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Biography in Honor of Roger E. Miller (1952−2005)†

Roger Erwin Miller, the John B. Carroll Professor of Chemistry at the University of North Carolina at Chapel Hill and one of the world's leading physical chemists, died of cancer on November 6, 2005, at his home in Chapel Hill. Roger was born in Kitchener, Ontario, on July 23, 1952, to Donald R. Miller (a problem-solver from whom he probably inherited his formidable experimental skills) and the late Ruth E. Peltz (who inspired him to pursue a career in science). Roger attended Kitchener Collegiate Institute High School and then, from 1971 to 1975, studied physics at the University of Waterloo in the adjacent twin city of Waterloo, Ontario. While there, he founded and led the Astronomy club of Kitchener, wrote a grant proposal to obtain the funds to build an observatory, and actually built a telescope, polishing the quite large mirror with his own hands.

From 1975 to 1980, Roger pursued first a Master's and then a Ph.D. degree at the University of Waterloo, with Terry Gough and Giacinto Scoles. It is worth noting that, at the end of his undergraduate education, Roger, like most outstanding Canadian students of that time, was advised to look south of the border to carry out his Ph.D. research. With the independence of judgment that later became one of his main characteristics, Roger first looked systematically around the university at Waterloo, liked the experiment that Gough and Scoles offered him, and decided to stay "to see if it could be done". What was this offered experiment? Gough, a spectroscopist, wanted to move beyond studying liquids, and Scoles, who at the time was working on molecular beam scattering experiments, wanted to investigate the infrared spectroscopy of molecular beams. This was a largely unexplored area, falling neatly between the microwave experiments of Bill Klemperer and Bill Flygare and the visible and UV studies of Rick Smalley and Don Levy. Roger's thesis was born from the crossing of his mentor's fields and introduced a new approach to infrared spectroscopy of molecular beams at unprecedented resolution. His studies at Waterloo established the trajectory of his later scientific endeavors, which contributed significantly to our knowledge of intermolecular forces.

† The references referred to in this biography refer to the List of Publications of Roger E. Miller published in this issue.

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After Waterloo, Roger Miller assumed the position of Research Fellow at the Australian National University in Canberra (1980−1985). Working in the research group of Bob Watts, a well-known theoretician, Roger was given the opportunity to create a molecular beam laboratory and introduce an experimental component to the group. Several months prior to his official start, Roger briefly visited Canberra and began designing his crossed molecular beam apparatus. By the time he arrived with family in tow, the equipment was well on the way to completion.

In short order, Roger, using the elegant method that he had developed at Waterloo to measure both the beam stream velocity and velocity distribution of a seeded HF beam with perpendicular and 45° intersections of the laser beam, measured the velocity distributions of HF and NH_3 molecules subliming from solid NH_4F (ref 17). Soon thereafter, Roger reported his first dissociation spectra of clusters formed from ethylene and other substituted hydrocarbons and he went on to obtain some of the first rotationally resolved cluster spectra in the 3 μm region. These studies were soon extended to other systems such as nitrous oxide, carbon dioxide, and water. In parallel with the cluster studies, Roger also completed a series of differential cross section measurements for He/HF, H_2/HF, Ar/HF, and H_2/N_2 with detailed scattering calculations to test models of molecular interactions. During his time at ANU, Roger contributed his first independent publication (ref 31) to the *Journal of Physical Chemistry*, a Feature Article, entitled “Infrared Laser Photodissociation and Spectroscopy of van der Waals Molecules”.

During his four plus years in Canberra, Roger co-supervised several research students in the Watts group, and with ANU serving as a strong focal point for quantitative molecular science in Australia, Roger and Bob Watts attracted a steady stream of leading researchers to Canberra. This influx resulted in fruitful collaborations with well-established scientists including Adi Ding, John Fenn, Gad Fischer, Brian Orr, and Udo Buck. His former colleagues in Canberra fondly remember Roger as enriching the local molecular physics and physical chemistry communities throughout Australia.

In 1985, Roger was hired as an Associate Professor of Chemistry by the University of North Carolina at Chapel Hill. He quickly established his new laboratory with excellent graduate students and postdoctoral fellows whose names are listed in chronological order in the Students and Postdoctoral Associates of Roger E. Miller in this issue. In just 3 years (1988), he was promoted to full Professor with tenure, having published 25 outstanding articles since his arrival at UNC-CH. In 1996, he was selected by UNC-CH to fill the John B. Carroll Chair of Chemistry, a position that he held until he lost his battle with cancer.

Roger had a remarkably productive career in physical chemistry. In 27 years, he published more than 200 highly significant papers, often showing the rest of us new limits for the techniques he employed and new possibilities that were thrown open by pushing those limits. In his early years at UNC, Roger extended the high resolution infrared studies of clusters in two important directions. First, he went beyond binary complexes to characterize the structures of $(CO_2)_3$ and two isomers of $(HCN)_3$ (refs 38 and 45), an important step in following the evolution of molecular properties from an isolated molecule to the condensed phase. Second, by incorporating a Stark cell in the experimental apparatus (ref 42), he was able to quantify the effect of electrostatic interactions between molecular species as a function of vibrational state. Being a physicist at heart, Roger was keenly interested in vibrational predissociation dynamics, as evidenced by measurement of mode-specific lifetimes and correlations to vibrational frequency shifts (ref 52). Through an elegant experimental approach (ref 61) with infrared laser dissociation and angular detection, Roger was able to observe J−J correlations in state-to-state dissociations. This in turn led to very accurate dissociation energies for a series of binary molecular complexes.

Roger established a new and important direction in his laboratory, the investigation of aerosols (ref 68). These studies rapidly expanded through productive collaborations, initially with outside researchers and then with UNC-CH colleague Tomas Baer, leading to over a dozen joint publications. Notable, among these, was the demonstration of depth profiling of coated aerosol particles using a variable power infrared laser to selectively vaporize the outer portions of the particle (ref 177). Although the techniques, the questions asked, and the types of information extracted in these studies were totally different from Roger’s more spectroscopically oriented projects, his insights in these studies were just as quick and perspicacious.

After learning about the characterization of pendular states in molecules (where the Stark energies exceed the rotational energy level spacings), Roger quickly realized and applied the principle to molecular clusters (ref 77). This led to a dramatic change in his approach to the study of molecular clusters in a variety of areas, orientational studies, simplification of complex spectra, identification of larger molecular aggregates, and measurement of their dipole moments. These techniques became as standard in his laboratory, as measuring vibrational−rotational transitions.

A second and equally profound change in Roger’s research evolved from helium nanodroplet spectroscopy introduced at Princeton in 1992 and beautifully developed in Göttingen in the following years. In 1995, Roger spent a sabbatical leave in Göttingen with Peter Toennies and Udo Buck, when the first high resolution spectra of molecules and clusters, encapsulated in helium droplets, were being recorded. Roger was stunned by the free molecular rotation of the species in superfluid helium droplets. He immediately recognized the great promise of this technique for studying molecular complexes and proposed using this superfluid medium as a reactor for synthesizing cluster chains of polar molecules and for the orientation of clusters in electric fields. Returning to Chapel Hill, he quickly started his own He droplet laboratory and the results that followed were among the many landmarks that Roger contributed to the field of physical chemistry. In his hands, helium droplets became an incredibly versatile technique that can now be used to study a large variety of physicochemical phenomena and systems, such as nonequilibrium clustering of molecules (refs 138 and 149), the chemical bonding of metal atom clusters (ref 161), and the chemistry of free radicals (ref 175).

Roger carried out a plethora of truly novel experiments. He asked profound questions about how molecules hydrogen-bond and self-assemble, how base-pairing and conformational dynamics occur in biomolecules, how highly reactive metals insert into chemical bonds, how chemical combustion can be probed by trapping intermediates, and how the path of a chemical reaction can be influenced with vibrational excitation. Roger’s work consistently set the highest standards for our community and produced many superbly trained graduate students and postdoctoral fellows. His talks and papers were always shining examples of scientific vision and razor-sharp thinking, delivered with remarkable pedagogy and a refreshing absence of hype. It was his experimental ability, however, that was and will remain legendary. This ability, which he was able to transmit to his students, ensured that the signal-to-noise ratio in Roger's experiments was invariably higher by an order of magnitude than that in similar experiments by others working in the same area. When asked how he achieved such performance, he would flash his famous sparkling smile and say "All you have to do is to tweak the knobs of the apparatus properly!"

Roger Miller's research was recognized with numerous awards, including the W. B. Pearson Medal in Physics (1981), the Alexander von Humboldt-Stiftung Senior Scientist Award (1995), the Earle K. Plyler Prize of the American Physical Society (1997), the William F. Meggers Award of the Optical Society of America (2000), and the Spiers Memorial Lectureship of the Royal Society of Chemistry (2001). He was a member of the Editorial Boards of the *Journal of Chemical Physics* (1998–2001) and of *Molecular Physics* (from 2003) and Editor of *International Reviews in Physical Chemistry* (from 2004). He was a Fellow of the American Physical Society, and just a few months before his death, he was elected Fellow of the Royal Society of London for the Progress of Natural Knowledge, the Science Academy of the United Kingdom.

Roger was active in The Church of Jesus Christ of Latter Day Saints, serving as the ecclesiastic leader of the student congregation on the campus of the University of North Carolina. He loved his association with young people. He loved fishing, golfing, and boating. He enjoyed woodworking and was an enthusiastic gardener. He loved music and the theater, especially plays and musicals in London. He was a devoted husband and father and especially reveled in his new role as a grandfather. His many and varied talents and interests revealed a zestful enthusiasm for all aspects of life.

Roger is survived by his wife, Deborah Ann Fraser (married in 1975); his father, Donald Russ; his brother, Gerry, and sister, Patti; his son, Lance, and daughter-in-law, Becca; his daughter, Rachel, son-in-law, Rylan Hansen, and his grand-daughter, Rylee Hansen; and his son, Roger Luke.

We are at a loss for words to describe the blow that the scientific community has experienced by Roger's departure. We cannot help feeling a certain sense of guilt that we are still able (although some slowed by age) to work and teach. The quality of research that Roger reached, routinely and apparently effortlessly, in his laboratory was not viewed with envy but with admiration. For those of us who were graced with his friendship, knowing Roger was a rare and special gift. Roger was a true prince among us, one who we will always remember and one who we will always and deeply miss.

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