Autobiography of Stephen R. Leone

It begins with the parents, the upbringing, even the religious background. Those who know me recognize the strong work ethic, responsibility and diligence instilled by my background. I wouldn't trade that upbringing or those parents for anything else. It ends with a willingness to change—not always easy, as my wife will say about me, but a willingness to accept others for what they want to be, the ability to perceive new scientific problems, the will to dismantle whole project areas in favor of something new, the ideas to create experiments and to investigate new things — this is at least partly how I am.

What follows is the obligatory sentence of the autobiography, the one I was avoiding: I was born in Queens, New York City, May 19, 1948. Both sets of grandparents had come over looking for a new life in the U.S. from Italy—Ellis Island played a prominent role in their entry. They found a way for my dad Dominic and mother Annie to meet for their marriage plans. My dad worked for Sears Roebuck for a few dollars a week. My mother was a schoolteacher. The first things I remember as a child were in a house in Rochester, New Hampshire—the snow plow digging out the streets, my getting hit with a large rock when playing in the backyard, the sledding in the backyard down to the river, getting locked out of the house, a really huge dog that turned out to be a beagle. My mother took care of us kids while my dad traveled to Illinois frequently while working for a company called Clarostat (Dover, NH), which made high voltage power resistors and potentiometers. The old radios and televisions had plenty of these components inside them.

At my age of five my parents packed up everything and the kids and moved to Batavia, Illinois, where I had all my elementary and high school education. My mother started teaching me to read and do math well before I went to the classroom, which was excellent. As kids my older brothers, Ray and Ron, were always involved with erector sets, model airplanes, fixing bicycles, and even old cars. I followed totally along in their footsteps, but in some ways I skipped the old car stage and went a long way with laboratory toys later in life. As a child, I had boxes of small electric motors and other devices that I took apart and put back together to learn how they worked. Building models was an exacting science for me, playing chess was great, plaster of paris was fun chemistry, and smelling the house up with a chemistry set that could never be purchased today became my routine. I totally believe that working with these mechanical and electronic things set the whole tone for my future. Tweaking knobs and taking things apart was in my nature. In high school, I took mechanical drawing. It was great—the closest thing to engineering that my high school had to offer. Boy did it come in handy when I designed my first machines for the laboratory later in life. What do I remember the most about growing up in Illinois — my Dad taking me to White Sox games.

Although there were no advanced placement courses in my high school, the education in Batavia was really good. The school system was so small that some innovative teachers took their own initiative to give some of us special opportunities to study beyond the normal grade school and high school classroom material. Thus in some classes I was able to work on math at a higher speed and physics at the college level, as long as I finished the high school material and took the high school exams too. I won the science award at graduation in high school, and I had a great photo of me taken in front of the smelly chemical bottles in the chemistry storeroom.

I was able to attend Northwestern University and make a pretty serious showing for a small town student. Calculus took some time to learn during the first year at the university, because I never had it before, but chemistry went really well—I hardly missed any points on the exams in my first term. By the time I went to see Ed King, the professor in my first course of chemistry at Northwestern, he was beside himself wondering when I was going to come in—I had done so well he wanted me to skip the rest of freshman chemistry and go right into organic chemistry. I took Professor King's advice and never looked back about what my major should be or why. My brother Ron had become a Ph.D. chemist and served as a wonderful role model for me.

I was part of a great group of undergraduates at Northwestern (Bob Bambara, Professor at University of Rochester, Bruce Henkin Ph.D. Berkeley, Chuck Franz Ph.D. UCLA, John Boras, M.D.) who always wanted more out of education, and the professors at Northwestern gave it to us—additional courses in advanced quantum chemistry, laboratory electronics, the opportunity to take graduate classes. We each joined research groups as undergrads and became total laboratory rats. I wanted to work with Joe Lambert, who had inspired me in my organic chemistry class, but the powers that be felt that Joe had too many undergrads in his group already. I therefore went to Du Shriver and asked him if I could do a physical chemistry undergrad project with him. Du was incredibly flexible and suggested great ideas, and I joined his group. Lo and behold, the recent addition of a laser Raman spectrometer at Northwestern allowed me to do the first polarization-dependent single crystal Raman study in the group, which resulted in my first publication. The trick was growing the crystals in a drawer in the laboratory by slow evaporation of the solvent. Some of the crystals were beautiful transparent truncated pyramids, while others had some flaw and deteriorated instantly in the laser beam. This introduction to lasers was another aspect of not turning back—the impressive possibilities of lasers made me interested in that technology for the long haul. The choice of undergraduate research was also a major turning point for me; rather than becoming a synthetic chemist, I went the route of lasers, electronics, and physical chemistry. Du, by the way, asked me to give a group seminar almost a few weeks after joining the group, on the subject of the G matrix in vibrational spectroscopy. This was a topic that I knew nothing about, not even what it was, but I gathered up books and learned. Basil Swanson and Greg Kubas taught me many things in those days; it is hard to look back without realizing how influential they were in my career.

The Vietnam war was raging and the country instituted the draft in my senior year in college. All of us were being called up for physicals. My brother Ray had served in Vietnam, and fortunately he came back without significant physical or emotional scars. Du Shriver said to me that it was the best thing I ever did when I came back on the EL from Chicago with the papers indicating that I (surprisingly) failed the physical exam. It permitted me to go to grad school! I visited grad schools on my own ticket in those days. It was the first time I was really away from home and there I was jetting around the country to

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look at schools. I think Du was hoping I would go to Harvard, but Berkeley's emphasis on immediate research was what I really liked—so Berkeley was my choice for grad school—no matter the Free Speech Movement or anything else.

It was difficult to leave Northwestern because of the friends and the great professors and the opportunities there. And where else could one have a color TV in a small dormitory where you could watch the original Star Trek show, or the first manned lunar landing. Maybe the transition to leave Northwestern was eased somewhat, ironically, when our senior year was shortened by the Kent State University shootings, which brought the whole issue of the Vietnam war into sharp focus for all of us. Even normally peaceful Northwestern became a place of activism. I had plans to work at the Lawrence Livermore National Laboratory during the summer before grad school, and now there were many things to consider about the war. I even received a phone call one day telling me that my security clearance had been approved for Livermore, but they wanted to make sure I was still ok with going there, given the happenings at campuses throughout the country.

I was honestly too straight and geeky about science to say no to the opportunity to work at Livermore. After graduation I was on a plane to the Bay Area and shortly thereafter I was being treated to discussions led by Edward Teller and many other smart people at Livermore. I also met great friends like Steve Bernasek (later to become Professor at Princeton), along with a bunch of other talented students during that summer at Livermore. The range of big science and the vision of the people at that laboratory were an eye opening experience for me. But what really caught my attention was a seminar by a visitor who talked about the wide range of applications of lasers. It hit me like ton of bricks—with my experience in laser Raman at Northwestern, and hearing this visionary seminar about what one could do with lasers, how could I work on anything else but lasers when I started grad school at Berkeley in the fall!

At Berkeley I had thought a lot about working for George Pimentel, but his group was rapidly being filled by other students even before I went to talk to professors. Where was I that whole summer when Bob Coombe, Bruce Ault and others already joined Pimentel's group? Brad Moore had just received tenure in the chemistry department, and he was emphasizing the use of lasers to probe molecular energy transfer, predissociation, and reaction dynamics. His research offered an ideal opportunity: a chance to start with a nonlinear optical parametric oscillator to make a tunable laser source for vibrational energy transfer studies. Maybe it was fortuitous that George's group had already become somewhat full, because it made me look more carefully at Brad. Indeed, Brad's group offered, for me, an excellent combination of cutting edge laser technology and chemical dynamics. Where else could I get to play with a brand new Nd:YAG laser and all the nonlinear optics I could handle to study chemical dynamics. Jack Finzi and I were the first students to pioneer this novel laser system and to make it productive for chemical science.

I recall telling Brad that all things being equal, I would like to work on reactive dynamics, to see molecular bonds making and breaking, rather than to do only energy transfer, which had been the mainstay of the group up until then. It turns out Brad gave me the opportunity to do both. Brad was a wonderful mentor, providing guidance when really needed and permitting freedom when it was not. Brad and I performed an experiment together later during my grad school experience, to measure the long path absorption of CO molecules on high overtone bands. Through this experience I saw why it was that Brad was so successful. He was passionate to succeed and a fantastic experimentalist. I am extraordinarily grateful to Brad; he continues to be so influential in my career.

During my four years in Brad's group, there were lots of students and postdocs in both the group and at Berkeley who became high profile scientists a few years later; I cannot possibly try to recall everyone, but a few examples were Ed Yeung (Prof. at Iowa State), Paul Zittel (Aerospace Corp.—Paul, by the way, taught me most of what I know about doing things in the laboratory with rigor and curiosity), Paul Houston (Prof. at Cornell, now Dean at Georgia Tech), Klaas Bergmann (Prof. at Kaiserlautern), Frank Wodarczyk (NSF program officer and my best friend—it is impossible to thank Frank enough for his insights and advice to me over the years, and for his tremendous friendship), Glen Macdonald (Argonne National Laboratory), Steve Bernasek (already mentioned) in Somorjai's group, and next door we had people in the Pimentel group like Curt Wittig (Prof. at the University of Southern California) and Mario Molina, soon to be Nobel Laureate. At Berkeley there were also other good friends in the biophysical groups, people like Jerry Babcock (one of my first teaching assistants, Prof. at Michigan State) and Bob Blankenship (Prof. at Washington University), and theorists like Steve ONeil (Tech Transfer University of Arizona) and David Yarkony (Prof. Johns Hopkins)—a diverse group and no shortage of talent. Upstairs there was also Tom Cech, soon to be Nobel Laureate. Tom and Carol Cech joined the faculty at Colorado, while I was there, and Jerry Babcock and I served together as Associate Editors of *Annual Review of Physical Chemistry*, before his premature passing.

Grad school is a driven time, when everything about projects working out or not matters so crucially to the students, and the interactions can get pretty intense. We of the Brad Moore group were no exceptions. Jack Finzi and I would race each other to the D level at odd hours to try get more time on the laser. We used liquid hydrogen fearlessly to obtain a factor of 4 improvement in infrared detection sensitivity. Rumors were that young assistant professors would slip into people's laboratories late at night to take instruments for their experiments. I recall vividly when Paul Zittel chided me about whether I was going to announce to the world that my main accomplishment was to make a parametric oscillator work (since it was essentially a commercial instrument), or whether I was really going to use it to measure something new. I think Paul made me determined to tackle bigger and better things. Later I was quick to set up anything new and give it a try. The bar was set high at Berkeley.

My initial studies were on vibrational energy transfer in HCl, by exciting $v = 2$ and watching it share half of its energy with another HCl to form two $v = 1$ HCl molecules, and also studying vibrational energy transfer between isotopes of $H^{35}Cl$ and $H^{37}Cl$. When Brad left for sabbatical, Frank Wodarczyk and I established a whole new direction of electronic-to-vibrational energy transfer. It was Frank's idea, and I built up the apparatus and made it work. I also pushed on to isotopically selective predissociation studies of bromine molecules (the fashion at the time was to do laser-selective isotopic chemistry) by using an etalon to narrow the YAG laser, and Glen Macdonald and I studied vibrationally enhanced reactions and relaxation of HCl($v = 1, 2$) with Br atoms until late in the morning by talking through a tygon tube speaker system between our two laboratories. Those were the days when anything and everything seemed possible.

When I considered possible postdoc positions, Brad Moore suggested that I could look for academic jobs directly from grad school. Young and naïve at the time, I believed him. My proposal for faculty positions would probably not pass muster today in the highly groomed world of academic candidates going around looking for jobs, but in 1974 I got a bunch of interviews and ended up at USC as an assistant professor with about $10000 as my set up for the laboratory. Once again there was an interesting turning point, because I had hoped to get an offer from Oregon or Yale, and by my delaying, the offer from USC was first pulled off the table, and then it was offered to me again. I know Brad Moore had a hand in getting the USC offer back. The setup support was not enough to get a laser laboratory established, and so I started building lasers, exploring new kinds of lasers, trying to establish laser-initiated reaction dynamics and energy transfer studies, all on a shoestring. My ambition level was far greater than my resources. The poor amount of setup funds, in restrospect, was very helpful, because it forced me to learn how to get grants. My videotape, delivered as a lecture to young faculty at Colorado, on "how to get grants" was legendary among at least a few young faculty. While the level of grantsmanship has also increased dramatically over the years, nevertheless, I learned enough about how to get funds that I could set up a laser laboratory.

In my independent career, I have been so fortunate to work with many, many talented students, postdocs, undergraduates, visiting scientists, and visiting students. I cannot try to relate something important about each of them. Nevertheless, I will ultimately note many areas of research that we did and I will refer to the names of many people. I apologize to all those whom I do not include by name—whenever I review the photos of everyone who passed through the group (if you have not checked this out on our Web site, it is a great tour of the Leone group history), I recall important contributions from every one of you and treasure the interactions I had with you! Another thing the careful reader will note is that sometimes this autobiography appears chronological, but at other times it is written as if there is no such thing as chronological time. Speaking about something that happened far back in time, suddenly topics as recent as yesterday are introduced. Unsettling as this might be, it is how I often view the progression of our research. In addition, rather than trying to tell the world about particular scientific results, I plan to focus on some of the broad themes that made the research fun and successful for us all and the people who shared in it. In the end, what I value the most are the successes of the people who worked with me. I celebrate their careers and dedicate this autobiography to my students and postdocs. I am so glad that I had the chance to influence your directions and provide advice.

I started out my first Ph.D. student Alex Horwitz on isotopically selective vibrational energy transfer with a home-built discharge chemical laser, Ken Kosnik on an optically pumped sulfur dimer laser, Ara Apkarian (my highly accomplished undergraduate) on constructing a discharge nitrogen laser, and Martin Braithwaite, a postdoc, on chemical reactions, in the first years at USC. Curt Wittig (at that time a Professor in EE at USC) and I also collaborated on a set of electronic-to-vibrational energy transfer lasers with Alan Petersen, and Curt helped me immensely to gather some first grant funds with him. I learned many ways to be efficient from Curt, as I was sure he was poised to take over much of USC in the future. Howard Schlossberg of the Air Force Laboratory gave me the first real funding of my own, and the Navy and Army granted us funds soon too. NSF funding came rather late in the scheme of things, not for lack of trying, but running aground of doubting referees.

While I was at USC, it was a bit like being in Hollywood, a wonderful place to get discovered. Somehow I think that what follows would never have happened if I had not been located at USC. In my second year at USC, the folks at the Joint Institute for Laboratory Astrophysics (JILA) and the University of Colorado, Boulder, notably Carl Lineberger, with help from the outside by Dick Zare, who I sometimes think of as a guardian angel, pointed me toward the possibility of a job offering at JILA with the National Bureau of Standards (NBS) and the University of Colorado. I recall Dick Zare saying, if you ever hear from people at JILA, pay attention. One thing led to another and eventually I had a very nervous interview in Boulder, in which I was worried the whole time about USC finding out. I really felt I had no intention to pull out from USC, and I know I did not do well on the interview. Nevertheless, I shortly learned that I would get an offer to move to Boulder. It was like heaven to consider the setup package that was being offered and the resources of the shops and other facilities at JILA. Long story short, I made the decision to move to Boulder. Bob Lee came to USC to document the equipment that would transfer, and this gave me my first glimpse of the incredible efficiency with which JILA did everything. Alex and Martin and Ken came with us, we packed up Ara after the move to grad school at Northwestern, and that started a fantastic 26-year career with wonderfully supportive colleagues, outstanding facilities, and a great outdoor living environment.

My research was somewhat foreign to the people in JILA; I brought in chemicals and big glass evacuated bulbs, while the rest of JILA was vested in shiny stainless steel apparatuses and precision measurements. But somehow we hit it off. I was pulled to JILA's way of doing things, and I think they found value in my chemistry. I was turning bolts next to my students for many years, establishing some of my own projects from time to time and, when successful, deeding a project over to a student, all the while juggling the NBS missions along with academic work and teaching at the University of Colorado.

I brought to JILA a unique expertise to detect infrared vibrational emission from molecules, using a circular variable filter to obtain modest spectral resolution with background-limited infrared detectors. We were busily trying to study vibrational emission to map energy transfer, photofragmentation dynamics by detecting the atomic emissions and vibrations in small molecules, and reactive chemistry. Barney Ellison, who was a postdoc at JILA at the time, said one day in my first year at JILA, "Why can't we look at infrared vibrational emission from ion reactions?" Within a few weeks I was setting up an infrared detector and modulation system on the flowing afterglow of Prof. Chuck DePuy in Chemistry, and together with Barney, Veronica Bierbaum, and visiting fellow Jean Futrell, we observed the first vibrational emission from an ion−molecule reaction, $O^- + CO$ to produce CO_2 and an electron. This led to a whole new campaign, and eventually a major series of new apparatuses, to measure state-resolved vibrational dynamics of ion reactions, ion mobilities, doubly charged ion reactions, and single collision ion reactive dynamics, with ever-bigger and more elaborate set ups over 25 years.

Bill Pence was the first to join the group at Boulder, and Hubert Hofmann came next as a postdoc, talking about Boulder being similar to landing in Tenerife on the day he arrived. Hubert and Steve Baughcum (also a postdoc) began studies of photofragmentation dynamics, while Bill Pence pioneered electronic energy transfer in alkali atoms, which developed later into orbital alignment collision dynamics, and Tim Zwier, Jim Weisshaar (postdoc), and Mark Smith were the key people to pursue the first ion dynamics investigations—Zdenek Herman was an important Visiting Fellow who could almost draw our data to make it believable in the ion experiments. David Nesbitt (now Prof. and Fellow at JILA and Colorado) discovered laser-initiated chain reactions. For his exquisite experimental and theoretical thesis, David was awarded the Nobel Laureate Signature Award of the American Chemical Society (together with Prof. Casey Hynes and me). Hubert Hofmann was perhaps the most focused of all the young students and postdocs in the early group at Boulder, always asking if a photofragmentation project would lead to an important publication and then setting about it methodically and rigorously.

Paul Houston (Cornell University) and I became partners in organizing a Gordon Conference in those early years, linked up from Berkeley days and resulting in continued interactions throughout our careers. Bob Field (MIT) and I became great friends over the years, my having first met Bob when he was a grad student in Klemperer's group at Harvard and I was visiting grad schools. Eric Weitz (Northwestern) was another important colleague and friend; he was the Ph.D. advisor of Ara Apkarian. We young professors had all started our research careers in basic chemical dynamics, molecular energy transfer, and spectroscopy at about the same time. Later I also met and became very good friends with Fleming Crim (University of Wisconsin). Charlie Parmenter (University of Indiana), who already had many accomplishments to his credit when I was starting out at JILA, became incredibly influential in my early career when he was a Visiting Fellow at Boulder.

Veronica (Ronnie) Bierbaum became my most valued collaborator and friend. I think she was surprised when she came to my office one day and realized that I had acquired a large mechanical drawing board and had already started designing a flowing afterglow specifically for infrared vibrational emission studies of ion reactions. The years I spent interacting with Ronnie Bierbaum are filled with good science and fantastic memories. She is extraordinarily influential in the way I do science. I have a great drawing from this time, "The beards and hairs of the Steve Leone group" by Zdenek Herman, which recorded some of the students and postdocs mentioned above, along with a few others in the early Leone group (Visiting Fellow Terry Cool, David Nesbitt, Bill Pence, Steve Baughcum, Jim Weisshaar, Tim Zwier, Heinz Herman, Dean Guyer, Frank Magnotta, Brooke Koffend, Veronica Bierbaum, and yours truly), on Zdenek's visit to JILA at this seminal time in my career. Many seemingly important discussions about life and science and good friendships occurred with people like Stew Novick and Pat Jones, who were in the Lineberger group.

Things move so fast, during those initial years of a scientific career, that many aspects blur together. The support of JILA and the NBS often permitted me a greater flexibility to pursue science. It seemed a time to be fearless in trying new things. The basic photofragmentation work and emission from free radicals merged into translational-to-vibrational energy transfer investigations and eventually time-resolved Fourier transform emission studies, the early investigations of electronic energy transfer (Bill Pence, Mike Hale) resulted in sophisticated orbital alignment research, nanosecond pulsed laser reaction chemistry became femtosecond wave packet dynamics, and so forth.

Steve Bernasek came to visit for a few weeks in my early days at JILA and we pioneered an idea to detect vibrational infrared emission from surface reactions, the catalytic oxidation of CO on platinum to form vibrationally excited CO_2. The experiment worked in an amazingly crude flow tube, and in the back of my mind I hoped that someday I would return to this interesting new direction. A few years later, Karen Carleton joined my group with a request that she work on something related to surface studies. Having almost no experience in this field at all, Karen and I undertook to build an ultrahigh vacuum apparatus for surface dynamics. Naturally, none of the initial experiments we thought about doing worked out that well, and the referees on the proposals were difficult, but our work eventually took off in a totally new direction of using lasers to probe the spin−orbit states of gallium atoms desorbed from silicon surfaces, and these initial successful studies led to a long and fruitful set of research projects on laser probing of etching, laser ablation to form kinetic-energy-enhanced beams for surface interactions, and laser probing of molecular beam epitaxial growth. Eventually these studies morphed into microscopies and nanomaterials studies, such as near field and apertureless near field microscopy to probe photolithographic materials chemistry.

Great successes came out of the early work on translation-to-vibration energy transfer (Frank Magnotta, Chuck Wight), photofragmentation dynamics (Heinz Herman, Steve Baughcum, Jamie Donaldson), and quantum yield studies using infrared fluorescence, as well as some first transient laser gain versus absorption measurements (John Smedley, Harold Haugen, Wayne Hess). Harold Haugen had little previous laser experience, but he became one of the best laser experts in the group; he also built incredible, specialized electronics boxes for all the experiments. The orbital alignment collisional energy transfer studies (several people said they would never work), which were pursued over the years by Wolfgang Bussert, Laurie Kovalenko, Ruth Robinson, Chris Smith, Harold Parks, Henrik Lorensen, Eileen Spain, and most importantly Jan Driessen, often with Ingolf Hertel contributing as a visiting professor, provided elegant pictures and an in-depth understanding of the little understood phenomena of orbital alignment effects on collisional energy transfer. We use crossed beams and the alkaline earth atom, Ca, for these vituoso studies. George Schatz, John Delos, and Larry Eno, among many Visiting Fellows, and Regina de Vivie Riedle (my one theoretical postdoc) helped us tremendously with the theory of orbital alignment effects and often mentored my own students in theoretical work. At the end of those investigations several years later, we had explored elaborate three and four vector correlations in collision dynamics, even making it difficult for theory to keep up. I also had a passion to study ion reactions under single collision conditions, and laser-induced fluorescence was introduced early on to investigate ion reactions under a variety of conditions to obtain more sensitivity (Mike Duncan, Chuck Hamilton, Dean Guyer, Guang-Hai Lin).

Building and modifying machines was a wonderful adventure with the JILA shop at hand. The machinists in those early days (Bill Lees, Dave Henry, Jon Andru, Hans Rohner) would take our most impossible ideas and turn them into gold mines of scientific output. Saul Lissauer and his electronics crew would design us new detector and data acquisition boards and repair our ailing instruments with great speed. The technology was very important to us. JILA folks respected technology, and it was encouraged. With a technological edge or new apparatus for a measurement, more and better results always came out.

Carl Lineberger was my ultimate mentor throughout my career. Even though it is always difficult to get a really clear answer from Carl, his advice, friendship, and help were invaluable. I look up to him and respect him because he is an incredible scientist and someone who really cares for and looks out for others. I was speechless at a very early age to receive the Pure Chemistry Award from the American Chemical Society, with Carl and Bill Reinhardt undoubtedly masterminding the nomination. Others in JILA and Boulder also had a great influence on my research directions: Alan Gallagher, Jan Hall, Art Phelps, Gordon Dunn, Keith Burnett, Eldon Ferguson, Will Castleman, Stan Cristol, Tad Koch, Dave Walba, Chris Shiner; also Barney Ellison and David Nesbitt were exceptionally influential in my early career; both later joined the faculty in Boulder and the roster in JILA.

Midcareer in some ways meant more of the same, but it also meant more responsibility, less time for my own research, finding myself no longer being a colleague working side-by-side with the students and postdocs, and being called "the boss" by students. Nevertheless, there were notable shifts in our scientific goals and approaches: rotational alignment and state distributions were measured for ions drifting in a rare gas, and laser probing of the moments of ion velocity distributions of drifting ions was an important new direction of study, as well as product state measurements of ion reactions (Andy Langford, Rainer Dressler, Hans Beijers, Henning Meyer, Steve Penn, Christian Lauenstein, Mike Bastian, Eric Anthony, Jian Li, John Husband); a selected ion flow tube led to intriguing cluster ion reactive studies (Karen Knutsen, Shuji Kato, Mike Frost, Joost de Gouw, "MK" Krishnamurthy, and Visiting Fellow C. D. Lin); the time-resolved FTIR technique opened up huge paths of higher resolution studies, such as photofragmentation and reactive events, five member ring intermediates, radical−radical reactions, and reactions that occur by avoiding the minimum energy path (Rick Fletcher, Eric Woodbridge, Paul Seakins, Chris Lovejoy, Jim Lundberg, Jody Klaassen, Jörg Lindner, Rich Loomis, Jonathan Reid, Tim Marcy, Viktor Chikan)—we benefited tremendously from the theoretical and experimental collaborations on these basic dynamics studies, especially with David Clary, Ian Smith, Dwayne Heard, George Flynn, Eric Weitz, Steve Klippenstein, and Larry Harding); surface epitaxy and etching studies were occupying much of our time and presented great opportunities for discovery (Bernard Bourguignon, Russ Smilgys, Doeke Oostra, Lisa Cousins, Rob Levis, Gabriela Weaver, Francis Campos, Paul Strupp, April Alstrin, Brenda Korte, Adina Ott, Sean Casey, Nick Materer, Matt Knowles, Rory Goodman, Cindy Berrie).

During this time, 1987 to be exact, Mary Gilles came to graduate school and joined Carl Lineberger's group, and we became a couple around my birthday in 1988 and married in 1993 (you could almost argue that Carl was not only my best friend and mentor, but he gave me my wife too). Nothing changed my life more than to be with Mary Gilles. She transformed me for the better, making me see the balance between work, family, and play. We are a team, helping each other when each is in need, and finding great opportunity to explore the world together. Even the simple things, like looking for whales or hiking, take on new meaning when I am with Mary. Mary taught me about the constant surprises of interacting with a large family of interesting brothers and sisters, and we had great experiences with many of her nieces and nephews, especially Dowdy Gilles, who helps to keep us young. It is also with great joy that Mary and I have had scientific collaborations and publications together.

A broad period of expansion of my research efforts led to further investigations with transient laser absorption versus gain probing that provided ever-exacting details about state-resolved collisions in energy transfer and photofragmentation events (Craig Taatjes, Joe Cline); single collision studies went beyond ions to reactions of metastable excited states in Penning ionization (Dave Sonnenfroh); single rovibronic states of bromine electronically excited molecules were probed in exquisite detail (Katsuyoshi Yamasaki); The doubly charged ion collaboration with Carl Lineberger and Steve ONeil came into full swing, discovering bond forming reactions of doubly charged ion reactions (Paul Miller, Michele Manning, Steve Price, Steve Rogers, Yin-Yu Lee); we continued to dabble with constructing new types of molecular lasers, often in collaboration with the Air Force Laboratory in Albuquerque (Harold Miller, Robert Pastel); the low temperature chemistry of Titan became a focus with important relevance to the Cassini mission (Brian Opansky, Jens Pedersen, Ray Hoobler, Seongkyung Lee, Andrei Vakhtin, Boris Nizamov, Fabien Goulay, Adam Trevitt). Throughout all of this time, the loyal funding support of the National Institute of Standards and Technology (NIST), NSF, DOE, AFOSR, the Air Force Laboratories, NASA, and ARO made it a joy to work in science.

Yet, something seemed missing from this productive effort. Others in the world were pressing to shorter timescales and coherent dynamics, and our group seemed to be standing still with proven techniques. I determined I had to get a femtosecond laser system into the group and find out what it could do for us. I felt that if we were a group that knew and understood kinetics at a high level, we also should be a group that pushed the boundaries of time dynamics. On quite a few occasions with the assembly of our earlier laboratories, I had invested a great deal of my own time to design and purchase equipment, and having brought the project to a state where things were initially working, I happily turned the experiment over to my students. My approach was to let the students manage their projects and the science themselves, and I took great delight in their successes. On more than a few occasions, I defined a nearly impossible project, yet the students and postdocs made it happen. It was the greatest reward for me to see the students succeed in this manner. Getting back to the first femtosecond laser in our laboratory, there was almost no great idea of what to do with one, but rather a few general directions. My hope was to get a Ti:sapphire master oscillator and look at the pulses myself and find out what new experiments were possible. It might have been impossible to set up such an experiment without the supportive environment of JILA and the National Institute of Standards and Technology. It renewed my interest in laboratory experiments to learn the possibilities with femtosecond lasers, which was great fun.

Shortly after we obtained a femtosecond laser oscillator, a combination of Visiting Fellows, Linda Young, Wendell Hill, and Steve Sibener produced the first semi-ultrafast result in our laboratory, a precision lifetime measurement of a fundamental Cs atom excited-state by single photon counting. With about 1 ns time resolution, this hardly qualified as a femtosecond experiment, but it got us started in a good direction. John Papanikolas and Richard Williams, along with Visiting Fellow Paul Kleiber, made the breakthrough that I was looking for. It was Paul Kleiber's idea to study lithium dimer in a high-lying avoided crossing state. The devil is in the details, but John and Richard were heroic in making a cw dye laser work to first excite a selective launch state from which they could prepare coherent wave packets in the lithium dimer with the ultrafast laser. The observation of simultaneous rotational and vibrational wave packets with state-selected detail opened up a whole new area for our research in the field of coherent dynamics. A series of successful studies, involving John and Richard, Radek Uberna, Munira Khalil as an undergraduate from Colgate, Zohar Amitay, Josh Ballard, Hans Stauffer, Elizabeth Mirowski, Xingcan Dai, and Eliza-Beth Lerch, pioneered a wide range of aspects of coherent wave packet dynamics, quantum information processing, predissociation, and the selective preparation and removal of individual members of coherent superpositions. Mark Ratner, Tamar Seideman, Bob Lucchese, Jeff Cina, Jiri Vala, and Ronnie Kosloff contributed in substantial ways theoretically as well.

Around this same time of the mid 1990s, I went to a Gordon Conference on Nonlinear Optics, and there was a long section on making high order harmonics. Many physics groups were making high harmonics in the 100 eV range by focusing a strong Ti:sapphire femtosecond laser into a rare gas, but groups were not doing anything with the high harmonics to study a molecular system or chemistry. I determined that we had to try to put together such a source. With the great help of the NSF group grant in JILA and NIST resources, but especially through the wisdom of AFOSR and program manager Mike Berman, our group got the chance to build up an apparatus to study time-resolved photoelectron spectroscopy in the soft X-ray regime. My idea was that atomic core levels convey a huge amount of site-selective chemical information in molecules through their chemical shifts, and if one could observe these shifts in real time, a new kind of transient chemical dynamics probing would be achieved. Lora Nugent-Glandorf, Jennifer Odom, and Mike Scheer were the bold and innovative people who took on this challenge and Lora first succeeded by measuring the time-resolved photoelectron spectra in the valence region of the directly dissociative state of the bromine molecule. These initial results expanded several-fold in my laboratory to include investigations of related valence band photoelectron spectra, superexcited states, and ionic liquids (Daniel Strasser, Christine Koh, Astrid Müller, Jürgen Plenge). A wonderful new apparatus was constructed to measure two-color and time-resolved photoelectron angular distributions (Louis Haber, Ben Doughty). Finally, the idea that I dreamed of, femtosecond time-resolved measurements of core levels, was successful with a new X-ray high harmonic transient absorption experiment of Zhi-Heng Loh—it was totally Zhi-Heng's idea and his innovative construction of this apparatus that produced the first femtosecond core level time-resolved investigations in the group.

I saw that our surface science efforts were primarily emphasizing ultrahigh vacuum epitaxy and etching, and yet the need for these kinds of investigations was waning with the excitement of colloidal nanoparticle research. At the same time, NIST was interested in nanotechnology, and near field microscopy had recently become practical. This gave me the impetus to establish over the years, with Yin-Yu Lee, Wolfgang Schade, Bogdan Dragnea, Laurie McDonough, Jodi Szarko, Jan Preusser, Ligian Muntean, and David Osborn, a new direction of near field optical probing, for example, using an infrared near field microscope for the first time to study the infrared bands in exposed and unexposed photolithographic films (collaboration with Bill Hinsberg and Frances Houle at IBM); we further explored the use of near field microscopy to probe defects in semiconductors by scanned probe photoconductivity. This work transitioned into apertureless near field microscopy studies of semiconductor nanostructures (Zee Hwan Kim, Larissa Stebounova, Yohannes Abate), broadband coherent anti-Stokes microscopy (Sang-Hyun Lim, Allison Caster, Olivier Nicolet, Adam Schwartzberg), and ultrafast pump−probe investigations of nanowire lasers (Jodi Szarko, Jae Kyu Song, Andy Caughey). The one remaining molecular beam epitaxy apparatus in our group is used to study novel doped nitride materials and provides samples for microscopy studies (Bing Liu, Takeshi Kitajima, Dongxue Chen, Yaroslav Romanyuk, Gabriel Dengel, Lukas Kranz, Daniel Kreier).

Without a doubt, the thrill of my life came with the call one day early in the morning from several friends at Boulder about my election to the National Academy of Sciences. In another coordinate, I was inevitably called into service as JILA Chair and as the Acting Division Chief of the Quantum Physics Division in NIST. Both were fun positions to fulfill and it gave me a great opportunity to read about business management and the methods of seeking change in organizations. Steve ONeil advised me on the books to read, because he was studying for an MBA. I learned that an idea can be great, but if the people remain unconvinced, the time is not right, or the culture will not change, it just does not go forward. In these administrative roles it was wonderful to have a chance to work with several exceptional JILA staff, such as Pat McInerny, Cheryl Glenn, Vickie Paulson, and Ed Holliness. I came to appreciate the tremendous strengths of these individuals, and it gave me great lessons in working with others. I have to also acknowledge the outstanding support of Lynn Hogan, Loree Kaleth, and Joanne Carney, who were my JILA group assistants over the years, with whose support and help the group prospered, and Maryanne Rasmussen (who was instrumental in pointing out my future wife, Mary Gilles, to me) and Lynn Sarkis in Chemistry, who I worked with, respectively, on graduate recruiting and program reviews. There was great satisfaction to help with the operation of the Institute, but the aspect of coming in on Monday morning only to drop everything to put out a fire was not as fulfilling as research. I developed a wide interest in business by serving in these management capacities, and it taught me a lot. I am often still inept in knowing how to properly advise and work with people in the group, but the experience in management definitely helps. It also showed me the political side of things well enough that I realized I would not want to emphasize administration throughout my career.

Mary Gilles and I had just finished remodeling our house through Charlie Sarkis in Boulder in 2001—we were certain that Boulder was the place we were going to stay for the rest of our lives—when I got a call from Dan Neumark at my office one Saturday asking me about my possible interest to come to the faculty at Berkeley. The Chemical Dynamics Beamline at Lawrence Berkeley National Laboratory (LBNL) needed a makeover, and some of the faculty had discussed the idea of getting someone who could both direct the beamline at LBNL and establish a strong chemical dynamics program as a faculty member in the chemistry department on campus. The number of steps in the process and the array of considerations were enormous, the negotiations and deliberations took a year, and it was a really difficult decision for Mary and me to make. Dan was heroic in pursuing the details of the offer and solving problems and setbacks at every turn. In the end we decided to give it a try and moved to Berkeley. I was able to retire from NIST and, in a sense, take up a whole new career, as professor of chemistry and physics, and Director of the Chemical Dynamics Beamline, and Mary was also offered an intriguing opportunity to become a staff scientist at LBNL. The hardest part was leaving the great colleagues in Boulder, especially Carl Lineberger. There were also aspects of leaving the minimal bureaucracy and excellent running operation of JILA for the higher cost of living and greater congestion and time required to navigate daily life in California.

Nevertheless, four 53-foot electronics vans pulled up to JILA, over a series of four days in late June 2002, and the entire laboratory was packed and shipped by the students and postdocs in the group, with the help of Herminio Peinado, who became our official packing helper, but more importantly Mary Gilles orchestrated the whole move for me. I can never repay her for the extraordinary work she did to make the move a success. There was one more van on the last day to take our household, and then the house that we loved and the laboratory operation at Boulder looked so lonely and forlorn. A move of this size affords the opportunity to throw away lots of old stuff. We went through the laboratories for months to decide what to take and what to excess. More importantly, such a move offers the chance to establish new directions—to make a fresh start in important new areas. I made a conscious effort to increase the emphasis in the microscopy areas and to build up a larger presence in ultrafast X-rays upon arrival at Berkeley.

I invested enormous time to get all the grants transferred successfully and to write new proposals. It might seem like a reasonable task, given that all the granting agencies tell you that it is easy, but the number of false starts and missteps that occurred while following the instructions faithfully was unexpected. Just the close out of all the grants and balancing the books was a never-ending task for financial wizards Barb Tennis and Pat McInerny. And still to this day I receive a call or email about some item of equipment from NIST or the University of Colorado that is lost in bookkeeping limbo and we have to deal with it.

Many, many hours of my time were spent asking the big questions about what projects were important to continue, discontinue, or to build anew. This led to an invigoration that is very difficult to describe to someone who stays in a static location with a set of operating laboratories. I have to characterize myself as a person who happily ends projects in favor of new ones. I cannot count the times in Boulder, and already in Berkeley, that I have seized the opportunity to end a whole line of work in a laboratory and build the laboratory up again from scratch to achieve a new goal. Yet the move led to a scale of transformation that even I had not appreciated, readily stretching the limits of what I could think about and do.

Moreover, I was about to become the Director of the Chemical Dynamics Beamline at the Advanced Light Source, and, without any prior experience in synchrotron experiments, I had to try to decide what new directions made the most sense and how to involve new outside users. To my great fortune, LBNL staff member Musa Ahmed was already at the beamline and exceptional in the day-to-day activities, and Musa is a real idea generator and a fantastic colleague. He and I saw eye to eye about what had to be done (smaller, mobile machines, designed for flexible use) and it made it much easier to set the broad new themes with Musa's enthusiasm behind me. Thus what could have been a difficult challenge to manage the beamline and establish new directions turned out to be a great scientific reward, a bright spot of moving to Berkeley.

Twelve brave students and postdocs (about half the group in Boulder) came with us to Berkeley (Astrid Müller, Jodi Szarko, Hans Stauffer, Josh Ballard, Laurie McDonough, Stefan Gilb, Bing Liu, Alan Arrowsmith, John Husband, Chuck Blackledge, Tianhao Zhang, Vil Nestorov, together with two new Berkeley students, James Clark and Xingcan Dai, and admin Cheryn Gliebe, who came to us from Bill Miller's generous offer of help [Cheryn is the only person who could have navigated the impenetrable bureaucracy at Berkeley to bring in our large group]). We acquired very soon several new postdocs, Boris Nizamov, Viktor Chikan, Vas Stavros, Zee Hwan Kim, and Sang Hyun Lim. Without their help and hard work it would have been extraordinarily difficult to set up the laboratories again. Extensive equipment lists were made, parts were numbered and coded, and digital photographs were taken, so even if a person was not coming with us for a particular experiment, the system could still be set up by others; the ideas for this superb documentation and all the work came from Mary Gilles. During the move, Mary did so many things, from troubleshooting the supply of bubble wrap and boxes, bringing food and drinks to the students packing, while at the same time helping to design efficient laboratory spaces and offices—she was incredible.

Arriving in Berkeley, the group was excited, but soon we were faced with the insurmountable array of boxes to unpack and equipment to set up again. First Josh Ballard and Hans Stauffer unpacked their equipment and put it out on the table, and soon others in the group got the urge to do the same, and we started to manage teams of people to set up each project in turn so that the job became easier. Both LBNL (Bill Wu) and the College of Chemistry (Alex Shtromberg) did a fantastic job of keeping the laboratory renovations on schedule, getting the laboratories ready in time for our arrival. It was challenging to get used to the large number of new people who joined the group around that time; there is a culture in a research group that takes a lot of time to establish, or reestablish, even though seemingly the publication of results rebounded rather quickly.

I had a dream to initiate aerosol studies at the Chemical Dynamics Beamline and to provide this new type of experimentation at the synchrotron to other groups. Jinian Shu built the first aerosol mass spectrometer to be coupled to a synchrotron light source, and he pioneered all the initial light scattering studies; soon after, a newly appointed LBNL staff member Kevin Wilson, together with Musa Ahmed, Christophe Nicolas, Eric Gloaguen, Jared Smith, and Erin Mysak, and also along with Profs. Thomas Baer and Eckart Rühl, were succeeding in novel studies of aerosol chemistry, photoelectron ejection, and light scattering. Musa Ahmed initiated biomolecule studies by photoelectron spectroscopy, and this has grown into a challenging, new project to perform imaging microscopy with Lynelle Takahashi, Jia Zhao, and Leonid Belau, based on secondary ion mass spectrometry desorption of species, but with the detection of neutrals by synchrotron photoionization. David Osborn, together with Craig Taatjes, both of Sandia Laboratories, came up with a great idea for an ultimate kinetics machine, using tunable single photon ionization to probe product isomers of reactions, and we succeeded to build it and establish another important new direction.

In the recent few years, there have been a number of highly successful and new ventures. Vas Stavros carried out coherent dynamics experiments using four-wave mixing for the first time in our group, and Stefan Gilb initiated a brilliant photoelectron angular imaging of Rydberg state wave packet dynamics (cover art). Xingcan Dai, Elva Torres, Eliza-Beth Lerch, and Ken Knappenberger pioneered new aspects of wave packet coherent control, and Ken obtained simultaneous electronic/vibrational wave packets in polyatomic molecules. Bing Liu, Dongxue Chen, and Yaroslav Romanyuk, along with visiting students from Würzburg, grew nitride species by molecular beam epitaxy and studied island and nanodot formation. Ken, together with Amy Cordones, also established another new area for our group, measurements of fluorescence intermittency and memory effects as a function of tunable wavelength excitation in semiconductor quantum dots. We also started collaborating on experiments with Mary Gilles, utilizing her expert new skills on X-ray microscopy, to study polymer photoresists (Ligia Muntean, Alexei Tivanski, and staff scientist Deirdre Olynick). Sang-Hyun Lin pioneered an exquisite broadband coherent anti-Stokes Raman microscopy, and Zee Hwan Kim developed a novel apertureless near field microscope. As already noted above, upon coming to Berkeley it was possible to expand the emphasis on ultrafast X-rays, especially with the ties to the synchrotron light source and the Center for X-Ray Optics. Our group built several new laboratories using high order harmonic generation, for the first time in transient absorption and with photoelecron angular imaging. Dan Neumark and I and several collaborators later established an Ultrafast X-Ray Science Laboratory, and we helped hire new LBNL staff members, especially, for us, Oliver Gessner.

Shortly after my arrival at Berkeley, Chuck Shank, the Director of LBNL, asked me to join forces with the accelerator group, in particular John Corlett, to establish a possible new facility proposal to DOE for an ultrafast X-ray light source. Together, John and I, with the help of John's expert team, made superb progress on the design of such an accelerator-based X-ray facility. It was called LUX and it had the potential for far-reaching new experiments, including attosecond dynamics, as pioneered theoretically by Sasha Zholents. It was a lot of hard work, scientific meetings, and deadlines, but I learned so much from the interaction that it forever changed my perspective about teams and the potential of a large-scale facility. It was also great fun. Unfortunately, the LUX project had to end, due to political considerations, but it gave me ideas for new directions in attosecond science that we might do on a laboratory scale.

Thus, our work on ultrafast soft X-ray studies with high harmonic generation led to a new laboratory and an especially productive close collaboration with Dan Neumark to produce attosecond pulses. With generous support from an AFOSR MURI grant (Mike Berman), and help from DOE and the Center for X-Ray Optics, as well as Jun Ye in Boulder, a new laboratory was recently constructed by Thomas Pfeifer, Lukas Gallmann, Aurélie Jullien, Phil Nagel, Mark Abel, Willem Boutu, and Justine Bell. Using a carrier-envelope phase stabilized pump laser, we produced isolated attosecond pulses that will be used to study the timescales of electron dynamics in atoms, molecules, and nanoparticles. I now have a dream to understand molecular dynamics on electronic timescales. Taking what I know from vibrational and state-resolved dynamics, and applying it to problems of the motions of electrons and electron correlations among orbitals, it will hopefully lead to a fertile new field of experimental science and become one of the playgrounds for the Leone group in the upcoming years.

The phases of my career do not yet seem complete, and therefore I have a lot of mixed emotions while writing this autobiography. I am tremendously honored that my former students, postdocs, and colleagues put together this issue for me. Mary arranged a spectacular birthday celebration and I was overwhelmed by the compliments about my mentoring, which has always been so important to me. I therefore hope that the publication of this special issue calls attention to the remarkable careers of my many former students and postdocs and the wonderful work they are doing. It is the students that give me the greatest satisfaction and measure of success of my career. You will undoubtedly recognize many names without being told where each of them is located now—former Leone group members having established award-winning careers in many areas: academics, government laboratories, industry, and even law, for which I am very proud. The group could not continue to be built and managed without the excellent help from my previous assistants, Marissa Tablan, Mary Holloway, and Cheryn Gliebe, and current assistants, Kathleen Fowler (the visa wizard) and Adam Bradford, and finance guru and business manager of LBNL Chemical Sciences, Angela Gill. Finally, I am looking forward to more years of training young co-workers, as well as pioneering new scientific directions—with health, grants, earthquakes, and Mary permitting. Love you, Mary, and thanks for all your love and support.

Stephen R. Leone

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