John Ross

1926–2017

BIOGRAPHICAL *Memoirs*

A Biographical Memoir by
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JOHN **ROSS**

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By Raymond Kapral

John Ross made outstanding contributions to many aspects of physical chemistry during his career. His work, combing experiment and theory, led to a deeper understanding of almost all aspects of reaction dynamics, ranging from the microscopic details of reactive collisions as revealed by molecular beam collision studies to the macroscopic properties of chemical networks and the dynamics of far-from-equilibrium reacting systems.

Ross was born in Vienna, Austria on October 2, 1926 and, as a consequence of religious persecution, immigrated to the United States before World War II. His studies at Queen's College in New York were interrupted by military service in the US Army from 1944 to 1946 where he served as a 2nd Lieutenant. After the war he returned to Queen's College and completed his Bachelor of Science in 1948.

He then went to the Massachusetts Institute of Technology for graduate work. Under Isadore Amdur he performed experimental studies of the transport properties of gases, completing his PhD in 1951. After one more year at MIT, working with James A. Beattie on gas thermometry, he went to Yale University for postdoctoral studies on non-equilibrium statistical mechanics with John G. Kirkwood. This dual background in experiment and theory served him well throughout his scientific career.

In 1953 Ross obtained his first faculty position as an assistant professor in the chemistry department at Brown University, thanks to the recommendation of Kirkwood.[1] This was an exciting period for research in reaction dynamics: it was becoming possible to probe microscopic aspects of chemical collision dynamics using a number of new experimental techniques. These new techniques allowed researchers to probe in great detail the processes that underlie the macroscopic rate laws that formed the basis for the description of chemical kinetics to that point. In collaboration with his colleague Edward Greene, he embarked on a program to study collision dynamics using molecular beam methods.

His experimental studies in this area were accompanied by theoretical papers dealing with atomic and molecular scattering theory, especially optical models for reactive chemical systems.

Ross spent the 1959-1960 academic year as a Guggenheim Fellow at the Lorentz Institute in Leiden where he wrote a wonderful paper with Peter Mazur, Some Deductions from a Formal Statistical Mechanical Theory of Chemical Kinetics, published in 1961 in *The Journal of Chemical Physics*. Using Boltzmann kinetic theory, Ross and Mazur carried out a detailed examination of chemical rate laws. This paper exploited his training in statistical mechanics and his knowledge of reaction kinetics to show the limitations of phenomenological rate theories.

In 1966, Ross joined the Chemistry Department at MIT. He served as the Frederick G. Keyes Professor of Chemistry from 1971 to 1980, served as Chairman of the Department from 1966 to 1971, and was Chairman of the MIT Faculty from 1975 to 1977. During his time at MIT his research developed in several new directions. At the beginning he was still heavily involved in molecular beam research and most of his group was carrying out experimental and theoretical studies on this topic. While never one to enter into the details of calculations, his insight was impressive. He had a knack for being able to see past the complicated details of a calculation and provide insights that helped allow the research to progress.

Ross occasionally had group parties at his house in Lexington where the MIT contingent discovered that they were only half of his group; a large number of students were still at Brown.

The theoretical component of Ross's group, along with those of Irwin Oppenheim; Robert Silbey; and, later, John Deutch, were housed in the "zoo" a windowless basement of MIT building 6. (In later years the theoreticians were promoted to a higher level.) Research meetings with Ross took place in his wood-paneled office with outer and inner rooms. The inner room had a fireplace and library and was a wonderful place for scientific discussions; in there it was easy to imagine oneself doing science in the 19th century. (There were many interesting rooms behind the doors on the long, plain corridors at MIT.)

Ross frequently remarked that this was his personal office, not that of the department chairman, so he would keep it after he finished his term as chair.

In the early 1970s, while continuing his work on molecular beam studies, Ross embarked on a new research direction: the investigation of aspects of chemically reacting systems that are driven far from equilibrium by external flows of matter or energy. Starting with a series of papers on chemical instabilities, coauthored with his post-doctoral fellow Peter Ortoleva (now at Indiana University Bloomington), he and his students went on to make many contributions to this developing field. This topic remained one of his major interests throughout the rest of his career at MIT and later at Stanford. Over the years, his research in this area focused on many areas of the diverse phenomena found in nonlinear chemical systems that are out of equilibrium. His contributions are so wide-ranging that only a few will be singled out here.

During the1970s, he recognized the importance of bifurcations that lead to oscillations or spatial patterns in chemically reacting systems. He provided one of the first theoretical descriptions of oscillatory phase dynamics and one of the first interpretations of periodic precipitation patterns in terms of instabilities of reaction-diffusion equations. Both of these developments spawned a large number of studies. One of his favorite topics was the study of the effects of external stimulation and perturbations on nonlinear chemical systems, and he used these to great effect in studies of reaction mechanisms. He carried out numerous studies of bistable, oscillatory, and chaotic states and showed how unstable states could be stabilized using external illumination with feedback and delay.

In his role as chair of the chemistry department at MIT he supported a lively and interactive atmosphere. A few examples drawn from the remarks of Irwin Oppenheim, John Deutch, and Robert Silbey in the booklet prepared for Ross's 70th birthday Symposium held at Stanford in 1997 provide hints about his personal interactions with his MIT colleagues.[2]

Both Ross and Oppenheim were in Kirkwood's group at Yale, Oppenheim as a graduate student and Ross as a postdoctoral fellow. Of this period Irwin Oppenheim wrote:

> *You know how important you were to my career – without your incarcerating me in your attic, I might never have written my thesis....I remember with great affection and joy our scientific discussions, our card games, our cooking adventures, and your joie de vivre. We were a perfect pair since you had an inexhaustible supply of jokes and I forgot the jokes as soon as you told them to me.*

John Deutch, who joined the MIT faculty in 1970 and is now an Institute Professor there, made the following comment:

> *Most of all, I appreciated your giving me the opportunity as a non-tenured faculty member, to learn from you each day when you had me drive you to and from work... All this while continuously pouring out interesting and important science, covering over the decade's major subjects: statistical mechanics, molecular beams and reactive scattering, non-equilibrium thermodynamics, and much more.*

It should be added that Irwin Oppenheim's postdoctoral fellow, Mike Weinberg, who lived in Lexington at the time, had the pleasure of driving Ross to and from work before John Deutch arrived at MIT.

Bob Silbey experienced Ross's dry humor:

> *My first year at MIT and my name, carefully painted on the outer door to my office, began to flake off, so I came to you, as head of the department to complain. You took me aside, in your avuncular manner, and informed me that there was 'tenure' paint and 'non-tenure' paint, and you were sorry to inform me that mine was the latter.*

In 1980 Ross left MIT for Stanford University where he continued research on nonlinear chemical dynamics and initiated work on other topics. He was the Camille and Henry Dreyfus Professor of Chemistry from 1983 and served as chair of the Chemistry Department from 1983 to 1989.

At Stanford, continuing work he began at MIT, he developed stochastic descriptions of non-equilibrium reacting systems on the mesoscopic level. This field is currently enjoying a high level of activity, largely because reaction dynamics occurring in biological cells cannot always be described by macroscopic mass action chemical rate laws since fluctuations arising from small particle numbers may be very strong. Ross continued to make important contributions to this area. On the macroscopic level, in collaboration with Katharine and Paul Hunt, of Michigan State University, he contributed to the notoriously difficult problem of the construction of non-equilibrium thermodynamic descriptions of far-from-equilibrium systems. Also at the macroscopic level, he showed how the logic elements of computers could be constructed from networks of chemical reactions functioning as a Turing machine.

Ross had a major interest in the determination of the mechanisms of chemical reactions from experimental data and developed new methods for this purpose. Knowledge of reaction mechanisms is central to our understanding of how chemical and biochemical systems operate. His work in this area had its origins in a number of his earlier studies, especially his work on the response of reactive systems to stochastic or periodic perturbations of the concentrations of chemical species, the observation that complex reacting networks are capable of computational logic operations, and work on stoichiometric networks to categorize species in reaction networks.

At the time, the standard methods for determining reaction mechanisms relied on the identification and study of individual reaction steps in the mechanism. Ross devised a method that utilizes the correlations among reactive species to map out chemical pathways. At Stanford, in collaboration with Adam Arkin and Peidong Shen, the method was tested experimentally on the first few steps of the glycolytic pathway, the results appearing in *Science* in 1997. Since his method provides a different perspective on how reaction mechanisms for complex systems can be determined, it can yield new insights into the operation of biochemical pathways.[3]

Ross was a strong defender of his research. On a number of occasions he remarked that one should consider scientific research like a battle campaign, and he was not shy about forcefully presenting his ideas. He always encouraged his students to speak and ask questions at conferences so that their work received due credit. He was a frequent speaker at the Gordon Conference on Chemical Oscillations and Dynamic Instabilities, the major scientific meeting on chemical dynamics far from equilibrium. Even when he was not a speaker, in keeping with his battle campaign, he always prepared a few slides to show as comments at the end of someone

else's talk dealing with material related to his own work, a practice not always welcomed by the speaker, but one that did make his point clear.

Ross thrived on collaborations with students and postdoctoral fellows, and he had a lot of them. The booklet from the conference held at Stanford in 1997 to commemorate his 70th birthday lists over 60 graduate students and 200 postdoctoral fellows, plus many other collaborators, who had worked with him to that time. And he continued to work for many years after that birthday!

He worked hard to find positions for his students and kept in touch with them over the years. He liked the telephone and it was not unusual to receive a call from him at odd times. He often had good advice to offer. On job hunting he had the following advice for Stefan Mueller, now a professor in the Otto-von-Guericke-Universität, Magdeburg: "Look, you have to pick up every pebble on your way and look under it. Never stop doing this!"[2]

Ross leaves an impressive scientific legacy and all of his many collaborators have benefited from his insight and knowledge. For instance Adam Arkin, then a postdoctoral fellow in Stanford and now a professor in Berkeley, wrote:

> *The thing I miss most being in John's group is the breadth of scholarship. Or, at least, the tendency of the group to believe it was qualified to discourse on anything…The people I met in John's group and John himself, of course, are some of the most curious, idiosyncratic, opinionated and smartest people I have ever met.*[2]

Ross's achievements did not go unrecognized and he was the recipient of many awards including the Irving Langmuir Award in Chemical Physics of the American Chemical Society (1993), the Peter Debye award of the American Chemical Society (2001), Sloan (1960-1964), and Guggenheim (1959) fellowships, Austria's Cross of Honor in Science and Art (2002), Fellowships in the American Physical Society, American Academy of Arts and Sciences, American Association for the Advancement of Science (1964), member of the National Academy of Sciences (1976), the US National Medal of Science (2000), among many others.

John Ross died on February 18, 2017 in Palo Alto after a brief illness. He is survived by his second wife, Eva, and his son, Bob, from his first marriage to Virginia Franklin. He and Eva traveled extensively and many of their adventures and meetings with other scientists are recounted in Eva's autobiography.[4]

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NOTES

1. As recounted by Ross in the special issue of the *Journal of Physical Chemistry* dedicated to John Ross on the occasion of his 70th birthday. *J. Phys. Chem.* 100:18885-18887 (1996).

2. Biographical Sketches and Reminiscences of the Graduate Students, Post Doctoral Associates, Collaborators and Colleagues of John Ross, April 12, 1997, Stanford University, compiled by Peggy Wood, Jim Sheates, and Darlene Bohm.

3. Ross's work in this area is summarized in his book, J. Ross, I. Schreiber and M. O. Vlad. *Determination of Complex Reaction Mechanisms: Analysis of Chemical, Biological and Genetic Networks*.Oxford University Press, 2006.

4. Eva E. Ross. (2006) *Making a Rose After the Diaspora*, New York: iUniverse.

SELECTED BIBLIOGRAPHY

1960 With E. F. Greene and R. W. Roberts. Variation of a chemical reaction cross section with energy. *J. Chem. Phys.* 32:940-941.

1961 With P. Mazur. Some deductions from a formal statistical mechanical theory of chemical kinetics. *J. Chem. Phys*. 35:19.

1962 With H. Mori and I. Oppenheim. Some topics in quantum statistics. The Wigner function and transport theory. In *Studies in Statistical Mechanics*, vol. I. Eds. J. deBoer and G. E. Uhlenbeck. pp. 217-298. Amsterdam: North-Holland.

1971 With T. F. George. Analysis of symmetry in chemical reactions. *J. Chem. Phys.* 55:3851-3866.

1972 With P. Ortoleva. Phase waves in oscillatory chemical reactions. *J. Chem. Phys.* 58:5673-5680.

1974 With A. Nitzan, P. Ortoleva, and J. Deutch. Fluctuations and transitions at chemical instabilities: The analogy to phase transitions. *J. Chem. Phys.* 61:1056-1074.

1976 With H. Metiu and K. Kitahara. Stochastic theory of the kinetics of phase transitions. *J. Chem. Phys.* 64:292-299.

1977 With G. C. Schatz. Franck-Condon factors in studies of dynamics of chemical reactions. I. General theory, application to collinear atom-diatom reactions. *J. Chem. Phys.* 66:1021-1036.

1980 With P. H. Richter and I. Procaccia. Chemical instabilities. *Adv. Chem. Phys*. 43:217-268.

With R. S. Berry and S. A. Rice. *Physical Chemistry*. New York: Wiley.

1981 With Y. Termonia. Oscillations and control features in glycolysis: Numerical analysis of a comprehensive model. *Proc. Natl. Acad. Sci. U.S.A.* 78:2952-2956.

1982 With S. Kai and S. C. Müller. Measurements of temporal and spatial sequences of events in periodic precipitation processes. *J. Chem. Phys.* 76:1392-1406.

1984 With E. C. Zimmermann and M. Schell. Stabilization of unstable states and oscillatory phenomena in an illuminated thermochemical system: Theory and experiment. *J. Chem. Phys.* 81:1327-1336.

1990 With K. L. C. Hunt and P. M. Hunt. Nonlinear dynamics and thermodynamics of chemical reactions far from equilibrium. *Ann. Rev. Phys. Chem.* 41:409-439.

1991 With M. Eiswirth and A. Freund. Mechanistic classification of chemical oscillators and the role of species. *Adv. Chem. Phys.* 80:127-199.

1992 With A. Hjelmfelt and E. D. Weinberger. Chemical implementation of finite-state machines. *Proc. Natl. Acad. Sci. U.S.A.* 89:383-387.

1993 With S. R. Ovshinsky and M. A. Fetcenko. A nickel metal hydride battery for electric vehicles. *Science* 260:176-181.

1995 With J. D. Stemwedel and I. Schreiber. Formulation of oscillatory reaction mechanisms by deduction from experiments. *Adv. Chem. Phys.* 89:327-388.

With W. Vance. Entrainment, phase resetting, and quenching of chemical oscillations. *J. Chem. Phys.* 103:2472-2481.

1997 With A. Arkin and P. Shen. A test case of correlation metric construction of a reaction pathway from measurements. *Science* 277:1275-1279.

2004 With M. O. Vlad and A. Arkin. Response experiments for nonlinear systems with application to reaction kinetics and genetics. *Proc. Natl. Acad. Sci. U.S.A.* 101:7223-7228.

2006 With I. Schreiber and M. O. Vlad. *Determination of Complex Reaction Mechanisms: Analysis of Chemical, Biological, and Genetic Networks*. New York: Oxford University Press.

2008 Determination of complex reaction mechanisms. Analysis of chemical, biological and genetic network. *J. Phys. Chem.* 112:2134-2143.

Thermodynamics and Fluctuations Far from Equilibrium. Berlin: Springer-Verlag.

2015 With A. D. Corlan. Kinetics methods for clinical epidemiology problems. *Proc. Natl. Acad. Sci. U.S.A.* 112:14150-14155.

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