by John Zhang, starts around 1985 and ends at the present time. The separation into two periods is, of course, somewhat artificial because Don has worked continuously within the theory of molecular dynamics for the last 35 years. No doubt he changed his order of priorities during this period but the changes were usually gradual and therefore it may not be easy to determine when it was exactly that he dropped one subject and started a new one. In other words, this partitioning has more to do with us and the way we knew him.

Donald J. Kouri: 1970−1985

When I screen all the members of the "older" generation in the field of molecular dynamics, Don emerges as probably the most characteristic scientist for the field of *quantum mechanical treatment of molecular dynamics*. His contributions range from inelastic through reactive bimolecular and molecular-surface processes and on to the more recent subjects related to Bose−Einstein condensation and confinement of material particles. The most characteristic feature in his doing science is to do it "*my own way*". And, indeed, his outstanding science is a direct outcome of doing things *his* way.

His starting point was the study of inelastic scattering processes between molecular species. In this respect his contribution led to the conversion of this subject from a package of sporadic papers into an active, respectable field of research. His name is associated with the two major breakthroughs in this subject: (1) Introduction of the coupled-states approximation (sometimes also termed the centrifugal sudden approximation), also known as the CS approximation, which reduces significantly the numerical effort required for deriving quantum mechanical cross sections at the expense of only a minor effect on the results. In this respect I mention his paper with Paul McGuire (*J. Chem. Phys.* **1974**, *60*, 2488), which can be considered as one of the most cited papers in molecular dynamics (∼700). (2) The "reduced dimensionality treatment" (termed in the 1970s the *infinite-order-sudden approximation* and known by its acronym, IOSA), which further facilitates the quantum calculation of the cross sections. Don was not alone in introducing these approximations but he undoubtedly had a major impact in developing and implementing them, first for inelastic bimolecular processes and later for reactive scattering,

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molecule−surface interaction, and electronic transitions. Moreover, he showed a profound insight and unusual understanding of their physical meaning, as is best realized from reading his excellent review chapter published in *Atom−Molecule Collision Theory: A Guide for the Experimentalist*, edited by R. B. Bernstein (Plenum Press: New York, 1979). Don's chapter is still considered a leading review on the theory of inelastic collisions in molecular systems. These two approximations, although introduced about thirty years ago, are still heavily used even nowadays, in particular in the study of reactive collisions.

I joined Don's group in the year 1970−71 for two reasons: (1) He intended to work on the quantum mechanical treatment for reactive scattering processes which in those days was in its infancy (and risky) and (2) he had two back-to-back articles that he published with his student Neal Sams (*J. Chem. Phys.* **1969**, *51*, 4809, 4815) in which they solved the multichannel Lippmann−Schwinger equation for inelastic scattering processes, *noniteratively*, in a most elegant way. After reading these papers, while still in Israel, I became curious about his approach and my interest grew when I found out that Don considered treating *reactive* scattering employing the Lippmann−Schwinger equation. Indeed, this is what we did during my postdoctoral year, namely, extend the Lippman−Schwinger equation to treat what in physics is termed a multi-arrangement-channel system and in physical chemistry a reactive-exchange process. This extension became known, some time later, as the Baer−Kouri equation and at a more advanced stage as the Baer−Kouri−Levin−Tobocman (BKLT) equation. As far as this equation is concerned, I mention two issues: (1) It was considered by the chemistry community as a *physical approach* (as compared with other approaches to treating reactive collisions) and, indeed, it was more popular among the physicists (see for instance: Adhikari, S. K.; Kovalski, K. L. *Dynamical Collision Theory and Its Applications;* Academic Press, Inc.: San Diego, 1991). (2) Around the middle of the 1980s, Kouri, Zhang, and others (see below) employed a modified version of the BKLT equation to derive rigorous (among the first of their kind) reactive transition probabilities and cross sections for three-dimensional molecular systems ($H + H_2$, $D + H_2$, $F + H_2$, etc.).

Toward the end of the nineteen seventies Don and I collaborated again in developing the IOSA version to treat (exchange) reactive collisions. This collaboration (together with Khare, Jellinek, and others) led to three-dimensional reactive scattering treatments of various systems and culminated in obtaining the first ever quantum mechanical reactive differential cross sections for $F + H_2$ (*Phys. Rev. Lett.* **1981** *47*, 1588), which compared reasonably well with Y. T. Lee's experiments. This calculation preceded by about fifteen years the rigorous treatments of this process.

These IOSA studies not only spread over several years but also enhanced our collaboration employing other approaches (see next section), which continued to fluorish through the 1980s and into the 1990s.

As a summary to this tribute I say the following: It is my belief that our field of molecular dynamics in general and reactive scattering in particular benefited greatly from Don's enthusiasm and insight and above all from his ingenious ideas. Moreover, I dare to say that without Don's contributions this field not only would have taken a different course, thus missing numerous beautiful theories he developed, but also would not have reached the peaks it reached with him.

Donald J. Kouri: 1985−2003

My graduate career at the University of Houston working with Don was one of the best periods in my life, which I enjoyed very much. Don was a wonderful advisor to me, both academically as well as personally. Being a graduate advisor, Don was very considerate to my needs as a student. In particular, he likes to give plenty of freedom to students and often encouraged me to explore new ideas and practice independent thinking, from which I benefited tremendously in my later career. Don likes to share every moment of joy with students whenever some new progress has been made, for which I truly appreciated being his student. Of course, what I learned and benefited from the most is Don's scientific insight and vision. The theory of quantum reactive scattering often eludes many students partly due to its uncompromising mathematical appearances. A great advantage I enjoyed is that I had this wonderful opportunity to learn the insight of reactive scattering theory from a top expert in the field. Into my third year in the graduate program, I was looking for a more exciting and ambitious project to work on as part of my thesis work. Don strongly encouraged me to tackle the 3D atom−diatom reactive scattering project by using algebraic methods to solve the Lippmann−Schwinger integral equation for reactive scattering. The original project was based on the BKLT (Baer−Kouri−Levin−Tobocman) equation that Don and co-workers developed in the seventies, but with new twists added later on to make it practical for numerical calculation. It is fair to say that without Don's strong support and encouragement, I would never have had the courage to tackle this project. What happened next is the well-known story of a successful collaboration between Don Kouri's and Don Truhlar's groups. This 3D integral equation reactive scattering project turned out to be a huge success and produced a series of benchmark results for atom−diatom reactive scattering in three dimensions. I personally benefited tremendously and learned a great number of ideas from both Dons through this collaborative project.

Don is a person with great foresight. At the time, while the time-independent approach to quantum reactive scattering was red hot, Don had another grand vision and began to promote the time-dependent (TD) quantum wave packet approach for quantum dynamics calculation. He formulated the popular coupled-channel wave packet (CCWP) approach for solving TD quantum dynamics problems. Working with postdoctoral fellow Rick Mowrey, they demonstrated the computational power of the TD wave packet approach in a paper in which tens of thousands of basis functions were included for inelastic scattering of a hydrogen molecule with a static surface. Later, by working with Michael Baer and Danny Neuhauser, they successfully extended the CCWP method to make it practical for atom−diatom reactive scattering. The application of the absorbing potential, originally introduced by Neuhauser and Baer, and later with Don, played a significant role in the success of the CCPW method. It should be pointed out that while Don was heavily promoting the TD approach as the most efficient method for future large scale quantum dynamics calculations, the majority of the reactive scattering works being done at the time were based on the time-independent approach. It is useful to note that today the time-dependent CCWP method has become almost the standard method of choice for studying complex quantum dynamics problems in the gas phase, at the gas−surface interface, etc. It should also be mentioned that the TD CCWP application is now widely applied in conjunction with the CS (centrifugal sudden) approximation, which is another powerful theoretical tool that gave Don a big name in the seventies.

Beginning in the nineties, Don found a new source of energy and branched out to many new fields of research, ranging from

quantum dynamics to applied math. A particularly important piece of his work during that period was the development of the distributed approximating functional (DAF) method that he co-developed with David Hoffman. The DAF method is in some ways related to the wavelet theory that has wide applications. Don and his collaborators have made significant progress to make DAF as versatile and practical as possible. In particular, Don has shown that DAF can be applied in many different fields such as in fitting of potential energy surfaces, signal processing, fluid dynamics, and reaction dynamics, etc. It is admitted, however, that the full potential of DAF is yet to be realized. Yet another interesting area of Don's recent research is in the fundamentals of quantum theory. Don has recently formulated a new generalization of the Heisenberg uncertainty principle, so as to include the idea of multiresolution analysis. This is important for nonlinear optics (e.g., preparation of optimal coherent light pulses), Bose−Einstein condensation and confinement of material particles, quantum computing, and many other areas. It has led to the development of generalized Gaussian and coherent quantum states. These promise to be important both theoretically and experimentally.

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