Charles B. Harris

1940–2020

BIOGRAPHICAL *Memoirs*

A Biographical Memoir by
Michael D. Fayer

NATIONAL ACADEMY OF SCIENCES

CHARLES BONNER **HARRIS**

April 4, 1940–March 10, 2020

Elected to the NAS, 2002

A pioneering physical chemist, Charles Bonner Harris spent an illustrious career investigating the detailed dynamics of chemical reactions and transient events. His efforts shed light onto the motions of molecules, atoms and electrons, resolving their actions into femtosecond timescales and picometer distances, and describing their behavior both in and on a variety of physical media. His scientific accomplishments had important ramifications for the advancement of materials science, chemical synthesis and the development of catalytic methods. When asked to identify his foremost accomplishment, Charles pointed to his students, who have gone on to populate the laboratories and offices of the nation's corporations and the faculties of its best universities, winning numerous awards along the way, including a Nobel Prize.

By Michael D. Fayer

For his contributions to the advancement of science, Charles was widely recognized. He was elected to the National Academy of Sciences of the United States in 2002 and the American Academy of Arts and Sciences in 1997. He received the Optical Society of America's Award in Applied Spectroscopy in 1967 and the Pittsburgh Spectroscopy Award in 1990. He was elected a Fellow of the Optical Society of America, a Fellow of the American Physical Society, a Fellow of the American Association for the Advancement of Science, and an Alfred P. Sloan Foundation Fellow. He spent the bulk of a more than 50-year career at the University of California at Berkeley, joining the faculty of the College of Chemistry there in 1967. Charles Bonner Harris was a gifted man. Some called him a man for all seasons, a Renaissance man, an appellation of which he would be proud. Certainly, he was enigmatic, a man of varied seasonings: brilliant but warmhearted, driven but patient, stubborn but sentimental, quiet but adventurous.

Early Life and Education

He was born on April 24, 1940, in New York City, a third-generation American. Around 1900, his grandfather, Yusef Hariz, emigrated from the small town of Zahale in the mountains of Lebanon, renamed by U.S. immigration officials as Joseph Harris. Before Charles' fourth birthday, his family moved first to Chicago and then to the affluent Detroit suburb of Grosse Pointe. His father, also Charles, was a "jack of all trades," able to run small newspapers as well as perform as a factory engineer. His mother, Brenda Bonner Johnson, grew up in Fort Worth, Texas, becoming the first female lawyer to work for the Texas state government. When the family moved to Grosse Pointe, she decided to forego the practice of law and instead became a third-grade teacher. Lebanese roots fostered a close-knit family of four, father Charles and mother Brenda, Charles, and his younger sister Sally.

By all accounts the youthful Charles excelled at everything he set his mind to do, displaying a breadth of curiosity and ability. He collected butterflies, studied piano, and joined the Boy Scouts. By age 13, he was an Eagle Scout. By age 14 he had mastered Beethoven sonatas and the Lebanese hand drum known as a derbake. Attending Grosse Pointe High School, he became both athlete and scholar. He played varsity football and joined the track team, becoming a state champion pole vaulter. Musical tastes broadened as he and his father often drove into Detroit to enjoy jazz music at the West End Hotel, where Charles occasionally joined in 2 a.m. jam sessions with well-known jazzman Yusef Lateef.

Like many young men of his age, high school adolescence also nurtured a penchant for getting in trouble, the kind of trouble fostered by an adventurous spirit. His father was once called to help him extricate the family automobile from the grip of a golf course sand trap. On another occasion, Charles had an unfortunate disagreement with a restaurant window and came away requiring numerous stitches. Then there was the story told by his childhood friend, David Roll, about a ski trip to northern Michigan, a nearly empty gas tank, and an attempt to fill it by siphoning what might be found in other vehicles. That story ended with Charles jumping from the car, trying desperately to smother his flaming jacket, and finally being saved by his friends' quick thinking and a roll in a snowbank.

Dealing with Charles' misadventures must have required more than a little patience and a measure of understanding from his parents. Some courage might also have been necessary, as when they allowed 16-year-old Charles, in what he later said was simply an attempt to avoid household chores, to move out of the family home and into a room in Detroit, insisting only that he continue high school and present himself each Sunday at family dinner. The difficulties of a commuter's life brought Charles' first attempt at independence to an end within but a few months.

Athletic pursuits, jazz jaunts, and misadventures still left enough time for Charles to complete high school with excellent grades and high SAT scores, and to earn himself a scholarship to Cornell University. A second shot at independence lay ahead.

College life seemed to be everything Charles had hoped for. A year of partying secured him a less-than-stellar grade point average—including an F in chemistry—and a swift termination of the scholarship supporting his newly independent life. At year's end Charles' father drove to Ithaca, talked to the dean, and then drove Charles back to Grosse Pointe. Conversation during their return trip must have been somewhat strained. Charles came home with two choices: the army or a job. Charles found a factory job.

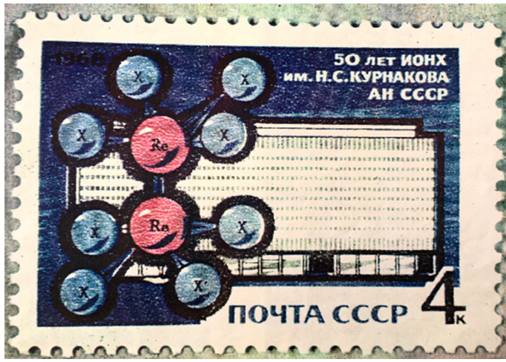
Armed with a failing grade in chemistry from Cornell University, Charles' first job gave him charge of the maintenance of chemical etching baths for a manufacturer of titanium components. This experience did not endow Charles with any great love for industrial chemistry, but it did result in a lay-off and the right to draw on unemployment insurance. Charles chose to use the time to return to school, attending summer classes at the University of Michigan and then enrolling full time at Henry Ford Community College. A year later, he managed to talk his way into the University of Michigan.

Despite preparing to restart his college career, Charles had not forgotten his love of partying, finding time to attend a few Detroit social events. At one party in particular, he met another graduate of Grosse Pointe High School, a young woman named Sara Ann Mikesell. Within the year, Charles and Sara were married and started life together at the university in Ann Arbor.

Still in need of a goal toward which to direct his talents, Charles studied philosophy and psychology and did well enough to earn another scholarship. Somewhere along the way he veered toward the pursuit of a premedical degree. Curriculum requirements dictated additional study in the physical sciences and ignited an interest in scientific research. At last aimed and armed with Sara's support, Charles graduated from the University of Michigan in 1963 with a degree in chemistry.

MIT and Berkeley

The decade of the 1950s saw the application of x-ray crystallography to the study of large molecules. In 1962, two Nobel Prizes were awarded for the crystallographic determination of large molecular structures. In this exciting atmosphere, Charles decided to attend the Massachusetts Institute of Technology (MIT) and study protein crystallography in the emerging new arena called biophysics. Once there, he became discouraged by the Biophysics Department's classroom requirements and began to cast a wider net in search of promising research problems. His efforts were rewarded when he found Professor F. Albert Cotton, an inorganic chemist who had also become interested in protein crystallography. However, before moving into proteins, Cotton noted the discovery by Russian scientists of a compound with a very short chemical bond and gave Charles the initial assignment of determining the crystal structure of a compound displaying this same type of bond. The task proved worthy of effort, resulting in an explanation for the short bond as the first proven example of a quadruple chemical bond. The Russians were so impressed that they put a picture of the compound on a postage stamp.



Russian stamp.

Protein crystallography faded into the background as this new discovery led Charles to explore more deeply the theoretical description of molecular electronic structures and the use of physical techniques, such as nuclear quadrupole resonance spectroscopy, for revealing them. More and more, Charles now wanted to understand the physics of chemical structures. Over the next three years, Charles and F. A. Cotton wrote and published a number of scientific articles in well-recognized technical journals. Later in life, Charles would tell the story that his Ph.D. thesis took him all of three days, since he had only to staple together the papers that he had already published.

While a professional life was taking form, Charles still managed to hold onto a rich and multi-faceted personal life. A Julliard graduate taught him the stand-up bass, which he occasionally played with a jazz trio, but MIT gatherings saw him principally sitting at the piano during regular Friday night performances. He also found time to become a pilot, learning to fly and earning an instrument rating. Sara's work helped pay for rented airplanes and on one occasion the two of them flew as far as the Bahamas. Much later, he often called the time at MIT some of the best years of his life.

An Atomic Energy Commission fellowship enabled Charles to stay on at MIT for a year longer, working in the Physics Department with John Slater and deepening his understanding of the theory describing the structure of electron atomic orbitals. Then in 1967, a very good offer in the form of an appointment as both an Assistant Professor in the College of Chemistry as well as a faculty scientist in what would become the Lawrence Berkeley National Laboratory (LBNL) brought him to the University of California, Berkeley (UCB).

UCB thought it was getting an inorganic chemist. In fact, the university acquired a physical chemist whose skills were developing rapidly and moving into spectroscopic arenas.

Initial years at Berkeley saw a flurry of activity. Building a new, independent research effort occupied his time but left enough irrepressible energy to spend hours wind surfing in the bay and riding his horse named Dan through the Berkeley hills. Jazz music continued to stir his imagination and prompted the frequenting of Sausalito jazz clubs. Home life also changed. A daughter, Heather, entered his life in 1968 and a second daughter, Sabrina, arrived in 1970.

When new spectroscopic methods showed up in a paper by J. H. van der Waals describing optically detected electron spin resonance, it sparked a fruitful collaboration between Charles and others at the University of California, Los Angeles (UCLA) and the University of California, Riverside. The collaboration sent Charles' research efforts in new directions, exploring the use of advanced spectroscopic techniques to uncover the structure and dynamics of molecular excited states. Soon thereafter, as laser methods added new power to optical spectroscopy, Charles began employing pulsed lasers to interrogate heretofore inaccessible transient phenomena.

These first few productive years at UCB set Charles' feet firmly onto a path toward success. His nascent student research group grew and flourished, producing volumes of admirable science and, in 1973, securing for Charles a tenured position as Associate Professor. Then, just as Charles professional life was beginning to settle into a steady pattern, his personal life took a turn. Charles and Sara divorced in 1974.

The early 1970s also saw Charles pursue collaborative work in the Netherlands. This opportunity became the first of many international ventures that attracted his attention. While working in the Netherlands, Charles found time to play the piano at an Amsterdam nightclub. There he caught the attention of another young woman, Ingrid Elmendorp. Ingrid and Charles returned to his home in Orinda and married in 1976. Their marriage lasted until the year 2000 and saw the birth of daughter Vanessa in 1977 and son Maarten in 1983.

Over a period of more than 40 years beginning in the early 1970s, Charles' laboratories became both citadels of scientific discovery and incubators of some of the best scientific minds in physical chemistry. Charles' mentoring would produce more than 70 Ph.D.s who would go on to populate the faculties of the world's best universities and the offices and laboratories of the world's top corporations. His students' success became a source of great pride. When asked of his greatest accomplishment, Charles would invariably identify his students.

His children's growth and accomplishments also fostered great pride. Heather married Aaron Dubois and became a bookkeeper and a creator of historical costumes. They live in Oakland, California. Sabrina studied fashion design. Her death to cancer in 2004 struck Charles more severely than any other event in his life. Maarten became a Doctor of Physical Therapy and practices in Vancouver, Washington. Vanessa became an M.D. and Ph.D. She conducts research on infectious diseases, working in Amsterdam, China, and Africa. She and her husband, Ole Bouman, have a son, Marius.

In 2002, with his election to the National Academy of Sciences, Charles succeeded in reaching a long-sought professional goal, a goal that had both enticed him and eluded him since his earliest days.

In 2003, Charles was asked to chair the Department of Chemistry and then to lead the College of Chemistry as Dean. He accepted these assignments with an uncharacteristic degree of humility and gratitude, honored by the university's trust and grateful for the opportunity to repay UCB for its support over so many years.

In 2011, Charles married his longtime childhood friend, Donna Day Westerman and gained a stepdaughter, Johanna, and four step-grandchildren, triplets Sam, Jack, and Catherine, and their sister, Anna. Donna is a professor emeritus at Orange Coast College and an exhibiting fine artist with studios in Oakland and Berkeley, where she is an ongoing Artist-in-Residence at the Kala Art Institute.

Even as he dealt with the loss of his daughter, the mid-2000s presented Charles with another heavy blow, a diagnosis of Parkinson's disease. As he had with so many new experiences in his life, he confronted the challenge with courage, twice enduring surgery to install Deep Brain Stimulation devices.

On the morning of March 10, 2020, in his home in Orinda, California, with his wife, sister, and children by his side, Charles succumbed to the complications of Parkinson's disease.

Scientific Career

Charles was trained as an inorganic chemist, receiving his Ph.D. under the mentorship of the renowned F. Albert Cotton. Charles began his career at Berkeley as an inorganic chemist with an interest in the physical properties of inorganic molecules. His desire to study the properties of inorganic molecules in their electronic excited states led him to employ sophisticated spectroscopic methods, particularly optically detected magnetic resonance (ODMR). However, his evolving interests rapidly led him to transition to the field of physical chemistry and chemical physics.

Charles and FA Cotton, ca. 1967.

The field of ultrafast chemistry became the major thrust of Charles' career. In the early 1970s, lasers that produced ultrashort pulses, initially picoseconds and then femtoseconds, were emerging from applied-physics labs, and a small group of pioneers began applying them to problems of chemical interest. Charles' first focus was on developing methods to create and measure vibrational coherence. Methods and insights developed in those early days continue to inform many areas of modern ultrafast science, including multidimensional infrared spectroscopy, femtosecond Raman spectroscopy, and vibrational coherence experiments.

As dye-laser technologies enabled picosecond time resolution, they were quickly adopted into Charles' lab and applied to increasingly complex molecular systems. At first, these were visible absorption studies of chemical reactions: diatomic dissociation, isomerization, and hydrogen-atom transfer. The goal of studying these initial systems was to test the new statistical mechanical theories that were being developed to describe the effect of solvents on chemical reactions. A key insight that emerged early on was that dissipation of vibrational energy released during chemical reactions played a critical role in the overall reaction dynamics, enabling bond-breaking and bond-formation steps.

When table-top femtosecond infrared methods became available, Charles immediately saw their potential to measure reaction dynamics in more detail and in more complex molecules. He began to move back to his inorganic roots and his early interest in organometallic photochemistry. Over the next two decades, Charles explicated the mechanisms of increasingly complex organometallic reactions. His contribution to understanding the bond-activation processes underlying catalytic reactions are particularly important.

Charles (right) with Alex Pines (center) and former post doc and Nobel prize winner, Ahmed Zewail (left).

Throughout his career, Charles believed that the best science relied on cutting-edge technology combined with advanced theory. He pursued these principles to the end of his career, collaborating with people developing new synchrotron x-ray methods, learning sophisticated computer-simulation techniques, and adopting new 2D-IR methods. The latter methods allowed him to finally achieve a career-long goal, the time-resolved visualization of the transition state in a chemical reaction.

The 1990s saw Charles broaden his research impact by taking leadership roles at LBNL, first as Director of Chemical Science and then as Deputy Director for LBNL. In collaboration with LBNL Director Charles Shank, Charles and Daniel Chemla helped establish ultrafast research technology and methods as bedrock tools for the Basic Energy Sciences program of the Department of Energy, a legacy that has grown in significance with time. The Harris research group activities paralleled these large-scale changes in experimental laser science with a commitment to femtosecond resolution measurements of molecular and chemical dynamics and defined the research conducted by the Harris group until his retirement. Charles' commitment to scientific leadership also proved to be a defining feature of his career, as he accepted the roles of Chair of the Department of Chemistry and Dean of the Department and College of Chemistry at Berkeley.

As established in the 1980s, research within the Harris group had two foci: chemical dynamics in solution and electronic excited state dynamics at surfaces and interfaces. The liquid side of his research transitioned from the dynamics of photodissociation,

Charles (right) with former student Paul Alivisatos, recently named President of the University of Chicago.

geminate recombination, and vibrational relaxation that had been the focus of research in the 1980s and early 1990s, to investigations of photocatalysis with organometallic complexes. This research used the photodissociation of CO from metal carbonyl complexes to create reactive metal centers in their electronic ground state capable of breaking notoriously robust CH bonds. This proved to be a critical research direction for the remainder of Charles' career. Specifically, this work demonstrated the mechanistic importance of ultrafast vibrational spectroscopy as a probe of photochemistry and also provided important insight into the role of the coordinative geometry and metal spin state of catalytic reaction mechanisms. In general, this activity fit within a larger theme of Charles' career, a commitment to establishing a conceptual framework for understanding chemical reactions in the condensed phase tested by experiment and systematized through the basic principles of statistical physics and quantum mechanics.

The research in solution-phase chemical dynamics happened in a densely populated and competitive research field that included many former members of the Harris group. For the investigation of ultrafast electron dynamics at surfaces and interfaces, Charles proved to be at the forefront of uniting the technology of surface science and ultrafast lasers to address the dynamics of electrons at the interface between metals and 2D molecular layers. This activity focused on time and angle resolution in two photon photoemission events. Charles brought a chemist's disposition to a field dominated by physicists, differentiating the focus of the research from others in the field. Much like the research in photocatalysis initiated in the 1990s, the investigation of metal-molecule interfaces with time-resolved photoemission persisted into the twenty-first century. Two areas of accomplishment rise above his other important activities. The Harris group demonstrated the sensitivity of the time-dependent energy and effective mass of image potential states, the interface analogue of a Rydberg state, to the conformational response of molecules adsorbed on a metal surface. The work also investigated the electronic structure and dynamics of interfaces between metals and organic semiconductors, information directly relevant to functional organic materials.

Research had a defining role for Charles and the group of scientists that he mentored, but his approach to research had as important an effect on his group as the specific research objectives they pursued. During the roughly twenty years Charles led large-scale efforts beyond his group, capturing his attention often proved challenging. After tracking him down, a discussion of a student's latest results often proved surprisingly brief if there was a failure to articulate a compelling motivation for the work. Such was the cost and benefit of the enormous research freedom he granted his people. Harris group members were empowered to pursue their own research directions but with the challenge of meeting Charles' exacting standards for significance and quality. His standards originated from his tremendous belief that he and the students he mentored would do research of significance by focusing on ideas he found timeless, deep, and interesting, with little concern for the fad of the day. As a result, many of Charles' former students and postdocs have been highly successful in pursuing new research directions, distinct from the research they conducted in the Harris group. Through their efforts, they keep Charles' spirit alive in the research they conduct and in the manner in which they conduct it.

During his long career, Charles mentored seventy-three Ph.D. students, eighteen post-doctoral students, and nine undergraduate students. He and his group published 246 peer-reviewed papers in top chemistry and physics journals. A selection of some of his most noteworthy articles is provided below.

Charles Harris group, 2012. From left to right: David Suich, Eric Muller, Ben Caplins, Alex Shearer, Son Nguyen, Adam Hill, Charles Harris, Justin Lomont, Molly Ryan, Matt Zoerb.

Charles at the piano, ca. 2015.

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