



John B. Fenn

1917–2010

BIOGRAPHICAL

Memiors

*A Biographical Memoir by
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and Charles E. Kolb*

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JOHN BENNETT FENN

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Elected to the NAS, 2003

The scientific contributions of John Fenn fostered new opportunities at the interfaces of chemistry, molecular physics, fluid mechanics, thermodynamics, spectroscopy, and mass spectrometry. Most dramatic was his development of electrospray ionization, a way to produce ions in vacuo from fragile and nonvolatile molecules in solution, so that they can be analyzed by mass spectroscopy. This technique proved extraordinarily fruitful, especially for molecular biology, because it enabled high-resolution studies of proteins and other macromolecules even in complex mixtures.



John B. Fenn

*By Dudley R. Herschbach
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From youngster to PhD chemist

Although he was born in New York City and lived in Hackensack, New Jersey, for more than a decade, John spent much of his boyhood in the small town of Berea, Kentucky. His family moved there in 1928, compelled by the onset of the Great Depression. His father, a 1910 graduate of Rutgers in electrical engineering, gratefully undertook teaching industrial arts in the middle and high schools allied with Berea College, a remarkable institution established to provide educational opportunities for needy students from Appalachia, whatever their race or creed. No tuition was charged and most students worked two hours a day in maintenance of facilities or affiliated services, including a bakery, dairy, truck garden, sheep farm, piggery, furniture shop, and weaving operation.

All told, he collected data on more than three thousand solutions. That was “a boring, pointless chore...[with results that] contained no surprises.”

entered the Berea school in the eighth grade. He often cited a favorite memory from a year later, the advice, in red ink, returned on his first exam paper in algebra: “Don’t ever try to be a scientist or engineer!”

In high school, John did well in his math courses, but the only formal science course he took was chemistry and he did not find it exciting. Yet, on entering Berea College, he decided to major in chemistry, although he admitted afterward that he wasn’t sure why. His choice was soon reinforced by Professor Julian Capps, who taught an engaging freshman chemistry course. John greatly admired Capps, both as an inspiring teacher of science and a witty raconteur wont to recite Milton and Shakespeare. Long after, when John became a professor, he took Capps as his role model.

In his sophomore year of college, John qualified as a teaching assistant and was pleased to be paid eighteen cents per hour, when meals were eleven cents each and living in the college dormitory cost sixty-five cents per week. As an extracurricular escapade, John also took a four-week summer course on welding, which certified him as a master welder. He completed his bachelor of science degree in chemistry in only three years, by dint of an ambitious academic schedule, including summer courses at the University of Iowa and Purdue.

Faced with a tough job market in 1937, John applied to several graduate schools and was gladly accepted by Yale University, with a teaching assistantship that met most of his expenses. As “a wide-eyed small town youth from Kentucky,” he was awed by the splendor of Yale’s buildings. He delighted “to wander around the gardens, gargoyles, and gates,” and from his room “to look out on the forest of chimneys, slate roofs, and Gothic towers.” As was customary, each of the six new graduate students in physical chemistry was promptly assigned to a research advisor, “with no opportunity to ask questions or indicate preferences.” John was assigned to Professor Gosta “Gus” Akerlof, who proved a congenial mentor and became a lifelong friend.

But John found the project assigned to him for his doctoral research was not at all congenial. It required routine measurements of the electric potential between silver and platinum electrodes in solutions of HCl, at various concentrations and in various solvents and at various temperatures. All told, he collected data on more than three thousand solutions. That was “a boring, pointless chore...[with results that] contained no

surprises.” His doctoral dissertation consisted just of forty-five pages of tables and three pages of text.

Despite his dismaying research project, John’s final year at Yale was blissful, made so by his marriage to Margaret Wilson, whom he had met nearly a decade before in Berea. Marriage was rare for graduate students in those days. Colleagues were startled, and also dubious because Margaret was ten years older than John. He liked to point out that since Yale did not give assistantships to third-year students, Margaret was his “fellowship” that year, supporting them both. “Endearing and enduring mates,” their marriage flourished for fifty-three years.

John completed his PhD in 1940, at age twenty-three, after only three years as a graduate student. But he departed Yale with “much diminished interest in scientific research,” owing to the disdain he had for his unappealing work on electrolyte solutions. Four decades later, John relished the irony in his return to solution electrochemistry to develop the electrospray technique that led to the award of his Nobel Prize in 2002.

Interim in industry

John’s first jobs as a chemist dealt with process and product development at Monsanto in Anniston, Alabama, from 1940 until 1943, and with Sharples Chemical in Wyandotte, Michigan, from 1943 until 1945. He next eagerly and boldly accepted an invitation by a friend, Jim Mullen, to help launch a company in Richmond, Virginia, named Experiment, Inc. Mullen had worked at Bell Labs on Project Bumblebee, a wartime project to develop a ramjet-powered antiaircraft missile for the Navy, and he then obtained a subcontract to do research on related aeronautical problems. Fenn served as vice-president of the company from 1945 until 1952, and the experience proved to be a springboard for his career.

He learned much about dynamics and thermodynamics of compressible fluid flow and combustion. With Mullen, Fenn published his first research paper, which dealt with ignition of flames in high-speed flow. It appeared in 1949, almost a decade after he received his PhD. Further papers led to “some recognition in the combustion community.” His company prospered and had a significant role in development of the first successful ramjet propulsion system. Fenn found, “in an about-face from my feelings at Yale,” that he enjoyed doing research.

His combustion research led to contacts that resulted in appointment from 1952 until 1962 as director of Project SQUID, a program administered for the Navy by Princeton

University to sponsor pure and applied research in fields of science related to jet propulsion. The program was named after the jet-propelled decapod of the sea. Some thirty laboratories in government, industry, and universities participated via subcontracts from SQUID during the decade John served as director. His role involved much travel, visiting laboratories, and interacting with many scientists in many fields, as well as people in the Office of Naval Research (ONR) and other agencies sponsoring research.

In 1955, John served for a year in the London branch of ONR as a liaison officer in combustion and propulsion. In effect a sabbatical, the interlude in London was a cherished adventure for his family. The children (Marianne was aged 13, Barbara was aged 11, and John, Jr., was aged 9) much enjoyed their schools and roaming London with Margaret while John visited laboratories in several countries, where he hoped to find more incisive techniques to study fluid dynamics and chemical kinetics.

Inadvertently, by leafing through journals, John learned of recent exploratory work on “supersonic nozzle” or “free jet” molecular beams formed by expanding gas at high pressure through a small nozzle and “skimmer” into vacuum. He was familiar with classic physics experiments, which had been done since the 1920s using molecular beams formed by effusion of gas at low pressure. Such beams provided a key advantage: the molecules travel independently, so properties of individual molecules could be determined by observing interactions of a beam with external fields, radiation, or other beams. But low intensity limited the scope of their applications. The supersonic free-jet beams offered much higher intensity and, by virtue of the high-pressure expansion, they produced much narrower velocity distributions and much lower temperatures than those in effusive beams. John was elated by the research opportunities opened up by supersonic beams; they “set the stage for the rest of my scientific life.”

Pursuing “Big Leaks”

Back at Princeton, John launched an adventurous program to exploit what he called “big leaks into vacuum.” Novel and productive, the program soon led to his appointment as professor of mechanical engineering from 1959 until 1963, and professor of aerospace sciences from 1963 until 1966.

The program was enlarged when he returned to Yale as professor of applied science and chemistry, a position he held from 1967 until 1980, when he switched to professor of chemical engineering, which lasted from 1981 until 1987. Throughout those three

decades at Yale, along with producing seminal research, John was a tireless evangelist for supersonic beams.

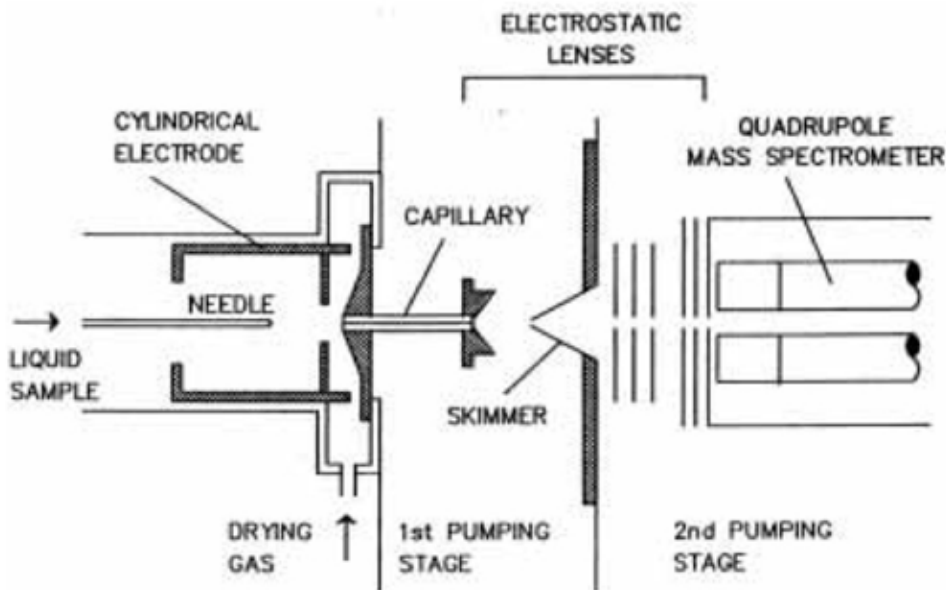
Early results at Princeton had elucidated the role of collision-induced processes in the supersonic expansion. Collisions in the high-pressure region organize the molecules of the jet to a remarkable extent. The emerging crowd of molecules thus may have mean separations of only fifty diameters, yet downstream acquire nearly the same velocity and direction and hence suffer almost no collisions. Consequently, the terminal state of the jet is far from equilibrium and the temperatures associated with relative translation and rotation of molecules within the jet are typically very low, in the range of one degree Kelvin or less. The vibrational temperature can be kept high or made low by choice of conditions governing the number of collisions in the nozzle. Free jet expansions greatly enhanced spectroscopy of polyatomic molecules from radio-frequency to optical regimes, because at low temperatures, chiefly just the lowest quantum states are populated.

Among studies done at Yale were a series on formation of molecular clusters, including clustering of water on hydrated protons. These exemplified how chemical reactions within the jet are in effect freed from the second law of thermodynamics. That happens because the entropy term in the free energy is suppressed by virtue of the extremely low relative translational temperature. Large mole fractions of molecular clusters can therefore be readily generated in supersonic jets, making accessible a vast realm of unexplored chemistry. For John, a favorite, triumphal example was generation of C_{60} in a supersonic jet of carbon clusters (although he was not involved in the discovery). Although C_{60} had long before been predicted to be a very stable molecule, efforts to synthesize it by conventional means had failed.



Fenn's "big leak" molecular-beam system, with two 32 inch diffusion pumps and one 16 inch "Jet Booster Pump," after it was moved from Princeton to Yale. Fenn, standing on the right, is accompanied by two postdoctoral fellows, S. B. Ryali, also standing, and S. P. Venkateshan, sitting.

(Photo courtesy The Nobel Foundation 2002.)



Schematic diagram of Fenn's second (and final) electrospray ionization apparatus built at Yale. Sample solution was sprayed from the hypodermic needle into a counter-current flow of dry nitrogen. The needle was at high potential relative to the cylindrical electrode and the metalized entrance end of a capillary. The ion-gas mixture enters the free jet expansion regime as it emerges from the exit of the capillary. Its center portion then passes through the skimmer into a second vacuum chamber containing a quadrupole mass analyzer.

(Photo courtesy The Nobel Foundation 2002.)

Another unique property of jet beams, thoroughly examined by John's lab, is acceleration by "seeding." The gas of interest is mixed with a large excess (typically one-hundred fold) of a light diluent gas such as helium or hydrogen. Collisions during the supersonic expansion thus bring the seeded molecules to about the same exit velocity as the carrier gas, and also concentrate the heavier species along the beam axis. Intense beams are thereby obtained with kinetic energy readily variable over a wide range extending well above typical activation energies for chemical reactions. The seeding technique and variants became pervasive in crossed beam experiments probing dynamics of molecular collisions.

Relished among John's many evangelical forays was a visit he made in the mid-1970s to Sandia National Laboratory. He had been invited to give a technical talk about how free

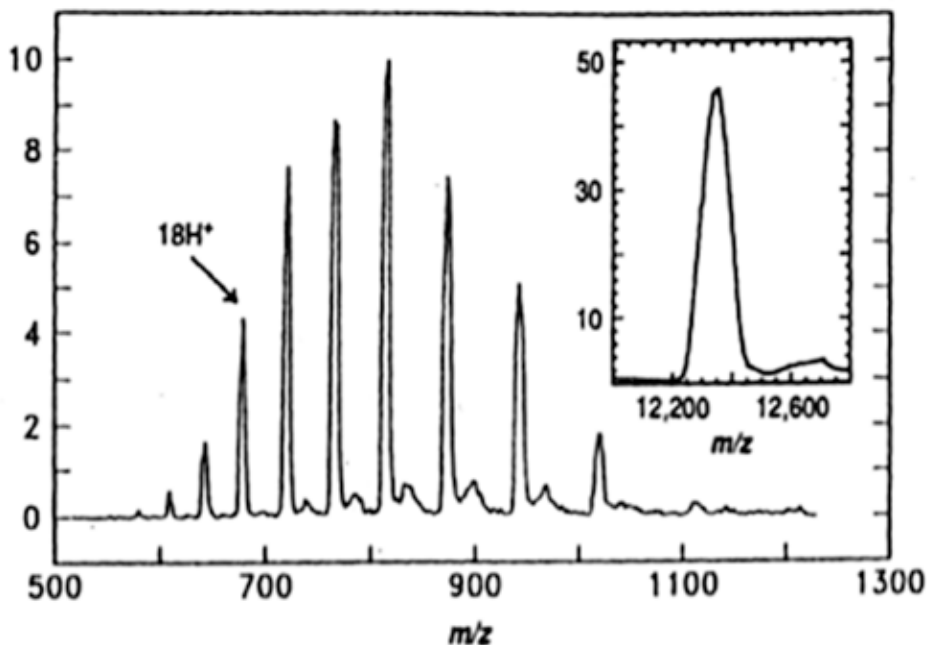
jets are used to study chemical reactions. Always keen to be unorthodox, he sent the title: “Information gained from Deliberate Leaks.” When advertised at the nearby Lawrence Livermore Laboratory (LLL), only the title was announced, not accompanied by the abstract. His host at Sandia was startled that John’s talk drew “a lot more people than we were expecting.” But then, he “noticed that many in the front two rows were from LLL’s intelligence and security areas.” They were baffled when John launched into his talk, deliberately laden with technical information attained from “big leaks.”

Electrospray ionization

The impetus for John’s pursuit of electrospray ionization came from a paper published in 1968 by Malcolm Dole, describing his attempt to measure the molecular weights of synthetic polymers by mass spectrometry. As applied for many decades, mass spectrometry involves ionizing samples, injecting the gas phase ions into a vacuum chamber, and determining the mass-to-charge ratio of the ions from their trajectories in electric and magnetic fields. The standard methods for ionizing neutral molecules employed knocking off electrons by collisions with high-energy electrons, photons, or charged molecules. But large polyatomic molecules, especially proteins and other complex biomolecules, could not be vaporized without extensive fragmentation and decomposition.

Dole’s approach was to introduce a dilute solution of the polymer in a volatile solvent through a hypodermic needle into a chamber through which nitrogen bath gas flowed at atmospheric pressure. A potential difference of several kilovolts between the needle and the chamber walls produced an intense electric field at the needle exit and dispersed the emerging liquid into a fine spray of charged droplets that are driven by the field toward the end wall of the chamber. A small fraction of the mixture of bath gas and ions passes through a nozzle in the end wall and skimmer, emerging as a supersonic free jet into a vacuum chamber housing a mass analyzer.

The essential feature that enables “soft” ionization of a large, nonvolatile polymer or biomolecule without fragmentation is evaporation of solvent from the charged droplets. Such evaporation requires a source of enthalpy, provided by gentle collisions with the thermal energy of ambient nitrogen bath molecules. The bath also serves to sweep away much of the emitted solvent vapor, along with other uncharged material, from the inlet orifice. As evaporation proceeds, the charge density on the droplet surface builds up to a critical value, termed the Rayleigh limit, at which Coulomb repulsion overcomes surface tension. The droplet then disintegrates into smaller charged droplets, each of which



Electrospray mass spectrum of Cytochrome C protein (nominal molecular weight 12,360 Da). The spectrum exhibits a sequence of peaks, each of which differ from its neighbor by one charge. The number of charges per ion is attributed to the number of adduct protons (H^+); it ranges from 12 to 19 for this spectrum. The insert shows the result of applying a deconvolution algorithm. It transforms the multiple peaks into a single peak that would be obtained if all the ions had a single charge. The m/z value at the apex of the single peak specifies the true molecular weight of the parent neutral molecule.

(Photo courtesy The Nobel Foundation 2002.)

continues to evaporate until the Rayleigh limit is again exceeded, resulting in further disintegration. This sequence repeats until finally the radius of curvature of the droplet is so small that the electric field at its surface is high enough to desorb a solute ion from the droplet liquid into the ambient gas that enters the inlet orifice to the mass spectrometer.

In the early 1970s, John's lab carried out variants of Dole's experiments, but they obtained discouraging results, in retrospect largely due to inadequate instrumentation for measuring ion currents and mass analysis. After a hiatus of nearly ten years, John decided

to revisit electrospray ionization (abbreviated ESI), not with macromolecules but with solute species small enough to be recorded using an available mass analyzer with an upper limit for mass at 400 daltons. (The mass unit of a dalton, or Da, is equivalent to 1 gram/mole or 1.66×10^{-17} kg). Results obtained for such species showed that, using solutions at concentrations of only a few parts per million, ESI provided a clean, single mass peak for each solute ion species with no evidence of fragmentation even for species that could not have been vaporized without extensive decomposition.

Borrowing a mass analyzer with range up to 1,500 Da led to further gratifying results. With excellent sensitivity and signal-to-noise, spectra were obtained for peptides having masses up to at least 1,200 Da. Moreover, some of the solute ions were doubly charged, an intriguing observation. It was provocative because the effective mass range of any analyzer increases by a factor equal to the number of charges per ion. John suspected that the number of charges on a solute ion formed by ESI might increase with the size of its parent molecule. That hunch was soon amply confirmed. By 1988, his group had applied ESI to solutions of protein molecules with molecular weights (M) in the range from 5,000 Da to nearly 40,000 Da. The mass spectrum for each protein comprised a sequence of peaks, each peak differing from its adjacent neighbors by one unit charge, corresponding to addition of a proton, H^+ . For example, the spectrum for bovine insulin ($M = 5,730$ Da) showed three peaks corresponding to parent solute molecules with four, five, and six added protons, whereas the spectrum for alcohol dehydrogenase ($M = 39,830$) had fifteen peaks corresponding to thirty-two to forty-six added protons. Thus, the resulting mass-to-charge ratios are within reach of a modest mass spectrometer with nominal upper limit of 1,500 Da.

John liked to recall, “Utter dismay was the initial response of every mass spectrometrists that saw our first spectra of proteins.” All were sure that the multiplicity of peaks for each species in a mixture would lower analytical sensitivity and make the spectra too congested to interpret. However, “after staring at these new spectra for a few days,” John realized that “each one of these peaks constitutes an independent measurement of the molecular weight of the parent molecule. There ought to be some way to average their information content to arrive at a more reliable and accurate value of that molecular weight.” He pointed this out to a new graduate student, Matthias Mann, who two days later presented John with a deconvolution algorithm. It enabled a desktop computer to transform the observed multiplexed spectrum into a single peak that would have been obtained if all the parent ions had been singly charged. Even the complex spectrum of



Photograph of Fenn's electro-spray ionization spectrometer apparatus, on display at the Chemical Heritage Foundation in Philadelphia.

(Photo courtesy the Chemical Heritage Foundation.)

a mixture of several proteins could be unraveled to produce a deconvoluted spectrum comprising a single peak for each protein species.

The results John obtained for proteins soon touched off an “electrospray revolution,” vastly extending applications of mass spectrometry, especially to biological and medical science. Within a decade or so, ESI had been shown capable of producing intact ions of macromolecules with molecular weights of more than 100 million Da. It was even found that viruses could undergo ESI without losing their viability. Thousands of research and analytical papers using ESI were appearing each year. Many variants became standard, such as direct input from liquid chromatography columns. The instrument built by John and his students while they were developing electro-spray ionization is now exhibited by the Chemical Heritage Foundation in Philadelphia.

In 1987, John reached age seventy, which was the mandatory age for retirement from the faculty at Yale. Fortunately, he was allowed to keep his lab long enough to achieve the results that touched off the electrospray revolution. Soon after, however, he was asked to give up most of his space. John responded by applying, in vain, for an open slot as an assistant professor. In 1992, Margaret died in an auto accident. Two years later, John moved to Richmond and married Freda Mullen, the widow of Jim Mullen who nearly fifty years before had recruited John to help launch Experiment, Incorporated. Virginia Commonwealth University provided ample lab space and welcomed him as Research Professor of Analytical Chemistry. In 2002, at age eighty-five, John received the Nobel Prize. A few months later, in April, 2003, he was elected to the National Academy of Sciences.

Multifaceted exuberance

In his many activities and roles, John gave full rein to his quick wit, intrepid questing, and zestful spirit. He particularly enjoyed and found fulfilling the teaching and mentoring of students. Thermodynamics was a favorite subject, although he had “barely passed” the only course he’d had in it as a student. After developing several unorthodox courses, John published a delightful book, chiefly directed at liberal arts students: *Engines, Energy, and Entropy: A Thermodynamics Primer* (1982, W. H. Freeman and Company). The impetus for writing it came from his association with Calhoun College, one of Yale’s residential colleges for undergraduates. When students requested that he offer a seminar course on repairing automobiles, John proposed instead to teach them about principles of heat engines. He designed his book “to appeal constantly to the reader’s own experience” and to enable students “to enjoy learning with their shoes off.” Along with lucid grounding in basic principles, the book is replete with whimsical cartoons and verses, a host of canny examples and refreshing historical excursions. Widely appreciated, it was translated into Russian and Chinese, and it is still in print with a 2003 edition released by Global View Publishing.

John’s approach was likewise exuberant in his personal, recreational pursuits. He was an avid reader of literature and the *New York Times*. Very fond of poetry, he often quoted from memory long passages from Shakespeare. He loved music; at scientific conferences, he sometimes led an evening song session. For years he was a daily jogger, taking it up well before it became vogue. He considered foreign travel as his “other addiction,” and his preparation for trips included listening to language tapes while driving to work.

As well as taking part in myriad conferences all over the world, John made some extended visits. First to London (1955), then as a visiting professor at the University of Trento, Italy (1974), the University of Tokyo (1977), and the Chinese Academy of Sciences (1987). Also, he was a visiting scientist at the North American Aviation Science Center (1965), the Indian Institute of Science in Bangalore (1979), and Latrobe University in Melbourne, Australia (1992). As the recipient of the Humboldt Senior Scientist Award, he visited The Max Planck Institute in Göttingen, Germany, during the springs of 1983 and 1984.

Always eager to probe problems and opportunities, John served as a consultant to several government agencies and a number of companies, including General Motors, Sandia Corporation, the Pfaudler Company, Sybron Corporation, Mobile Corporation, Thermal Research and Engineering Company, AeroChem Research Laboratories, and Relay Development Corporation. He also gave forthright advice to academic administrators, not always by request. At a celebratory “FennFest” dinner at Yale in 1983, President A. Bartlett Giamatti earnestly thanked John for “the galvanizing effect of his fierce loyalty and his great capacity for indignation,” as well as his “unconventional views of what chemical engineering is and what it might become in the future.”

Characteristically, John placed great emphasis on the contributions to his scientific work by his students and colleagues. The “FennFest” issue of the *Journal of Physical Chemistry* (1984) includes a roster of about a hundred associates described by him as those he had “worked with, learned from, and leaned on.” More are cited in his 2002 Nobel Prize lecture. Vastly larger is the host of friends, within and apart from science, who are grateful to have known him and been inspired by his exemplary love of life and learning.



Cartoon suggested by the title Fenn gave his Nobel Lecture: "Electrospray Wings for Molecular Elephants." Looking on quizzically is Charlie the Caveman, who appears often in cartoons Fenn included in his book on thermodynamics. Drawn by his daughter, Marianne Steinberg.

Ode to Electrospray Mass Spectrometry

Large molecules refuse to soar

Alone in the air they won't vie

Heated up, they buckle and break

Acting like an egg you fry

Spectrometers need gases charged

Before mass measurements they try

Big molecules drift far apart

When very wet instead of dry

Aha said Fenn, I will charge them

After they're dissolved awry

Droplets from his electrospray

Shot into vacuum quickly die

Done in by Coulomb explosion

As solvent boils the solutes sigh

Highly charged, alone at last

Mass identity they supply

Like Disney's Dumbo did before

Elephant ions learned to fly.

--C.E.K.

AUTHOR'S NOTE AND REFERENCES

This memoir is drawn chiefly from much fuller accounts published by John, his “Biograffiti” (1996) and “Wings for Molecular Elephants” (2003), listed below with four other articles that describe further aspects or episodes. Our liberal use of quotes is intended to convey the cheerful tone of his writing.

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