



James L. Kinsey

1934–2014

BIOGRAPHICAL

Memoirs

*A Biographical Memoir by
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and Bruce R. Johnson*

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NATIONAL ACADEMY OF SCIENCES

JAMES LLOYD KINSEY

October 15, 1934–December 20, 2014

Elected to the NAS, 1991

James L. Kinsey was a pioneering physical chemist, educator, leader, and friend to generations of scientists. As a scientist, he used deep insights into spectroscopy, theory, and scattering to connect the power of spectroscopy to molecular reaction dynamics in bold new ways. As a person, he brought his rigorous intellect and keen sense of humor to every interaction as an insightful colleague, mentor, and wise friend who made an enormous difference in each life he touched.

Kinsey led research in the dynamics of chemical reactions by developing novel ways to investigate their intimate details. He performed some of the first state-to-state measurements using laser-induced fluorescence to probe individual quantum states of reaction products. He and Bob Field invented stimulated emission pumping as a means of reaching and characterizing previously inaccessible, high-lying vibrational states in polyatomic molecules, providing a new method for investigating molecular dynamics.

After a molecule gains enough energy to dissociate, under what circumstances does it immediately fly apart or linger a bit? Kinsey showed how a dynamical view of resonance Raman spectroscopy provides new insights into this process. He interpreted his data using the theoretical descriptions of these half-collisions developed by Eric Heller to learn about the dynamics of excited- and ground-state molecules.



By F. Fleming Crim,
Robert F. Curl,
and Bruce R. Johnson

Education, Professional Appointments, and Family

James Lloyd Kinsey was born in Paris, Texas on October 15, 1934, the only child of two West Texas schoolteachers, Lloyd and Elaine Kinsey. The family moved around West Texas and settled in San Angelo, where Jim grew up. After high school, he entered Rice University, which was then Rice Institute and known as the elite institution for the most talented students in the region. He set out to study chemical engineering,

but the attraction of a course in physical chemistry during his junior year turned him into a chemistry major and launched him on the scientific journey of a lifetime. After completing his undergraduate studies in 1956, he remained at Rice for graduate study and received his Ph.D., working on the microwave spectroscopy of chlorine dioxide under the direction of his fellow Texan Bob Curl.

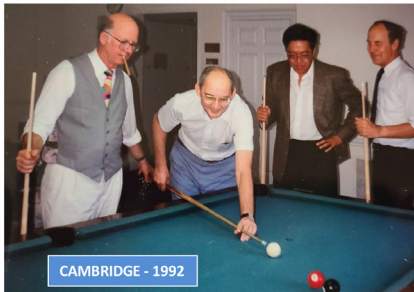
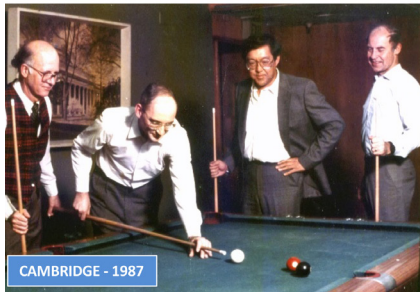
Bob had received an Assistant Professorship at Rice in January 1958, replacing George Bird, who had moved to Polaroid Corporation and left behind a microwave spectrometer and two students. Provided with this flying start to his academic career, Bob accepted immediately and flew to Houston to assess the situation at Rice, learning that Bird's spectrometer was working well and that the faculty regarded the young Ph.D. student Jim Kinsey as exceptional. Furthermore, Kinsey's thesis topic, the microwave spectrum of chlorine dioxide, introduced the rotational spectroscopy of polyatomic free radicals as a new area in need of theoretical treatment.

Jim and Bob worked their way through A. R. Edmonds' *Angular Momentum in Quantum Mechanics* to learn about the coupling of angular momentum and decomposition of reducible representations of the group of all rotations in a three-dimensional space. Understanding these Wigner $3-j$, $6-j$, and $9-j$ symbols was essential for the analysis of the rotational spectrum of ClO_2 , and the insights they acquired became a cornerstone of both of their research careers. (In a touching gesture, Jim presented Bob with his well-worn personal copy of Edmonds at the end of his research career.) In January 1959, they finally cracked the ClO_2 assignment problem, with only work on parameter fitting and publication preparation remaining. Jim had a sound Ph.D. thesis, and senior faculty at Rice, particularly Ted Lewis, successfully urged Bob to graduate his exceptional student in June 1959 to allow Jim to accept a postdoctoral appointment in Sweden.

Jim claimed that he vowed never to do spectroscopy again after he completed an NSF post-doctoral fellowship with Per-Olov Löwdin's Quantum Chemistry Group at the University of Uppsala. The publication that resulted from his sojourn in Uppsala was a treatment of the H_2^+ ion and the isoelectronic two-proton, mesonic ion without using the Born-Oppenheimer approximation (an inadequately recognized achievement). Although one cannot know how seriously he took his vow, which he certainly broke later when he did spectroscopy in the service of dynamics, spending time doing theoretical chemistry fit into the pattern of his entire career. He was a consummate experimentalist because his skills and worldview rested on deep theoretical insights. In fact, about

a quarter of his papers are theoretical, and although Jim never claimed to be a “real” theorist, his fluency with theory came through in almost every scientific conversation and paper.

The next stop was Berkeley, where Jim spent two years with the nascent research group of Dudley Herschbach as the first Miller Research Fellow in chemistry. The “band of zealots” in the Herschbach group aimed to observe the intimate details of chemical reactions using crossed molecular beams. The collection of talented coworkers and fellow zealots, such as Jim Norris and George Kwei, became life-long friends, and the intellectual ferment in the group set Jim firmly on the path of studying molecular collisions and chemical reaction dynamics. The collection of photos in the figure shows a pool game with Kinsey, Norris, Kwei, and Herschbach from 1960 that they staged again on several occasions over the years. It shows the evolving appearance and continuing friendship of the “band of zealots.”



The recurring pool game with Kinsey, Norris, Kwei, and Herschbach.

The time at Berkeley was rewarding beyond science, as Jim met an organic chemistry student, Berma McDowell, and they married in the living room of friends in the Berkeley Hills in July 1962. A home movie of a happy picnic in the summer of 1961 at Tilden Park in the hills over Berkeley with Jim, Berma, Bill Tolles, Bill's wife, and Dudley is one of Bob's mementos of that time.

That same year, Jim accepted a faculty position at the Massachusetts Institute of Technology (MIT), and their honeymoon was a cross-country trip to Cambridge to begin a career of more than 25 years at MIT. Berma and Jim shared a sense of humor and the ability to tell a good story that made them delightful company and created a warm environment for their friends and colleagues. Their family grew in Cambridge with the arrival of Victoria in 1966, Samuel in 1968, and Adam in 1970.



Jim and Berma, Berkeley, 1962.

The roles of teacher, researcher, mentor, friend, and colleague were not separate endeavors for Jim but rather an integrated whole. The rigorous and incisive thinking he brought to science permeated all of his interactions, and his humor and good will leavened every aspect. He joined a group of physical chemists at MIT that early on included Bob Silbey, John Deutch, Irwin Oppenheim, and John Waugh. Jim loved to collaborate and built notable partnerships with Dave Pritchard and Bob Field as he began melding the ideas of spectroscopy, scattering, and reaction dynamics. These topics seem inextricably linked today, but as described below, Jim's work was critical to building those connections. At MIT, Jim began scattering experiments to study reaction dynamics but found himself inevitably drawn to its spectroscopic and theoretical underpinnings. A sabbatical at the University of Wisconsin built connections to Dick Bernstein and Raphy Levine and thoughts about microscopic reversibility and information theory. In the mid-1970s, the pull of spectroscopy triumphed, as he became one of the early and most successful practitioners of laser-induced fluorescence as a probe of state-resolved reaction dynamics. Over the next decade, scattering experiments on laser-excited atoms and dimers (with Pritchard) and the invention of stimulated emission pumping (with Field) further married dynamics and spectroscopy. Thinking about dissociating molecules as undergoing "half collisions" led Jim to use the theoretical formulation of Eric Heller to understand emission spectroscopy as a means of following the evolution of dissociating molecules.

A “temporary” stint as Chair of the Department of Chemistry at MIT when John Deutch went to Washington in 1977 became a full five-year term and put Jim’s skills as a leader on full display. There are many tales of his spotting the heart of an issue and employing his dry wit to build a consensus. After proving to be both tough and fair in his role as Chair, Jim was in the running to succeed Deutch as the Dean of the School of Science, and, at the same time, Rice University beckoned once more. President George Rupp convinced Jim to come back and, in 1988, he returned to his alma mater as Dean of the Weiss School of Natural Sciences.

Jim’s return to Rice gave Bob Curl and others a colleague with whom they could discuss theory, and it was clear to everyone that Jim brought a sophisticated, keen, and realistic view of science, the university, and the world. As had been the case with his colleagues at MIT, people came to rely heavily on Jim for guidance in dealing with personal as well as technical and scientific difficulties. Bob had an experience as chair of the Chemistry Department that was similar to that of most of his friends and colleagues, who found Jim to be a tough boss, even with his longtime friend and mentor. His remarkable insight and understanding allowed him to be simultaneously a friend, advisor, mentor, and leader in ways that few others could manage.

Jim thought it was time that Rice’s Chemistry Department made plans for its future, and he arranged a weekend retreat for the department to consider the future systematically. At the retreat, future Nobel laureate Rick Smalley came forward with his vision that much of that future should focus on the development of nanoscience and nanotechnology. The willingness of the department, and later the university, to accept Rick’s vision of nanoscience as an important part of the institution’s future was crucial as Jim dealt with many attempts to lure Rick away. Ultimately, Rick decided that he could influence the nature of Rice in a way that he could not do at a larger university. Rice provided everything he asked for in terms of salary, laboratory space, support for the expensive equipment needed for nanoscience research, and a commitment to hire new faculty doing research in the area, and so Rick spent the rest of his career there. The selection of Rick Smalley, Bob Curl, and Harry Kroto for the 1996 Nobel Prize in Chemistry is just one measure of the wisdom of keeping Rick at Rice.

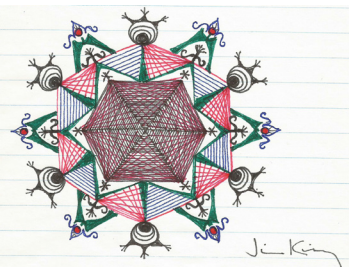
As Dean, Jim did much for the institution. He was fair and professional with faculty and was especially supportive of people starting out. He got to know everybody, especially science and engineering colleagues in the Rice Quantum Institute. Jim could also be tough when needed, and even turned down Rick Smalley on occasion, not generally a

simple task. Rick still credited Jim with always giving each of his ideas a serious hearing. Jim was an exceptional listener with the interest and intellect to appreciate a very broad range of science.

Jim served as Dean for ten years, including a year as the Interim Provost, combining his leadership skills with deep scientific knowledge. The “secret” door connecting his Dean’s office to his laboratory was a symbol of that pairing. Jim brought Bruce Johnson and Carter Kittrell with him to Rice, and their value to the Chemistry Department had a strong positive effect that continues to this day. Jim was generous in allowing others to use his laboratory and to work extensively with both Bruce and Carter, even though Jim was paying their salaries.

Both at MIT and Rice, group meetings with Jim were highly instructional, as was almost any conversation. When a student or postdoc presented interesting results, Jim was very clear in his praise, and he was equally clear about poorly constructed arguments or science that did not meet his high standards. By any measure, Jim Kinsey had a remarkable sense of humor. Students at both MIT and Rice knew that he would return copies of their theses full of red ink pointing out issues both scientific and grammatical. For example, “‘access’ is not a verb” and “the word ‘data’ is plural” (both of which used to be true). If their written misdeeds were too egregious, Jim had a series of special rubber stamps to vividly add emphasis. Junior faculty occasionally found these stamps used on drafts of proposals as well. The mildest one said merely *nihil obstat*, a phrase used by the Roman Catholic *ensor deputatus* indicating that a manuscript contains no doctrinal errors. At MIT, students coveted finding this stamp on their thesis drafts rather than the alternatives, and receiving it was a joyful event. At Rice, President Rupp, a theological historian by training, suggested to Jim that the stamp conveyed too little enthusiasm and should be retired.

Another unique practice of his was attending thesis defenses and seminars with paper and multiple colored pens, doodling while the speaker was talking. He crafted very intricate patterns during the talks and appeared not to be listening. At the end, nevertheless, he would usually ask incisive questions showing that he had been paying attention all along. He would then sign the doodle and give it to the speaker. In 1994, for his 60th birthday celebration, an elaborate surprise party with Berma keeping the secret, copies of a large number of the doodles from all over were collected in a memory book in his honor.



A Jim Kinsey Seminar Doodle.

The accompanying figure shows an example he gave Fleming Crim after a seminar at Harvard.

After his time as Dean, Jim served as Chair of the Scientific Advisory Board of the Welch Foundation from 2006 to 2012. Jim continued living life fully and died at 80 years of age on December 20, 2014, after a brief illness. He touched and enriched the lives of many through his science, his leadership, and his friendship.

Scattering, Spectroscopy, and Chemical Reaction Dynamics

Jim was a pioneer in bringing the power and rigor of molecular spectroscopy to chemical reaction dynamics, a path that his deep insights into scattering, spectroscopy, and theory allowed him to follow with both skill and determination. His keen intelligence and way with words made it all seem effortless and obvious, and Jim made the intellectual heavy lifting both inviting and fun. He had the ability to motivate, challenge, and inspire his coworkers with a casual comment or salient observation.

When Jim arrived at MIT, molecular beam scattering was still in the “alkali age,” which relied on the ability of a hot wire detector to ionize alkali-containing species with near unit efficiency. He was soon observing long-lived complexes in the scattering of K and Cs with SO_2 , CO_2 , and NO, but he also ventured into new territory by studying the reaction of TBr with K and Cs in experiments that relied on the detection of the β -decay of the tritium product collected on plates arrayed around the vacuum chamber. By detecting the lighter product of the $\text{K} + \text{TBr} \rightarrow \text{KBr} + \text{T}$ reaction, he measured the angular distribution of the reaction products and overcame the unfavorable kinematics that constrained the alkali halide to a narrow angular range in the laboratory frame. During this time, he and John Waugh were writing about molecular reorientation and spin-lattice relaxation, and he was also writing about momentum autocorrelation functions with John Deutch and Bob Silbey, his theoretical bent emerging once again. Jim wrote a particularly useful paper on the design of slotted disk velocity selectors, and he used that design in experiments to measure the total scattering cross section of D_2 by He as a function of relative speed in order to probe the intermolecular potential.

During a 1969 sabbatical in Madison, Jim thought about the state-resolved scattering experiments that were on the horizon (and which he would soon perform) and wrote

about microscopic reversibility in reactions with partial resolution of reactant and product states. This work connected closely with the information theory approach that Raphy Levine was developing, and they subsequently published a paper together, one of several on different topics. Three themes flow through Jim's work at this stage: extracting potentials from scattering data, state-resolved reactions, and extracting upper-state potentials by analyzing emission from chemiluminescent systems. Here we see the emergence of the view that spectroscopy would give insights into potentials and dynamics.

The first laser-induced fluorescence (LIF) measurements on reactive systems by Dick Zare in the early 1970s were raising exciting new possibilities, and in 1975 Jim's lab published its first state-resolved reaction study detecting the OH product from the reaction of H atoms with NO_2 , $\text{H} + \text{NO}_2 \rightarrow \text{NO} + \text{OH}(v, j)$.

The OH product was a particularly rich molecule to study. The spectroscopic resolution of individual rovibrational states in the OH product provided unprecedented detail and gave an incisive view into the dynamics of the reaction, and the population of the lambda doublet states carried information on the stereodynamics. A 1977 article in the *Annual Review of Physical Chemistry* became a touchstone for early practitioners of the art, and, characteristically, Jim used Wigner $3-j$ symbols to discuss the intensities. The $3-j$ and $6-j$ symbols always seemed to be at his fingertips, and his fluency reflected his deep understanding of angular momentum coupling. During this period, Jim spent a summer at Los Alamos National Laboratory and refined his work on using Fourier transforms of high-resolution spectral measurements of recoiling molecules to extract three-dimensional velocity distributions. This approach was a harbinger of the enormous amount of information to eventually come from Doppler-resolved measurements on recoiling molecules. The summer included camping and gathering mushrooms in the mountains with the family, and Jim loved to tell the story of a friend in Boston asking if he needed a passport to go to New Mexico!

Collaboration with Dave Pritchard from the Physics Department at MIT provided an example of using Fourier-transform Doppler spectroscopy to obtain the angular distribution in the scattering of electronically excited Na with Ar, but at least as important, it led to a series of experiments in which they and their coworkers observed the inelastic scattering of sodium dimers to obtain rovibrational-level specific scattering cross sections. Jim relished collaboration and more than once said he began a collaboration because "it would be fun to do something together." With spectroscopy now at the heart of much of his work in molecular dynamics, Jim began yet another enormously fruitful collaboration.

Lasers were clearly at the heart of experimental physical chemistry in 1981, when Jim, Bob Field, and their coworkers performed the first “stimulated emission pumping” (SEP) experiments. They used one laser pulse to excite I_2 molecules to an electronically excited state that would relax to a range of vibrational levels in the ground electronic state by spontaneous emission. In addition, they used a second laser pulse to control that process by stimulating emission to a selected vibrational state, using this stimulated emission to populate preferentially a state that was otherwise inaccessible. This approach was a powerful means of preparing states containing large amounts of vibrational energy and of opening the door to unexplored regions of the molecule’s potential energy surface. The real power of the approach lay in studying polyatomic molecules, and two—formaldehyde (H_2CO) and acetylene (C_2H_2)—proved particularly rich. The measurements produced spectroscopic constants to characterize the molecules to very high energy levels and allowed the exploration of state-resolved unimolecular reactions, energy flow in colliding and isolated molecules, and the timely topic of chaotic motion and isomerization at high levels of vibrational excitation. Once again, the *Annual Review* article that they wrote became the bible for practitioners of the art and craft of SEP. The SEP acronym also had a charming connection to the *Hitchhiker’s Guide to the Galaxy* by Douglas Adams, who also refers to SEP. In his lexicon, however, it stands for “Somebody Else’s Problem,” which was definitely not a possibility in the Field and Kinsey groups.

The theme of optical probes and preparation as a means of understanding the nature of molecules in interesting energy regimes as well as their dynamics was fully realized in Jim’s work from this era. Observing emission from electronically excited molecules in the form of resonance Raman scattering was underway in his laboratory. The idea was to use the absorption and emission of photodissociating molecules to probe the dissociation dynamics in extremely short-lived species. At MIT, Jim and his collaborators had observed very long vibrational progressions in the resonance Raman spectroscopy of ozone (O_3) and methyl iodide (CH_3I). The theoretical framework of Eric Heller allowed Jim and his coworkers to look back into the early time dynamics to make intuitive connections between the spectroscopic features and the underlying dynamics. These “half-collisions” connected the spectroscopy and the scattering dynamics, and the analysis particularly emphasized the equivalence of the time and frequency information in these experiments. Once again, Jim was looking at the world through the window of the Fourier transform.

There was some initial skepticism in the scientific community about the experiments. Because the molecules dissociate on the femtosecond time scale, faster than vibrational

periods, some argued that Raman vibrational progressions would be washed out and unobservable as a function of emission frequency. In a now-classic paper from 1984, Jim and his group reassured the community that their results were not artifacts because, in narrow-band resonance Raman scattering with well-defined initial and final molecular states, energy conservation dictates that the spectrum of emitted photons should be unaffected. Later experiments by others duplicated the results and removed any residual doubts.

The experiments produced unique information about these molecules. The broad absorption spectrum of a photodissociating molecule primarily reflects the very short-time dynamics, but the emission spectrum reveals more details that encompass intermediate times. The emission spectrum contains information about both the excited-state and the ground-state potential surfaces, and the intensities of fundamentals, overtones, and combination bands reveal properties such as forces and gradients on the excited-state surface. The observed energies and band contours of the same features reveal characteristics of the ground-state surface. The dispersed emission spectrum of ozone showed that the molecule began dissociation without bending, the signature being the absence of emission into bending vibrations of the ground electronic state. Methyl iodide illustrated a different lesson. The C-I bond elongates during the dissociation, with higher and higher overtones of the C-I stretch reflecting later and later dynamics. The progressions showing excitation of the CH₃ umbrella bending appeared later in the dissociation after extension of the C-I bond allowed the methyl group to evolve toward its planar equilibrium geometry. Jim showed how these dispersed spectra opened a window on the half-collision, and his approach illustrates his characteristic melding of spectroscopy, dynamics, and theory.

The dissociative resonance Raman results spurred further research. The group measured progressions in the case of methyl iodide at a number of distinct laser frequencies (Raman excitation profiles) and modeled their variations, providing a direct measure of the interference between overlapping excited electronic states. After moving to Rice, the Kinsey group studied iodobenzene to obtain detailed information about a very different dissociation mechanism. Another example from that time is Raman scattering from dissociating nitrosyl chloride using multiple excitation wavelengths. The experiment accessed several excited vibrational levels of the ground state for the first time, and calculations with a large-amplitude quantum mechanical vibrational model resolved disagreements among previous analyses.

During this time, Jim and his colleagues also examined the weak oscillations atop the broad Hartley absorption band of ozone by Fourier transformation and, unusually, made assignments in the time domain. The transform of the oscillations showed a series of small, early time features whose periods correlated well with a family of unstable nearly periodic orbits in the classical dynamics. The two equivalent O-O bonds start stretching in concert (still without bending), but prior to dissociation, one bond goes through a single oscillation while the other oscillates anywhere from one to four times. Experimentally, it was especially tempting to seek even more detailed, vibration-specific information about the later dynamics by measuring Raman excitation profiles of ozone. Jim's retirement intervened in what proved to be a very involved analysis. Nonetheless, others performed a number of theoretical wave packet studies along these lines. The history of his research illustrates Jim's great talents in bringing people and ideas together. A review of the dissociative resonance Raman spectroscopy research up to 1996 appeared in a special *Journal of Physical Chemistry* James L. Kinsey Festschrift issue.

His contributions brought Jim many honors and recognitions. He was elected to the National Academy of Sciences (USA) in 1991 and was a Fellow of the American Physical Society and of the American Academy of Arts and Sciences. He received the Earle K. Plyler Prize of the American Physical Society and the E. O. Lawrence Award of the U.S. Department of Energy. The American Chemical Society recognized him and Bob Field as the mentors of Yonqin Chen, who received the Society Nobel Laureate Signature Award in Graduate Education. His friends and colleagues have come together to endow the James L. Kinsey Memorial Lecture Series in the Department of Chemistry at Rice University. Modeled after Jim's own practices, the series annually chooses a speaker from among talented young scientists at early stages of their careers.

Leader, Colleague, Mentor, and Friend

Most scientists see their profession as opening doors to a rich intellectual life and to a rich collection of personal relationships. Jim combined those two features to a remarkable extent in his roles as colleague, mentor, and friend to many. All who dealt with Jim recognized that he was incredibly smart and profoundly wise. These characteristics came through particularly in his sense of humor. He was amusing (and insightful) about topics ranging from science to the state of the world to the personal, and his droll comments often went to the essence of the discussion on any topic. His approach to people and life made him an adept and joyful collaborator, and it seemed that interesting ideas would emerge from casual conversations. The note that he and Bob Silbey wrote

in the *Journal of Chemical Physics* titled “On the preponderance of near-prolate rotors among polyatomic molecules” has the feeling of a lunchtime conversation that got out of hand. Jim was an astute and sympathetic friend to colleagues at MIT and Rice and in the broader scientific community.

Jim was an effective colleague and leader because he understood both people and organizations very well. His talent for seeing the larger picture both personally and professionally made him a particularly valuable resource for young people early in their careers. He was a natural leader in both formal and informal roles with a personality and wisdom that drew people to him. He could make hard decisions and guide groups to good conclusions because he did not separate the intellectual rigor of doing science from the requirements of leadership, and he brought his considerable insight to both. Jim liked to play poker, and more than one person has said that playing poker with him, literally or figuratively, was a scary proposition. He knew how to both encourage and caution his colleagues as he advised them. The sympathy, kindness, wit, and intellect that Jim brought to the world made him truly a “*mensch*,” a person of integrity and honor. It was a term he liked and reserved for people he admired. His friends applied it to him with admiration as well.

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