Autobiography of Robert W. Field

I was born June 13, 1944 in Wilmington, Delaware, the first of two children of Kay and Edmund Field. It was an inauspicious beginning. The obstetrician's first words to my mother were "Young lady, you should have had a Caesarian", words meant to deflect his responsibility for my broken clavicle, injured facial nerve, and brain damage. My brother, Jay, was born (by Caesarian) three years later, just before the Field family returned permanently to Chicago.

My father began working for DuPont as a (Quantum Mechanics-free) Physical Chemist immediately after receiving his 1936 Ph.D. at Northwestern University (supervised by Ward Evans, of later fame in the trial of J. Robert Oppenheimer). Before my birth, my mother had been an elementary school teacher in New York City, largely to escape the cultural and political iron grip that DuPont exerted over Wilmington. With such superb chemist and educator role models, I may have been predestined to become a professor of chemistry.

My father's parents (Noah and Marie) emigrated from Kharkov and my mother's parents (Arthur and Anna Huebsch) from Bohemia and Hungary. Ed and Kay met at Northwestern. In Kay's first Chemistry course, Professor Ward Evans announced, "Young lady, I know that you are only in this course to find a husband". This made her so angry that she got the highest grade in the course, *and* a husband. In the lab my mother had produced an extremely precarious glassware set up, and her TA (Ed of course) asked, "Young lady, just what do you think you are doing?", and they were off and running.

One of my earliest memories is when my mother put me, for the first time, in a "Baby-Tenda" (a small table with a recessed baby seat in the center) and gave me my first taste of bacon. My joy was exceptional and I began making an enthusiastic, singing-like noise that became known in the family as my "bacon song". So began my love of both food and control over an initially small region of the visible and reachable world around me. I also loved music (especially Leadbelly) and considered phonograph records to be almost sacred objects. One day, perhaps because my fondness for food had resulted in some rapid growth, my mother told me that I had broken a record, upon which I became inconsolable because I was unaware that "record" has two meanings. Once this was explained to me, I continued collecting and breaking the two kinds of records with bacon-like joy. I grew fast and read many books, the Doctor Doolittle and Wizard of Oz series being two of my favorites.

One Halloween, my mother created a "professor" costume out of an old cap and gown with a light bulb (as a cartoon symbol of ideas) attached to the cap. I loved this costume and remember the one time I wore it as if it were yesterday. More predestination?

I attended the Avalon Park elementary school through fifth grade. The Chicago public schools in the 1950s were a laboratory for how *not* to foster curiosity, learning, or two-way dialogue between student and teacher. Once I got a grade of "F" in "Science". There had been one of the infamous "film strips"; this one showed a house in Holland that had a tile-covered roof. I put up my hand (something that shy Robin never did in those days) and held it up long after the "discussion" of the filmstrip had ended. I *had* to tell the class that our house at 8211 Kimbark also had a green tile roof, which (in addition to the small hill at the front of the house) was a unique architectural feature in the neighborhood. The teacher became furious that I would not put my hand down until I shared this remarkable fact with the class. She could not have understood how important it was for me to feel special by being different.

The Lab School. I transferred from Avalon Park to the University of Chicago Laboratory School at the beginning of sixth grade. It truly was a laboratory *for* (rather than against) education. This was one of the most important turning points of my life. During the first few weeks at the Lab School, I was pleasantly astonished by the profound change in the relationship between students and teacher from what I had come to accept at Avalon Park. The role of the teacher was not merely to keep order in the classroom and to "cover" a fixed curriculum. Morton Tenenberg, my homeroom teacher, was genuinely curious about what each of his students thought and felt about *everything*. Learning was by dialogue, not pontification, and certainly not organized around standardized tests. Mr. Tenenberg ("Teney") made all of us believe that our childish ideas were worth something, and that when we brought something new and special into the class, we would get the attention and intellectual reinforcement that we all so desperately wanted. No child was left behind.

A few years later, I found myself once again in Mr. Tenenberg's classroom, this time freshman geography. He did something that seemed very strange, but I now recognize as an educational stroke of genius. We discussed *whether* we would study each of the countries in the world. Each student was assigned to be an advocate for one country. We spent the entire year discussing which countries we should study, but never made the transition from deciding to studying. Of course, such a transition would have been superfluous.

So how did I end up as a chemist? I was torn between two extremes: working with people, ideas, and emotions or alone in a laboratory with objects and facts, respectively reflecting what seemed to me to be the essence of my mother and father as role models. But, as I would eventually discover, this was a false dichotomy.

In high school I took courses in Physics and Chemistry, but avoided Biology. I was nearly swept away by the enthusiasm of my Chemistry teacher, Richard Bartosiewicz. He created a three-track system, with separate problem sets for each track. I voraciously consumed all three sets of problems every week, but Chemistry seemed too easy to be respectable. I really was a pompous snot.

I remember many visits to my father's lab at Standard Oil of Indiana (now BP Amoco) in Whiting, Indiana. I was fascinated by his many small and large toys, but I never got a sense from those visits of the nature of research or even that there was anything more to Chemistry than making stuff by a cut-and-try Edisonian process. I was more interested in my father's lab instruments and in the frog's leg lunches at Phil Schmidt's Restaurant. About 15 years later while I was a postdoc, when for the first time we both attended an ACS meeting, my father told me the story about his invention and successful pilot plant scale demonstration of a process to make high density polyethylene (a process which, in the infinite wisdom of management, Standard Oil declined to pursue). I saw, for the first time, my father's passion for research and his transformative joy at achieving his objective. This came at a time when I too had

10.1021/jp909444e CCC: $40.75
Published on Web 11/19/2009

begun to achieve research objectives far beyond my imagining. But I am getting ahead of my story.

I had summer jobs in chemical laboratories. At Nalco Chemical (1960−1962) in Morrie Mindick's lab, I cleaned glassware making lavish glove-free use of acetone and benzene, performed friction tests on cardboard treated with various silica sols, and measured the capacity of ion-exchange resins. In Herb Hyman's inert gas fluoride group at Argonne National Laboratory (1963−1965), I had my first tastes of spectroscopy and building a scientific model. I prepared solutions of ultrapure SbF_5 in ultrapure anhydrous HF and recorded fluorine and proton NMR spectra of these solutions in Joe Katz's laboratory. The tantalizingly complicated dependence of the chemical shifts on SbF_5 mole fraction demanded some sort of physically plausible model, which I attempted to formulate. This was my first real taste of chemical research. It was not a matter of filling in blanks in a table. And it was not simple to demonstrate whether the model was true or false.

During my senior year at U-High, I had one of the great musical experiences of my life. I attended the First University of Chicago folk festival. The highlight for me was a workshop on the Carter Family, conducted by the New Lost City Ramblers.

Amherst College. When I arrived at Amherst College in 1961 I was determined not to major in Chemistry. It was going to be History, Economics, or English—something *difficult* that involved ideas, emotions, discussions, writing, and certainly not chemicals. I remember pushing a snotty letter under the office doors of Professors Robert Whitney and Marc Silver, who taught the Organic Chemistry course. I was outraged that the principles and concepts content of the course seemed to be dwarfed by facts that could only be memorized. I took a Quantitative Analysis course from Cooper Langford. I hated this course, and I was too much of a klutz to make measurements at 0.1% accuracy. But it was a discussion with Langford that changed my scientific life. I asked him a question that somehow boiled down to whether a transition was allowed or forbidden. He picked up a tiny scrap of paper and proceeded to perform an elementary group theoretical calculation. I wanted to know what he had written on that scrap of paper. I wanted to be able to make such simple yet powerful calculations. I was hooked.

I ended up majoring in Chemistry and doing my undergraduate thesis project with Cooper Langford. The goal was to discover how complexation of aniline by a transition metal (Co or Pd) modified aniline's electronic structure and how this modification differs from that caused by simple protonation. Another challenge to formulate a model! I had become an electronic spectroscopist!

Graduate School. When I applied to graduate schools, I specified my area of interest as "Theoretical Inorganic Chemistry", the naiveté of which should have ensured the universal rejection of my application. I decided to go to Harvard, with the expectation of working for Professor William Lipscomb. But, after my first day in the Chemistry 160 (Structural Inorganic Chemistry) class at Harvard, I knew that Bill Klemperer's vision of how molecules work was going to be my vision. And, in addition, there was Bill's extraordinary enthusiasm. When I told Bill that I wanted to join his research group, I noticed a moment of hesitation, and I am sure he was thinking of the nonsense about Theoretical Inorganic Chemistry in my application essay. I never spoke to Bill Lipscomb.

During my first year at Harvard I served as a Teaching Fellow in the Physical Chemistry course taught by Professor George Kistiakowsky. His lecture on the *wrong way* to blow glass, equipped with a spring-loaded, Mg-filled, "accidentally" burning tie was memorable. At least five of the students in Kisty's course figured prominently in my Harvard and post-Harvard life. Jeremy Thorner, Walter Struve, and Ed Laws enrolled as graduate students at Harvard and became my roommates for three years. Bill Gelbart and I were roommates during the summer after my first year at Harvard. Bill (a cellist) introduced me to the two Schubert piano trios, which ignited my passion for string chamber music, especially Beethoven and Shostakovich. Last but certainly not least, Kate Kirby introduced me, in 1983, to my wife, Susan Geller.

The Klemperer group was a marvelous place to become a spectroscopist. We all believed that we were the center of the spectroscopic universe, and perhaps we were. We developed unconventional experimental methods to measure unconventional quantities, from which we constructed unconventional models for the structure of unconventional, often floppy molecules. The group was divided into two mostly noncommunicating subgroups: superhigh resolution molecular beam electric resonance spectroscopy upstairs in the basement and optical spectroscopy and collisional energy transfer ("clunk work") downstairs in the sub-basement. Despite the >20 person size of the group, every day Bill walked through the lab and made suggestions to almost everyone. They were great suggestions, but it was an embarrassment of riches. We cautioned each other that if Bill proposed the same idea three days in a row, it was probably the idea of a lifetime. Also, Bill liked to make bets about what we would observe: $1 million if he was just blowing smoke or a nickel if he was certain.

My project was to measure the electric dipole moment of the CO $A^1\Pi$ state by radiofrequency-optical double resonance Stark spectroscopy. In those pretunable laser days, the CO was to be excited with rovibronic selectivity via an accidental coincidence with an atomic emission line. Eventually this morphed into electronic excitation via CO $A^1\Pi{-}X^1\Sigma^+$ resonance radiation generated in a microwave discharge lamp. Since the radiative lifetime of the CO $A^1\Pi$ state was only 10 ns, extremely high intensity radiofrequency radiation was required to stimulate the Λ-doublet transition at a rate competitive with radiative decay. The combination of VUV radiation, >100 V/cm radiofrequency electric fields, and 50 mTorr CO pressure resulted in frequent lightning strikes in direct view of the photomultiplier.

It was an extremely difficult experiment and I had a very wide Λ-doubling frequency region to search. In order to reduce the search range, I decided to fit to an effective Hamiltonian model every scrap of spectroscopic information that might have been relevant to the CO $A^1\Pi$ v, J-dependent Λ-doublet transition frequencies. I fitted *everything* known from 1930−1970, especially spectroscopic perturbations, for *all* of the valence states of CO. I got more than just a little bit carried away. What had begun as a side project became my Ph.D. thesis.

Spectroscopic Perturbations. When the dust settled I had a complete collection of fitted perturbation matrix elements between the CO $\sigma\pi^*$ $a^3\Pi$ and $A^1\Pi$ states and the $\pi^3\pi^*$ $^{1,3}\Delta$, $^{1,3}\Sigma^-$, and $^3\Sigma^+$ states. I was able to show that each observed matrix element was the product of a constant electronic factor and a calculable vibrational overlap integral. This "matrix element method" was useful for predicting the magnitudes of unobserved perturbation matrix elements and, later, for assembling a group of perturbing vibrational levels into one absolutely vibrationally numbered electronic state.

But there was one treasure embedded in my fit results that I did not know how to capture. I was sure that there had to be a fundamental relationship among the magnitudes of the electronic factors for all perturbations between states belonging to the $\sigma\pi^*$ and $\pi^3\pi^*$ electronic configurations. I asked Bill Klemperer for help with this problem and, without hesitation, he outlined how to reduce many-electron inter-*state* matrix elements to sums of one-electron inter-*orbital* matrix elements. To this day I have no idea how Bill knew this trick, which has been, for almost forty years, the most used tool in my theoretical armory.

I liked perturbations, but collecting perturbation data seemed like picking through other spectroscopists' garbage. Level shifts, "intensity borrowing", interference-based intensity anomalies, and "extra lines" were aspects of broken patterns. Assignment of spectra is necessarily guided by patterns. Most spectroscopists seemed to regard perturbations as occupational hazards. Perturbations made assigning spectra difficult, they made tabulating spectral data awkward, and they seemed to convey little physical content other than the existence of an accidental degeneracy. There were a select few spectroscopists who shared my fascination with perturbations: Richard Barrow, Albin Lagerqvist, Joelle Rostas, Herb Broida, and Hélène Lefebvre-Brion. I met all five as a result of our shared interest in perturbations, and each profoundly influenced my scientific trajectory.

It took me many years to appreciate that regular patterns indicate simple, separable dynamics and broken patterns encode complex, hence interesting, dynamics. Eigenstates are stationary. Spectra are transitions between eigenstates. Therefore spectra might seem to reveal only static molecular structures. The physical chemistry community was beginning to get bored with structure and infatuated by dynamics. It became fashionable for frequency-domain spectroscopists to use the phrase "structure *and dynamics*" without any concrete idea of the meaning of this word "dynamics". Perturbations imply that the usual approximate constants of motion are no longer conserved and cannot ensure the Born−Oppenheimer separability of electronic states and vibrational modes. Population might be collisionally funneled from one electronic state to another via a very small number of mixed eigenstates. Perturbations *are* dynamics! There are many kinds of broken or nonstandard patterns in a spectrum, some of which might be called meta-patterns. *Assignment* of perturbed spectra requires nonstandard experimental strategies, especially multiple resonance schemes. *Access* to key perturbations can require a full career to develop the necessary tactics. Unusual properties must often be sought in order to *recognize* a special class of target level. My graduate students continue to teach me new tricks for assignment, access, and recognition of dynamically special states. Once again, I am getting ahead of my story.

Even after I had completed my fits of all of the CO spectra and finished writing my thesis, I had not given up hope that, after 6 1/3 years of attempts, my CO RFODR-Stark experiment would work in its final implementation at Harvard. It failed one final time. But I was not ready to give up. I had chosen H. P. Broida's research group in the Quantum Institute at UCSB for my postdoc. I knew that Herb Broida had done pioneering microwave-optical double resonance spectroscopy (with Harry Radford, Ken Evenson, David Pratt, and David Harris) on the CN $B^2\Sigma^+(v=0)\sim A^2\Pi(v=10)$ perturbation. These experiments exploited a discharge source of electronically excited CN, "doorway mediated" collisional transfer between the A and B states via the A∼B-mixed perturbed levels, and steady-state nonequilibrium population distributions that resulted from the very different radiative lifetimes of the CN A and B states. I knew that Herb had several lasers in his lab, and I knew that David Harris had a well-equipped microwave setup. So I planned to assemble a new version of my CO experiment at UCSB. However, Herb Broida had different secret plans for me!

Postdoc at UCSB. A few years before my arrival at UCSB, Katsumi Sakurai had completed an enormously successful postdoc in Broida's group. During that postdoc he collaborated with Broida's student Stan Johnson in the construction of an early version of the extremely versatile "Broida oven" source of refractory metal oxide and halide diatomic molecules. He discovered that there were several coincidences of BaO $A^1\Sigma^+$-$X^1\Sigma^+$ rovibronic transitions with Ar^+ laser lines, including one (496.5 nm) located at a very low J'' value (the R(2) line of the (7,0) vibrational band). Katsumi and Herb knew that this BaO, Ar^+ laser combination was ideally suited for microwave-optical double resonance spectroscopy in both the X and A electronic states of BaO.

Herb Broida realized that I was a super arrogant, possibly talented Harvard graduate. He knew that I was determined to pursue my vendetta against CO. So Herb decided that he would trick me into believing that a BaO MODR experiment was *my idea*. On my first day in the Broida lab, Herb suggested that I read Katsumi's paper on BaO A-X transitions excited by various Ar^+ laser lines. He never said one word to me about Katsumi's idea for a MODR experiment, nor was anything written about this idea in the paper Herb had given me to read. Herb's stratagem worked! The next day I proposed to try exactly the same experiment that Katsumi had proposed to Herb.

Herb had a magical laboratory stockroom. He had the idea that it should be possible, using standardized components (monochomators on wheels, instrument racks on wheels, all laser beams 1 m above the floor, 0.5″ diameter precision machined stainless steel rods cut in standard lengths), to set up a new experiment from scratch in one day. There is a story about Herb visiting the Zare lab and teaching the students, with a few steel rods and lenses that he had carried in his briefcase, how to image laser excited fluorescence into a monochromator.

Neither Broida nor Harris had the necessary microwave components in the 37 and 45 GHz frequency ranges required respectively for the $J'' = 1-2$ and $J' = 3-2$ rotational transitions. Herb told me to call a friend of his, who worked in a General Motors research lab in Santa Barbara, to ask whether he could lend us the necessary components.

The next day we had everything we needed and I began to learn how to detect, scan, and measure the frequency of microwave radiation. At Harvard, I had worked exclusively in the <2 GHz radiofrequency region, and now there were many new tricks I needed to learn. Foremost among these was how to measure the frequency of radiation from a free-running klystron by beating it against a harmonic of a lower-frequency oscillator, the frequency of which could be measured using a HP frequency counter. For convenience in setting up microwave scans over each 10 MHz wide region, I calibrated an ∼1 MHz precision wavemeter and found that the true frequency differed from that on the wavemeter dial by 30 MHz.

Lagerqvist and Barrow had reported an analysis of the extensively perturbed optical spectrum of BaO in 1950. This was an interesting coincidence because Lagerqvist and Barrow had been my spectroscopic heroes ever since their perturbed CS spectra had played a crucial role in guiding my only successful experiment at Harvard, the RFODR-Stark spectrum of electronically excited CS. Their BaO A-X molecular constants permitted me to restrict the MODR search region to only ∼30 MHz. About 4 days after arriving at UCSB, two of Broida's students (Bob Bradford and John West) and I began searching for a MODR signal in BaO. We had learned how to optimize BaO generation in the Broida oven, imaging of the Ar^+ laser induced BaO A-X fluorescence onto the PMT, and scanning the microwaves. We also verified, from the dispersed fluorescence spectrum, that the Ar^+ laser line was exciting BaO out of the $v'' = 0$, $J'' = 2$ level. Everything was working perfectly. But we saw no MODR signal after a full week of scanning.

A week was nothing in comparison to my 6 years of failure at Harvard. But I had run out of ideas for things to optimize. It seemed as though it might be time to give up and try a completely different experiment. Then I discovered that I had incorrectly remembered the sign of my 30 MHz correction of the wavemeter. Writing coherently in lab notebooks is an art I have never mastered. The first scan, centered 60 MHz away from where we had initially been scanning, yielded a very strong MODR signal. Within two weeks of my arrival in Broida's lab, *I had succeeded* at an experiment reminiscent of my failed Klemperer-lab experiment. I was on top of the world. *I* had made it happen. It was *my* idea. And Herb Broida never reminded me that I had borrowed two of his students, exploited his stockroom, and been artfully fed Katsumi's idea.

Herb told me that I should apply for some local Quantum Institute funds to purchase needed improvements of the microwave setup. Why should I have to waste time doing this? I was busy being superproductive in the lab. I wrote a short proposal and, undoubtedly because Herb had prearranged the whole thing, the request was immediately approved. Now I could set up what I considered to be a state of the art microwave system for MODR experiments.

And then Herb kicked me out of his lab and took away his two graduate students who had been helping me with the experiments. He told me to rebuild an improved MODR setup in an empty laboratory room that was available. I was self-righteously furious. Why should an experiment that had been so successful, more so than any of the other current experiments in Herb's lab, be forced to undergo such a momentum destroying transformation?

And almost as an afterthought, Herb informed me that, Don Jennings, a sabbatical visitor from the National Bureau of Standards in Boulder, would be setting up a continuous wave (cw) dye laser in my new lab. I agreed that a tunable laser would be vastly better suited to MODR experiments than accidental coincidences with Ar^+ and Kr^+ laser lines, but it seemed unlikely that such a laser would have the intensity, stability, reliability, and ease of tunability needed for Broida oven MODR experiments. By this time I should have known that Herb was forcing me to do what I was too stubborn to do for myself and, at the same time, always making it possible for me to believe that *I* was the one who had sole responsibility for making all of the good things happen.

As it turned out, Don Jennings was among the world leaders in the development of cw dye lasers. His home-built lasers were easy to assemble, reliable, and convenient to operate. A huge amount of data began to flow out of the new lab. But there was always a tension between Don's goal of improving the laser and mine of recording more spectra. We learned to live with it. And Don was a great bluegrass banjo picker.

Captain (USAF) C. Randy Jones was also a visitor in the Broida laboratories. The Air Force was interested in developing chemical lasers as weapon systems that could be flown in airplanes. The simple idea behind this was that the technology for building rocket engines that burn about 1 lb/s of fuel was well-known to the Air Force. An order of magnitude calculation suggests that fuel combustion of 1 lb/s corresponds to a power of more than 1 MW. A chemically powered device would be much more "portable" than an electrically powered one. If it were possible to convert a significant fraction of the chemical energy into optical radiation, a new class of flyable weapon system would be created.

Herb Broida assigned Randy Jones the task of measuring the absolute quantum yield for converting the exothermicty of a gas phase metal oxidation reaction into visible wavelength chemiluminescence. It turned out that the $Ba + N_2O \rightarrow BaO^* + N_2$ reaction had a 30% quantum yield for chemiluminescence in a perturbed vibrational band of the BaO A-X electronic transition. I *owned* the BaO A-X system. I was an expert on perturbations. Herb's CN MODR experiments had exploited nonthermal population distributions that were the result of population funneling through perturbed levels in CN. The most intense BaO chemiluminescence seemed to originate from $A^1\Sigma^+$ levels locally perturbed by $a^3\Pi$ levels. All of a sudden spectroscopic perturbations had the potential of being transformed from other people's garbage into the key to a militarily important new technology.

I set out to prove that the vibrational levels of the BaO $A^1\Sigma^+$ state that were most prominent in the $Ba + N_2O$ chemiluminescence were those that were perturbed by vibrational levels of the $a^3\Pi$ state. Albin Lagerqvist had observed and rotationally assigned extra lines at many local perturbations in the BaO $A^1\Sigma^+$ state but had not made electronic ($^1\Pi$, $^3\Pi_0$, $^3\Pi_1$, $^3\Pi_2$, $^3\Sigma^+$) or vibrational assignments of the perturbers. I set out to make these perturber assignments using the "matrix element method" that I had demonstrated in my Ph.D. research. In order to make this unconventional method of electronic and vibrational assignment more convincing, I decided to extend it to MgO, CaO, SrO (all of the raw assignments were from Lagerqvist), and BeS (raw assignments from Barrow).

I was astonished at the completeness and reliability of Lagerqvist's and Barrow's rotational assignments. Their work became my ideal of spectroscopic scholarship. Even when a complete assignment picture could not be drawn, using available tools, from the raw data, the analysis was pushed to the limit of what could be established with certainty, and all of the fragmentary data was published in a transparently usable format.

My analysis of the CaO perturbations was challenged by Leo Brewer, based on the qualitative difference between the energy order of electronic states in the isovalent CaO and C_2 molecules. This was my first experience of scientific disagreement. Leo Brewer set a wonderful example of gentlemanly disagreement. In order to convince him that my analysis was correct, I had to extend the analysis to all extant data on molecules with eight valence electrons and I had to find a reason why C_2 was so different from CaO. This led me to appreciate the profound difference between the zero-order electronic structures of covalently and ionically bound molecules, which in turn pushed me toward what I consider to be my most important contribution to electronic structure theory: the atomic ion-in-molecule ligand field model, first elaborated with Steve Rice and Hans Martin (a Lagerqvist student!) for the Ca monohalides and then with Mike Dulick and Richard Barrow (!) (with Air Force Geophysics Lab support from Ed Murad) for the rare earth monoxides.

Largely as the result of my intense immersion in spectroscopic theory and my discovery of Jon Hougen's wonderful monograph NBS #115, I modestly told Herb Broida that I thought, in one or two group meeting lectures, that I could present a unified picture of *everything that was useful* in Herzberg's diatomic molecules book. Herb smiled inscrutably and immediately offered me two group meeting slots. Of course, I failed to accomplish even a small fraction of my arrogantly stated goal. This led quickly to my first experience teaching a graduate course at UCSB and the creation of a set of typed handouts that became the backbone of the graduate level spectroscopy course (5.76) that I taught for many years at MIT and THE BOOK that I cowrote (twice) with Hélène Lefebvre-Brion. Once again, Herb Broida knew exactly how and when to push my buttons.

I was enjoying a level of productivity and intellectual growth as a postdoc at UCSB that was far beyond anything I could have imagined during my graduate years in the Klemperer group. Herb Broida told me that I had to go to Washington to speak to people at ONR, AFOSR, NSF, and ARPA about writing a large-scale research proposal to support the UCSB program on electronic transition chemical lasers. Why should I, the superproductive one, have to waste my time trying to raise money? Why should research cost so much? What is this nonsense called overhead? Of course, Herb was right again. I went to Washington, learned a lot, and laid the foundation for a UCSB proposal that was funded shortly after I unexpectedly made my transition from UCSB to MIT.

Herb Broida told me that he wanted me to give his lecture at the annual AFOSR Molecular Dynamics contractor's meeting. This was one of the few things that Herb did for me that I did not fight. I was aware that this was a great opportunity for scientific visibility. After I gave my talk, Jim Kinsey asked me whether I might be interested in a faculty position at MIT. I was very interested!

As soon as I accepted the faculty position at MIT, my relationship with Herb Broida changed profoundly. It was six months before I would leave UCSB, and I expected that my enormously rich and supportive interaction with Herb would continue, perhaps elevated to a new level. Ever since my arrival at UCSB I had been meeting with Herb in his office for several hours every week. Herb cut me off cold. It hurt a lot! But, once again in retrospect, I know that this was the best possible thing Herb could have done. I had to define myself as a scientist. I had to decide how to create a research program at MIT that would be distinct from what Herb helped me to build at UCSB. I had to continue asking myself: why should anyone be interested in my research? Where is the surprise? What big picture questions can I answer using new technology and, most importantly, drawing on my unique vision of how molecules think? What, if anything, lies beyond tables of eight-digit molecular constants? What is the spectral simplification provided by double resonance good for? It is certainly suited to looking at high excitation energy, where textbook patterns are either entangled or broken and new classes of patterns await recognition.

MIT. At MIT, Jim Kinsey extended the education that Herb Broida had ignited. Although Jim had begun his career as a microwave spectroscopist, he was certainly not interested in spectroscopy as an end in itself, especially not *high-resolution* spectroscopy. Jim and I were very different in our choice of scientific problems, student mentoring styles, and approach to scientific results. I desperately wanted Jim's respect, not merely because earning it might be one prerequisite for being granted tenure at MIT. I wanted his respect because he was really smart, rigorously honest, and not afraid to say what he thought. Jim did not have to trick me into doing the right things, as Herb Broida had done. Jim helped me to want to do the best possible science. Jim and I were magnificently complementary. Jim left MIT to return to Rice University in 1988. I miss him.

I decided to build my MIT research program around the Broida oven, cw dye lasers, and optically pumped cw electronic transition lasers. I vowed to deny myself the pleasures of two very tempting technologies: molecular beams and microwave spectroscopy.

The Broida oven was a simple and versatile source of MX diatomic molecules from a huge swath of the periodic table: M = *all metals*, X = all halides, oxides, sulfides, and a few nitrides and hydrides. It was a 0.1−2 Torr vacuum flow system built from inexpensive and interchangeable components. The number density and ~300 K rotational temperature of MX molecules were ideally suited for fluorescence excitation (LIF) and dispersed fluorescence (DF) spectroscopies and the operating pressure made it possible to reduce rotation-changing collisions so that they did not interfere with the rotational selectivity of OODR and DF spectroscopies.

In 1974 there were very few cw tunable lasers in academic laboratories. I intended to build at least three replicas of Don Jennings' cw dye laser for use in optical−optical double resonance experiments. The sub-MHz resolution, ~100 mW average power, and few percent amplitude stability would allow me to run circles around the N_2 laser- and flash lamp-pumped dye lasers (and Optical Parametric Oscillators) in use in all other gas phase Physical Chemistry research groups.

I chose cw optically pumped electronic transition lasers for several reasons: I wanted to avoid competition with Herb Broida's group in the chemical laser area, I thought that optically pumped lasers (OPL) might eventually find use as gain probes for electronic transition chemical lasers, and I thought that OPLs would generate a huge number (many v',v'' bands and ~100 rotational transitions from each vibrational band) of fixed frequency laser lines at accurately predictable frequencies spanning the visible and near-infrared regions that were currently inaccessible to cw lasers. In fact, I attempted to patent the concept of cw, electronic transition OPLs. As it turned out, OPLs opened up new scientific directions for me that were vastly more interesting than my initial ideas.

My avoidance of molecular beams and microwave spectroscopy at MIT lasted for more than 30 years. It grew out of my suspicion that I would be incapable of resisting the temptation to engage in ultrahigh resolution spectroscopy. Esoteric electronic quantities, such as hyperfine coupling constants, electric dipole moments, magnetic *g*-values, lambda doublings, and spin-splittings, were instant yawn-producers among my colleagues. I wanted to find "big picture" ideas (ideally having some relevance to Chemistry) while remaining a card-carrying high-resolution spectroscopist.

MIT provided $40,000 (1974 dollars) to set up my lab. I purchased an Ar^+ laser and a 1 m Spex monochromator and spent the rest on home-built dye lasers, copies of Broida-lab style optical components, instrument racks on wheels, and a laser table with a 10″ diameter hole (to accommodate a Broida oven inside a laser cavity). I made a bet with one of my colleagues that I would observe a dispersed fluorescence spectrum of BaO before Christmas break of my first year at MIT. A few days before the deadline, all of the necessary equipment was in place and working, but the machine shop had not yet finished welding together the rack-on-wheels for the bulky and heavy monochromator. We borrowed a forklift, moved the monochromator into position near the Broida oven, and won the bet.

My first two graduate students were Rick Gottscho and Brooke Koffend. Rick "owned" the Broida oven, BaO, and OODR. Brooke achieved miracles with the cw optically pumped I_2 laser. The lab was only marginally air-conditioned and we used city water to cool the Ar^+ laser (often requiring replacement of large and expensive water filters, two every hour). During the summer the temperature in the lab would often rise above 100 °F, and only Rick was tough enough to work long shifts in the lab, usually stripped to his underwear. He had a magical ability always to finish a shift in the lab having achieved a valuable experimental result, regardless of which subset of equipment failed to work on any given day.

Rick Gottscho demonstrated the fundamental capabilities of OODR. He showed that the first laser produced velocity- and M_J'-selected populations in the intermediate v', J' levels, which were sampled in the directly pumped and collisionally populated $J' \pm 1, 2, ...$ rotational levels by the second laser. I was very proud of Rick's measurements of velocity- and alignment-relaxation in rotation-changing collisions of BaO with Ar and CO_2. Rick also showed that the OODR effect could be detected as a dip in the fluorescence intensity from the intermediate level when the second laser was tuned through resonance. Rick was the first to exploit perturbations in the intermediate state in order to gain access to classes of otherwise unobservable electronic states. He observed intensity anomalies in the ratios of R($J - 1$), P($J + 1$) rotational transitions in the dispersed fluorescence from an upper level that was of $^1\Pi,^1\Sigma^+$ mixed character. These anomalies were an example of quantum interference between parallel and perpendicular type transition amplitudes. I am a collector of interference effects.

Brooke Koffend's I_2 OPL experiments were astonishing. He used the Ar^+ laser 514.5 nm line to longitudinally pump a 2-m long I_2 cell. The side fluorescence was very intense. The first time Brooke aligned the resonator mirrors at the front and back of the I_2 cell, the side fluorescence practically vanished, owing to competition between the spontaneous fluorescence over 4π steradians and the stimulated emission exclusively in the forward direction. Brooke had two marvelous ideas for experiments with his OPL. One led to a collaboration with Professor Shaoul Ezekiel of MIT. If we used a single longitudinal mode Ar^+ laser, locked to an ultrastable frequency reference (a Lamb dip associated with an I_2 hyperfine component), to pump our I_2 laser, and we locked the length of the I_2 laser resonator to maximize the OPL output intensity, we produced I_2 laser lines throughout the red and near-infrared regions, the frequencies of which were knowable to the same absolute accuracy as the ultrastable frequency reference. The second idea was to use the pressure-dependence of the gain of the OPL to measure the rate of rotation−vibration collisional relaxation of the population in the lower level of the laser transition. This lower level was a very high-v'' vibrational level of the I_2 electronic ground state. Since vibration−rotation and pure rotation transitions are electric dipole forbidden in homonuclear molecules, measurement of the pressure dependence of OPL gain provided an exceptionally convenient way to survey collisional properties of highly vibrationally excited molecules. One might have naively expected the collisional properties of high-v levels to be very different from those of low-v levels, but they were not.

I cannot resist another image from those early days in the lab. Brooke was a chain smoker. He developed amazing skill at drop-and-drag cleaning of a laser mirror while holding a smoking cigarette in the same hand as the mirror. No one could clean laser optics faster or better than Brooke. Mike Dulick used exactly the same cigarette-aided procedure; could Brooke have taught him?

Stimulated Emission Pumping. These early OODR fluorescence-dip and OPL side fluorescence-dip experiments (~1976), and the pressure dependent OPL gain measurements were pointing the way toward Stimulated Emission Pumping, but it took one more fortuitously bad idea to push me toward SEP. Jim Kinsey and I were very impressed with Robert Le Roy's long-range atom-in-molecule theory for the near dissociation rotation−vibration constants of diatomic molecules. We thought that it would be wonderful to develop a similar theory that related the near-dissociation level structure of small polyatomic molecules, such as S_2O, to the properties of the separated dissociation fragments, $S(^3P)$ and $SO(X^3\Sigma^-)$. We suspected that such near-dissociation levels of polyatomic molecules would likely be difficult to access and nearly impossible to assign.

Stimulated Emission Pumping was the ideal way to gain Franck−Condon-selective access to highly vibrationally excited levels of small polyatomic molecules. The only other game in town was high overtone pumping, which was ill suited for all but R−H stretching vibrations. S_2O was not well-known spectroscopically, too heavy to have conveniently large rotational constants, and not a permanent gas. Jim and I chose formaldehyde and acetylene as the warm-up polyatomic molecules to demonstrate the feasibility of SEP. What a pair of lucky choices! So we were off to the races.

We did our first SEP experiments on I_2 in 1978. I had originally planned to call the upward transition "PUMP" and the downward transition "DE-PUMP," but Jim Kinsey and the rest of the group insisted on the vaguely inappropriate "DUMP." I required very little convincing. So SEP became known as PUMP and DUMP, which turned out to be a great way to attract attention.

Formaldehyde was Brad Moore's molecule and acetylene was the joint property of Bill Klemperer and Kevin Lehmann. Jim Kinsey and I thought at the time that we were merely borrowing these molecules to demonstrate the capabilities of SEP. I would never have dreamt that 30 years (and perhaps 100 graduate student years) later I would still be working on acetylene.

In the very early days of polyatomic molecule SEP, Hai-Lung Dai applied for a postdoc in the Field−Kinsey research group. It was a no-brain decision to hire him, but I had no money. So I turned him down. Hai-Lung would not take "no" for an answer (and Brad Moore telephoned asking me whether I was crazy). Eventually I offered Hai-Lung the postdoc with the proviso that I might not be able to pay him! MIT is not known for its charity toward inadequately funded young faculty, and I knew that Hai-Lung and I were taking a big risk. That risk grew into a lifelong friendship and collaboration.

I took the absurd step of dividing my research group into three subgroups: diatomics, formaldehyde SEP, and acetylene SEP. The H_2CO and HCCH subgroups competed to be the first to record SEP spectra. It was a photo finish, H_2CO winning by a nose. But I had created a monster. The to-the-death-competing H_2CO and HCCH subgroups did not talk to each other. The only thing they agreed on was that diatomic molecules were beneath their threshold for tolerance. The diatomic subgroup dismissed the H_2CO and HCCH subgroups, accusing them of engaging in "spectrology". As it turned out, the HCCH subgroup, in its fascination with "quantum chaos" and my insistence that the $E_{vib} = 28\,000$ cm^{-1} spectra of HCCH were "intrinsically unassignable", was engaged in the purest form of spectrology.

The early years of H_2CO and HCCH SEP were very exciting. Every experiment we tried on H_2CO worked beautifully, but the results were spectroscopically and dynamically unsurprising. Every experiment we tried on HCCH also worked beautifully, but *all* of the results were tantalizingly mysterious.

There was one essential difference between the initial H_2CO and HCCH experiments. The H_2CO (Hai-Lung Dai, Pat Vaccaro, Dave Reisner, Carter Kittrell, and later Scott Halle, Caba Korpa, and Friedrich Temps) and HCCH (Evan Abramson, Dan Imre, Yongqin Chen, Carter Kittrell, and later Chuck Hamilton, Peter Green, Jim Lundberg, George Adamson, Stephani Solina, Jon O'Brien, Sarah Jane Cohen, Bhavani Rajaram, Michelle Silva, and Richard Duan) experiments were respectively "bottom-up" and "top-down".

Our H_2CO SEP experiments sampled the lowest vibrational levels and worked systematically upward, assigning and fitting the energy levels to the traditional anharmonic spectroscopic model. The occasional intermode anharmonic or Coriolis interaction could be understood in the framework of simple few-level local perturbations. We achieved an essentially complete sample of the normal mode and diagonal anharmonicity constants (ω_i, x_{ij}) and were able to compute all of the Coriolis interaction parameters using the standard normal mode model. We always knew in advance the normal mode assignment of every highly excited vibrational level we selected for studies of the Stark effect or collisional depopulation rates. Despite elegant and truly heroic PUMP−DUMP−PROBE and sub-Doppler resolution pulsed/cw laser experiments (Pat Vaccaro and Friedrich Temps), the highly excited vibrational levels of H_2CO *never* exhibited properties unexpectedly different from those of the zero-point vibrational level. H_2CO was too cooperative, so we eventually turned all of our efforts to HCCH.

30 Years of Acetylene. We began our HCCH SEP experiments with the highest vibrational levels (near $E_{vib} = 28\,000$ cm^{-1}, well above the acetylene ↔ vinylidene isomerization barrier) of the linear S_0 state into which we had good Franck−Condon access from low-lying, assigned vibrational levels of the *trans*-bent S_1 state. The SEP spectra told two quite different stories. Our 0.03 cm^{-1} high-resolution SEP spectra were incomprehensible in the jwesense that we were unable to recognize any $BJ(J + 1)$ rotational energy level patterns when we made stacked term energy plots of SEP spectra originating from successive J', $J' + 1$, $J' + 2$, etc. upper levels. In contrast, our 1 cm^{-1} low-resolution SEP spectra exhibited multi-eigenstate "clumps" and all of the clumps followed simple $BJ(J + 1)$ term energy curves.

The absence of rotational patterns in the high-resolution SEP spectra of acetylene suggested that all vibrational levels were of profoundly mixed normal mode character and that these mixtures change drastically from J to $J + 1$. This led me to make my infamous statement that the highly vibrationally excited rotation-vibration energy levels of acetylene are "intrinsically unassignable". These are strong words for a spectroscopist! Quantum chaos was a hot topic in those days, and we set out to prove, using a variety of statistical measures (foremost among these was the adjacent level-spacing distribution), that our SEP spectra of acetylene were the first molecular spectra to exhibit quantum chaos, which is just a fancy way of saying "intrinsically unassignable". However, we found that all of the statistical measures were corrupted by inadequate resolution and dynamic range and, most importantly, by the existence of several *a priori* unknowable *approximate* constants of motion. In retrospect, any chemist should have known that most chemical bonds were strongly conserved quantities and that no molecule could reach the "bag of atoms" limit at energies lower than the first dissociation asymptote.

The presence of $BJ(J + 1)$ patterns in the clump spectra suggested the existence of a special class of multieigenstate "feature states." Each feature state is a group of strongly anharmonically mixed zero-order normal mode states that derives its intensity from one special Franck−Condon bright zero-order state. Why is the brightness confined to a J-invariant group of states? It took several years and a trans-Pacific battle for the concept of polyads to emerge.

During a short visit to MIT, Kaoru Yamanouchi (a postdoc from Professor Soji Tsuchiya's laboratory) suggested that it would be a good idea to record dispersed fluorescence (DF) spectra of acetylene. I was dead set against this idea because I was certain that the intensity of fluorescence that could be excited by a pulsed dye laser would be too weak to yield dispersed fluorescence spectra at resolution sufficient for vibrational assignments, except at very low E_{vib}. In addition, 20 cm^{-1} resolution DF spectra would likely be inferior to our low-resolution SEP spectra. The *only* good things about DF spectra would be the ability to cover a broad spectral region quickly and, when using an ICCD array detector rather than a photomultiplier, obtain meaningful relative intensity measurements. As usual I was quite wrong.

Being wrong is usually instructive, as it proved to be in this case, for two reasons: (i) low-resolution means early time dynamics and (ii) relative intensities enable statistical methods for pattern disentanglement. Twenty cm^{-1} spectral resolution meant that we were blind to dynamical processes that occurred more than 3 ps after the initiation or "pluck" of the dynamics on the S_0 potential energy surface from a well characterized and systematically selectable S_1 vibrational state. The nature of the initial pluck was perfectly known and, within a limited range of *trans*-bending and CC-stretching normal mode displacements, systematically variable. The spectral splittings and relative intensity patterns in the 20 cm^{-1} resolution DF spectrum would eventually provide a description of the *earliest-time dynamics* subsequent to a *perfectly specified initial pluck*. But first we needed to understand that the early time dynamics was exclusively intrapolyad dynamics and, in order to restrict our view to the dynamics within only one polyad at a time, we needed to figure out how to disentangle the overlapping spectra of different polyads. The key to this disentanglement proved to be the relative intensity information that was vastly superior in the DF spectrum to what could be obtained from SEP spectra. Once again, I have gotten ahead of myself. We needed to learn about *polyads*.

Kaoru was collaborating with David Jonas. David realized that several very strong anharmonic resonances had, for many years, been known and understood in the infrared spectra of acetylene. The existence of polyads (groups of zero-order vibrational states so strongly linked by off-diagonal matrix elements that diagonalization of an effective Hamiltonian matrix rather than second-order perturbation theory is required to represent the observed vibrational eigenstates) was accepted dogma among vibrational spectroscopists. But polyads were a big surprise to us. The "resonance vector" analysis of Fried and Ezra and Kellman identified all of the approximately good quantum numbers that survive the complete set of quantum-number-destroying anharmonic resonance mechanisms known in the S_0 state of HCCH. These are the *polyad quantum numbers*.

David Jonas showed how to apply the polyad model to our DF spectra of acetylene. One nontrivial obstacle to David's realization of the appropriateness of the polyad model is the qualitative difference between how infrared and SEP spectra access each polyad: the nature of the "bright" state. In the infrared spectrum, the bright state in each polyad is a normal mode zero-order state that has an odd number of quanta in one of the IR-active normal modes, ν_3 (the antisymmetric CH stretch) or ν_5 (the *cis*-bend). In our DF spectra, the bright state is a zero-order state that has zero quanta in all of the Franck−Condon inactive modes (only ν_2 and ν_4 are Franck−Condon active). A more daunting obstacle to the use of the polyad model was collegial. David had to convince Kaoru Yamanouchi and Kaoru had to convince his advisor, Professor Soji Tsuchiya, that the standard vibrational assignment procedure used by electronic spectroscopists, picking out progressions in the Franck−Condon active normal modes, must crash and burn at high E_{vib} in polyad-forming molecules. Of course, the traditional progression-following procedure is physically sound, but it fails to deal explicitly with the systematic increase vs. E_{vib} in off-diagonal intrapolyad anharmonic coupling matrix elements. These off-diagonal matrix elements affect the DF spectrum in two ways that were familiar to me from the analysis of simple spectroscopic perturbations: intensity borrowing and level-repulsion. The nominally dark states borrow intensity from the Franck–Condon-bright state and are shifted out from under the artificially broadened bright state. Each Franck–Condon progression must inevitably split into fragments too numerous for unambiguous assignment.

Even though the polyad- and Franck–Condon-progression-following-assignment schemes were both founded on the same set of Franck–Condon-active bright states, it seemed to the participants that no mutually acceptable set of assignments could be negotiated. A trans-Pacific battle of the titans ensued. The best way to gain a deep understanding of a scientific concept is to explain it to a colleague, especially to one with whom you have a profound but respectful disagreement. The breakthrough came when Kaoru proposed what were ultimately proven to be correct vibrational assignments built on $2\nu_5$ (two quanta of the *cis*-bend) and David realized that the brightness of these nominally dark states originated in the 2:2 anharmonic (Darling Dennison) interaction in which two quanta of bright ν_4 are exchanged for two quanta of dark ν_5.

The polyad model has led my research group on a magical mystery tour. Originally, because all known HCCH S_0 anharmonic resonances were embodied in our polyads, we called them "super-polyads". Kevin Lehmann objected, provocatively suggesting that the use of the modifier "super" inappropriately evoked "mine is bigger than yours". Kevin was right.

Matt Jacobson realized that each polyad is illuminated by exactly one bright state. This means that the pattern of relative intensities of all transitions into a given polyad does not depend on the S_1 vibrational level from which a particular polyad is viewed. But when polyads 1 and 2 occupy overlapping energy regions, the members of the two polyads may be disentangled by viewing the two polyads from S_1 state vibrational levels A and B. The different polyad 1:2 intensity ratio from level A vs B enables a remarkable statistical procedure, "spectral cross-correlation (XCC)", by which an initially unknown number of unknown spectral patterns may be deconvolved.

The existence of a large number of XCC-separated polyads provided input for a multiresonance effective Hamiltonian fit model that accounts for a huge body of DF and SEP spectral data. Matt Jacobson, in separate collaborations with Howard Taylor and Mike Kellman, employed Heisenberg's version of the correspondence principle in reverse to convert the HCCH reduced-dimension, quantum mechanical effective Hamiltonian to a classical mechanical pure-bending Hamiltonian. Using the "surface of section" tool of nonlinear classical dynamics, the structure of pure bending phase space can be systematically mapped, showing the emergence of regions of chaos and the birth, at bifurcations, of qualitatively new classes of quasi-periodic orbits. One particularly interesting class of stable, large-amplitude motion is the local-bender.

Both the spectroscopic and the classical mechanical effective Hamiltonians show that the local-bender motion becomes increasingly stable at high vibrational excitation energy. This local-bending coordinate is striking in its resemblance to the acetylene ↔ vinylidene minimum energy isomerization path. The chemical significance of large amplitude eigenstates organized along an isomerization path is astonishing. If such eigenstates actually exist, they could be used to spectroscopically map the infamous "reaction coordinate" of introductory chemistry courses. They might also provide a rational basis for the design of efficient external control schemes.

The spectroscopic effective Hamiltonian is a reduced-dimension fit model. It is not the exact Hamiltonian. Do large-amplitude local-bender eigenstates of the *exact* Hamiltonian actually exist at high E_{vib}? An experimental answer to this existence question requires solution of extraordinarily difficult issues of access, recognition, and input of fit results to a suitable isomerization coordinate-mapping procedure. One of my original motivations for combining multiple-resonance with spectroscopic perturbations was to gain access to special classes of otherwise inaccessible levels. The search for large amplitude local-bender eigenstates in HCCH is more difficult than looking for one needle in a very large haystack. Since a large amplitude local-bender state is localized in a profoundly different region of coordinate space than the vast majority of "ergodic" eigenstates (which are small-amplitude and complex), access to a local-bender state by SEP requires a "local-bender pluck" vibrational level of the S_1 state. Both turning points of the S_1 ν_6 (*cis*-bending) normal mode resemble the half-linear geometry of a local-bender. So we had our local-bender pluck, or did we? Transitions from low vibrational levels of S_0 to $\upsilon_6 \neq 0$ vibrational levels of S_1 are Franck–Condon forbidden.

The HCCH S_1 state has *trans*-bent equilibrium geometry, with a relatively low in-plane barrier to a *cis*-bent conformer. Our only hope for access to $v_6 \neq 0$ local-bender pluck states was to exploit the strong anharmonic interactions between the Franck–Condon bright ν_3 (*trans*-bend) and Franck–Condon dark ν_6 (*cis*-bend) levels caused by the low barrier between *trans*- and *cis*-bent conformers of the S_1 state. My usual out-of-control optimism led me to propose to Adam Steeves and Hans Bechtel that it should be possible to locate and characterize suitable local-bender pluck states by subjecting the S_1 state vibrational structure to only a small amount of deperturbation analysis. We soon found that reality is most unkind.

Undaunted, we called in the Master Deperturbator (a name that Rich Saykally had bestowed on me, and which I now cede to a vastly more deserving spectroscopist): Anthony Merer. Anthony subjected every scrap of spectroscopic information about the HCCH S_1 state to a global analysis. This analysis dwarfed, in complexity and completeness, my long-ago deperturbation of all of the valence states of CO. Soon, Anthony Merer led Adam Steeves, Hans Bechtel, Nami Yamakita, and Soji Tsuchiya on an exhaustive exploration of the S_1 state. We employed every spectroscopic and analysis trick in our books. I violated my solemn pledge never to do molecular beam or microwave spectroscopy at MIT. By far the most useful tool was simple, supersonic jet, one-laser laser induced fluorescence, because it had a dynamic range of 10^4! We were able to observe the $K_a = 0$, 1, and 2 rotational submanifolds of *every* vibrational level of HCCH, from the zero-point vibration up to near the top of the *trans*–*cis* barrier. The web of anharmonic and Coriolis perturbations was so insidious that the only way to achieve definitive assignments was to observe all of the levels. Approximately 100 journal pages later, we have what is probably the only essentially complete analysis of the vibration–rotation structure of an electronically excited polyatomic molecule. And we know the term energy and vibrational character of several local-bender pluck states.

The completeness of our spectroscopic data set enabled us to engage in the "Xmas Eve Assembly" method of assignment. After fitting our spectroscopic observations into what we knew was a *complete* rotation-vibration manifold, there were spectra of a few vibrational levels left over! These levels had to belong to the previously unobserved HCCH S_1 *cis*-bent conformer. Josh Baraban, with help from Professor John Stanton, has assembled convincing theoretical evidence in support of these assignments.

With candidates for local-bender pluck S_1 levels in hand, the problem of *access* was solved. But we had to ask how we might *recognize* a large amplitude S_0 local-bender level. Adam Steeves had an idea of a lifetime that a large change in nuclear geometry must be reported on by changes in electronic properties, such as electric dipole moment or hyperfine structure. Electronic properties are *embedded reporters* on large amplitude motions! But electronic properties generally give rise to very small energy splittings. How would one search for a needle in a haystack? You make the needle very large!

We needed a technique capable of searching over a huge number of vibrational levels yet capable of revealing diagnostically significant small splittings. Bring on Brooks Pate's chirped-pulse (CP) microwave spectroscopy! Owing to its center of inversion, S_0 acetylene has no pure rotation spectrum. But vibrational local-bender states come in degenerate g,u pairs. Only the local-bender states of acetylene will exhibit $J = 0-1$ pure rotation-like vibration−rotation transitions in the millimeter wave region. Our collaborator, Professor Hua Guo, has confirmed this by full dimensional *ab initio* calculations.

The CP technique permits us to search, in a single 1 μs pulse, a 10 GHz wide region with 100 kHz resolution. CP-mm-wave spectroscopy is the ideal complement to 10 Hz pulsed supersonic jets and Nd:YAG pumped tunable lasers. Adam Steeves, Barratt Park, and Kirill Kuyanov have already demonstrated that CPmm spectroscopy is ~1000 times more sensitive than our previously optimized single-resolution-element sequential scan schemes. Kirill is building a slit-jet CP spectrometer that we expect will increase this sensitivity by another factor of 1000. I am in my second scientific childhood!

HCP. The only molecule in which we have actually observed bond-breaking isomerization is HCP. Bhavani Rajaram, Haruki Ishikawa, and I thank Kevin Lehmann for lending HCP to us, but we never returned it. Like acetylene, HCP forms bend-stretch vibrational polyads. As the amplitude of the bend increases and the H starts to shift its bonding interaction from the C to the P atom, a qualitative change in the polyad structure occurs. The "isomerization states", the states organized along the isomerization path, suddenly fall far below the low energy region of each polyad. This led Mike Kellman and Reinhard Schinke to ideas about bifuractions in the classical dynamics and statistical signatures in the quantum adjacent level spacing distribution. HCP provided many of the key ideas that have guided our quest for signatures of isomerization in HCCH.

Laboratoire Photophysique Moléculaire (LPPM) and the Perturbation Books. My first visit to LPPM was in the summer of 1975. I had begun a perturbation-centered correspondence with Hélène Lefebvre-Brion while I was a graduate student. I was an experimentalist curious about unconventional electronic properties and she was a theorist who wanted to use theory in unconventional ways to interpret experimental results. Predestination seemed to be at work.

At about this time, an editor from Academic Press had the idea that someone should write a book about perturbations. She contacted Dick Zare for his advice about suitable victims. Dick suggested me! But I was in my first year as an Assistant Professor at MIT and the last thing a sane experimentalist person would do is to agree to write a mostly theoretical book. I had the brilliant idea to suggest Hélène Lefebvre-Brion. Who could be better qualified? But Hélène's prompt and mostly positive response was that her English was not good enough to be sole author of a technical monograph in English. So she suggested that the ideal arrangement would be for me to coauthor the book with her. So I was had!

The original idea was that I would spend a few months during summers at LPPM and the book would be finished in 1977. By the end of 1977 I had many pages of mostly illegible notes, some hopelessly bad ideas for framing the subject, and no text at all. I knew that, if I were to achieve tenure at MIT, my first sabbatical would occur in 1981. Surely, if I were to spend half of it in Orsay, Hélène and I would be able to finish the book. Wrong again! Out of desperation, we resorted to an ingenious book-production plan where Hélène would write rough text for *all topics*, those she knew a lot about and those about which she knew next to nothing. I would polish the English and expand the explanations for the former and, driven by terror and confusion, completely reconstruct the latter. All of this was done on opposite sides of the Atlantic. Hélène was *usually* very gracious and accepted most of my revisions and new material with enthusiasm.

I did all of my book-writing work in intense 1−2 week bursts in non-MIT locations, mostly with Stew Novick at Wesleyan and Rich Saykally at Berkeley. I worked 80-hour weeks, just as I had as a graduate student. It gave me special pleasure to show that "I still had it", at least in short bursts. "Perturbations in the Spectra of Diatomic Molecules" was finished in 1984, seven years after the promised 1977 completion.

I made many visits to LPPM. At the top of my list of favorite perturbation-related papers was one by Marcel Horani, Joelle Rostas, and Hélène Lefebvre-Brion. Joelle was one of the first people I met at LPPM. I am fortunate to include Joelle and her husband François among my closest friends. We have also collaborated many times on some of our favorite molecules: CO, MgO, and CaF. Marcel Horani holds a special place in my memory, because he was the only one at LPPM who was willing to have lunch with me every day at the campus cafeteria and *speak English*! Dolores Gauyacq and Guy Taieb have also been great friends and collaborators.

At an October 1997 symposium in honor of Hélène Lefebvre-Brion's retirement from her CNRS research appointment, I suggested to her that it would be a good idea to make some minor corrections and revisions of our then out of print book. My original idea was that it would take about one year to produce a revised and perhaps 10% expanded edition. The publisher (Academic Press had become a part of Elsevier) wanted at least a 30% expansion and some hot new topics. I had wanted to make the dynamical aspects of spectroscopic perturbations more explicit and I also wanted to show how to extend the methods, models, and concepts to small polyatomic molecules. A 50% expanded new book, "The Spectra and Dynamics of Diatomic Molecules", was promised for 1998 and finished in 2004.

The perturbation books were unique in that they were intended as user's guides for both experimentalists and theorists. Hélène and I were never hesitant to express our often strong opinions about the quality or appropriateness of methods and goals.

Rydberg States. Ever since my Ph.D. thesis study of the CO valence state perturbations, I was fascinated by unexpected relationships between molecular properties. I believed that I had discovered that *all* of the electronic perturbation matrix elements between the states of the $\sigma\pi^*$ (2 states) and $\pi^3\pi^*$ (6 states) configurations were described by only two electronic parameters.

Atomic-ion-in-molecule ligand field theory reproduced the electronic properties of all of the low-lying electronic states of the alkaline earth monohalides and all of the valence states of the rare earth monoxides and monohalides. Despite my passion for finding few-parameter models that explain many properties of many states, I was profoundly uninterested in Rydberg states. Arrogantly and naively, as was my habit, I had dismissed Rydberg states as an infinite collection of cookie-cutter-identical states. See one, you have seen them all! I had even tortured my graduate quantum mechanics class with the brilliance of Robert Mulliken's characterization of the series-invariant nature of the inner part of Rydberg orbitals: "ontogeny recapitulates phylogeny". I had heard of multichannel quantum defect theory (MQDT), but I believed that quantum defects were merely fudge factors. I could not have been more deluded.

Members of my group (Peter Bernath, Steve Rice, Jim Murphy, Nicole Harris, John Berg, Jeff Norman, Zygmunt Jakubek (BaF), Jason Clevenger (CaCl), Chris Gittins, Serhan Altunata, Bryan Wong, Steve Coy, Vladimir Petrovic, and Jeff Kay) and I have been studying CaF since 1972, seven years longer than my beloved acetylene. After many years of patient effort, Christian Jungen finally convinced me that the closed shell nature and extreme polarity of the $Ca^{2+}F^{-}$ ion-core made CaF an ideal test case for MQDT. Chris Gittins and Jeff Kay assembled a huge CaF data set and then, using Christian Jungen's MQDT fit computer program and his dazzling insights, we assembled a *complete* representation of the electronic structure of CaF: all states and all dynamical processes. The (mostly) core-penetrating s∼p∼d∼f series (four Σ, three Π, two Δ, and one Φ) form a series of supercomplexes that is represented by an internuclear-distance- and energy-dependent quantum defect matrix (20 diagonal and off-diagonal quantum defect matrix elements, 20 d/dR matrix elements, and 20 d/dE matrix elements). The core nonpenetrating f, g, h, i, ... states are represented by a long-range model. In what is essentially inside-out ligand field theory, the Rydberg electron samples the multipole moments and polarizability of the CaF^{+} ion-core. The Rydberg states of CaF exhibit an interconnectedness of parameters and properties that dwarfs anything my research group has observed in any other molecule, including acetylene.

CaO. The electronic structure of CaO combines that of the Rydberg electron on CaF with the O^{-}-localized core-hole of the alkali oxides. The two open-shell structures give rise to legendary complexity in the spectrum, but only apparent complexity of electronic structure, owing to the negligible spatial overlap between the Rydberg electron and the core-hole. The two open-shell electronic structures are additive and the resulting electronic states are multiplicative (the six-fold degeneracy of the ^2P-hole on O^{-} converts every state of CaF into six electronic states of CaO). Many group members contributed to the analysis of the fiendish spectrum of CaO (Rick Gottscho and Ron Marks analyzed the "Gottscho−Marks bands", Ron Soltz, Hartmut Schweda, Jeff Norman, Keith Cross), but the final victory belonged to Dave Baldwin. The simplicity of the electronic structure model permitted us (Ernest "Foss" Friedman-Hill and Dave Baldwin) to extract the electron affinity of O^{-} from the relative energies of the deperturbed $Ca^{2+}O^{2-}$ and $Ca^{+}O^{-}$ potential energy curves, which gave rise to my only paper in the *Journal of the American Chemical Society*. I like to think that this was my first venture into patterns of patterns: "meta-patterns".

Li Li. Li Li was among the first group of Chinese scholars to make extended research visits to the United States after the Cultural Revolution. She had been working in a laboratory in Qinghai (perhaps a one week train ride to Beijing, where her husband and daughter lived) in which even the most routine research components were unavailable. When she arrived in 1982 for her first of many visits to my group, she was beside herself with excitement at the number of lasers and opportunities for spectroscopy.

I think I have written more papers with Li Li than with anyone else. She has an amazing instinct for seeing something special in a spectrum and then presenting that thing in a pictorial way that exactly fits my spectroscopic mental receiver. Mostly Li Li worked on Na−Na. She had the idea of doing perturbation facilitated OODR in a heat pipe oven. She exploited well-known $A^1\Sigma^+_u \sim b^3\Pi_u$ perturbations in order to explore the completely unknown high lying triplet states by OODR fluorescence excitation spectroscopy and all of the mostly unknown bound vibration−rotation levels and continuum of the $a^3\Sigma^+_u$ state. She exploited extremely weak singlet∼triplet perturbations to examine collisional transfer through doorway states. She observed and made simple models to explain the hyperfine structure of triplet states. We reorganized the triplet Rydberg series of Na_2, replacing the inappropriate separated-atom state names by united atom state names, which made the core-penetrating/nonpenetrating nature of the Rydberg series usefully transparent.

Li Li sent many of her students and colleagues for research visits to my laboratory. Her own visits to Bill Stwalley's and Marjatta Lyyra's laboratories created explosions of new projects. I remember spending a week visiting Li Li at Tsinghua University. During that week she presented me with a series of spectroscopic gems that quickly resulted in five papers!

I am a few years older than Li Li, but when I visited her in China, she was my "Chinese mother".

Diatomic Molecules. At the core of my spectroscopic identity one finds mostly diatomic molecules. One never forgets one's first love.

Mike Dulick announced to me that he was going to spectroscopically characterize *all* 14 of the rare earth monoxides. These were exceptionally difficult molecules to make sense of, because all of the very numerous low-lying electronic states had almost the same rotational constant, the electronic states did not arrange themselves into the usual $^{2S+1}\Lambda$ multiplet state patterns, and some initially unknown selection rule prevented linking partial Ω-submanifolds into one global level diagram. Mike's spectra of CeO and PrO revealed the level patterns, hidden quantum numbers, and selection rules that demonstrated how a simple atomic-ion-in-molecule form of ligand field theory explains the electronic structure of all rare earth diatomic monoxides and monohalides (many thousands of electronic states). The rare earth project started by Mike Dulick (funded by Ed Murad of the Air Force Geophysics Lab) has provided opportunities for spectroscopic heroism to Professor Colan Linton (a frequent visitor from the University of New Brunswick), Steve McDonald, Harold Schall, and postdoc Leonid Kaledin.

Mike McCarthy, Mingguang Li, and Hideto Kanamori (visiting professor from Tokyo Institute of Technology) developed a sputter source for molecules that could not easily be produced in the venerable Broida oven (mostly transition metal hydrides), invented FM-MRS (frequency modulation-magnetic rotation spectroscopy), a double-null form of spectroscopy ideally suited for ultrasensitive high resolution spectroscopy of molecules in high temperature environments, and a unique form of sub-Doppler spectroscopy (sideband optical−optical double resonance Zeeman spectroscopy: SOODRZ) capable of automatically assigning low-*J* lines of complicated spectra. Jeff Gray applied our supermultiplet model to the Zeeman spectra of NiH. A great deal of fun was had by all, especially by me!

Richard Barrow and Albin Lagerqvist. I had many wonderful visits with Richard Barrow and Albin Lagerqvist. I was one of the few young laser spectroscopists who appreciated the elegance, difficulty, and honesty of their diatomic molecule spectroscopy, especially the perturbations. Richard told me a story of how he and Albin met at the beginning of their careers and became great and unconventional friends. Albin's English was good, but idiosyncratic. One of his favorite phrases was that he had been served "an elegant sufficiency" of food. Richard told me about how they worked together assigning the spectra of CS and BaO. They would eat and drink a lot, then go up onto the roof of Richard's apartment building in Oxford and assign the perturbations in the light of the moon. I never found an error or even a misprint in their published tables.

I had one visit to Stockholm when both Richard and Albin were present and interacting with each other in the special way that they had cultivated over their 50-year friendship. They were both rather formal in speech and bearing, but humor was always lurking in every gesture and phrase of their spoken interactions. Albin would serve us very strong coffee and then he would put a few tablets of artificial sweetener into his and then ask if we would also like some arsenic in our coffee.

Some Stories. The atmosphere and character of a research group is profoundly influenced by the stories that are passed from one generation to the next. Here are some of my favorites.

My Ph.D. thesis was 388 pages long, but that includes 250 pages of computer output. Not to be outdone, Rick Gottscho's thesis (the second from my research group) was 627 pages, including 118 pages of computer output. Pat Vaccaro spent a full year on his magnum opus, 510 pages *without any computer output*. Not only did Pat beat Rick by a nose, but David Pratt wrote a review of Pat's thesis concluding with "Every Physical Chemist should read this thesis".

Half of my research space was a 40' × 40' laboratory room in the old Harrison Spectroscopy Laboratory. The temperature in that lab was a steady 55° F, year round. Parkas rather than lab coats were the typical working attire, partly to keep warm and partly as protection from the large population of mosquitoes (that had colonized the insides of our laser tables). Almost every optical and electronic component had a tag "touch this and you die" courtesy of Evan Abramson. Evan is also famous for having spent several months living in the laser lab (he did not like his apartment), sleeping in an old laser crate. One day, when Michael Feld was taking a visitor on a tour of the Spec Lab, they encountered Evan fast asleep in his crate. Michael was not amused, but Dam Imre was hysterical with laughter. Evan's first meeting with my then girlfriend Susan Geller was also memorable. At Susan's first dinner with my research group, Evan asked me (meant as a compliment) "Where did you find this chickie?" This innocent remark had an emotional resonance of only slightly weaker impact on Susan than the three "young lady" remarks had on my mother.

Carter Kittrell, the group's "super-postdoc", played a central role in moving my research into the pulsed laser world. He was an amazing experimentalist. I remember how he proved that the resonator of our pressure-scanned Molectron dye laser was flexing as we changed the SF_6 gas pressure. He built an etalon with one mirror glued to the top of the resonator and another mirror mounted to the laser table on a magnetic chuck. As we changed the pressure in the resonator he observed the motion of fringes from his etalon. From that time on, we had one of the most stable pressure-scanned Nd:YAG pumped dye lasers in the world. Another of Carter's special characteristics was that he was a world-class packrat. One could not walk down any corridor at MIT without seeing a pile of discarded junk outside a lab door with a sign "Save CK" on it. One lab's junk is another lab's treasure. But on the night of October 20, 1996 there was a rainstorm of the century in Cambridge. MIT's infamous steam tunnels filled with water, water penetrated under the asbestos cladding of the steam pipes, and traveled to the termination of the steam pipes in my lab. The lab filled with mineral-laden steam, and everything optical, mechanical, and electronic was destroyed. There is no more efficient way of solving the problem of obsolete laboratory equipment than an involuntary steam bath.

Pat Vaccaro used to work in the lab well past midnight almost every night. There is some folklore at MIT that the best signals are achieved after the subways stop running. It is my opinion that Pat liked working alone. One night around midnight a stranger opened the door to the lab expecting to find the lab unpopulated and a good source of resellable objects. It is difficult to imagine who was more surprised, Pat or the intruder. Pat yelled, the intruder bolted, with Pat in hot pursuit. Some versions of this story have Pat merely chasing and others have him catching the culprit. Pat informed me that he actually did catch the culprit in a once-in-a-lifetime flying tackle. The nickname for Pat that I proposed, "Pat-man", seemed appropriate, but never caught on. Pat tells me that this was not the first lab intruder he had encountered. He reported the first one to Campus Police and they showed him some photos for possible identification, but they pulled two photos back saying, it could not be this one, because he had been injured in a shoot-out the previous week, nor this other one, because he was already in prison for manslaughter. The lesson is, never attempt to get between Pat Vaccaro and his lasers.

Daisy Chawla was an extremely cheerful and enthusiastic student with an exceptionally "can-do" attitude. It was impossible for her not to participate in any adjustment of her apparatus, even if it was an extremely intricate multistep alignment process. Serge Churassy and Li Li would have had their own stories to tell about their collaborations with Daisy. Here is a story of Daisy and the Spectra Physics Ar^+ laser repairman. A capacitor needed replacement and Daisy volunteered to do it. But she installed it with the wrong polarity. When the power was turned on, the capacitor exploded like a grenade, fragments barely missing the head of the serviceman. But the noise caused temporary deafness and a coffee-times-1000 level of whole-body anxiety—cured by going outside the lab to smoke cigarettes with Mike Dulick.

For reasons I have never fully understood, people enjoyed teasing Stewart Cameron. He was doing an experiment on CO and kept a large quartz cell filled with CO on his laser table. Jeff Gray found an enormous dead cockroach and placed it next to Stewart's CO cell. Stewart was not amused. Stewart's experiments involved a lot of expensive polarizing optics and opto-electronic components. He kept these and many other treasures in a locked cabinet (with a very large and prominently displayed padlock). What he did not realize is that the doors to the cabinet could easily be removed at their hinges without disturbing the lock. So, every night, the components in the cabinet were massively rearranged. The group did everything in their power to drive Stewart crazy, with minimal success.

When I was on sabbatical in France I used to send gargoyle postcards to the group asking about progress in the lab. Twenty-five years later, on a visit to France, Vladimir Petrovic sent gargoyle postcards to the group, but I had no clue why I thought that this was so clever.

In 1977 I visited several European spectroscopy laboratories. On my first visit to Albin Lagerqvist's lab on Vanadisvägen in Stockholm, Albin met me at the airport, drove me to the Physics Institute, showed me the room that would be my office for a week, listed all the people in his group and their projects on the blackboard, expressed his hope that I would have a good week, closed the door, and left me alone. There was also a guest room in the Physics Institute, and that is where I slept each night in an otherwise unpopulated building. I discovered that there was a room called "Café Planck", where members of the Lagerqvist group would have coffee together. I had a wonderful week, but I was stunned by the extreme respect for my privacy. I traveled directly from Stockholm to Nice, where I was the guest of Professor René Stringat. In Nice I was given a room in one of the most off-scale hotels I had ever seen. I was taken to one fantastic meal after another. It seemed as though they were trying to kill me with food, especially cheese and wine, and kindness. But I kept wondering when we would have some time to talk about molecules. Apparently, there are two very different styles of spectroscopy in Europe! After my visit to Nice, I visited Lyon (Jean d'Incan and Roger Bacis), where the balance between molecules and superb food was right where I wanted it to be.

The first year I taught Freshman Chemistry (400 students in room 10-250), I kept accidentally kicking a wastebasket that was cleverly located at the front of the room near the blackboard. It made me look like a klutz and the class was becoming more interested in when I would kick the can than in what I was writing on the board. Eventually after I kicked it for the last time, I picked up the offending wastebasket and threw it out the main door of the lecture room, to the best round of applause that I have ever received. The class presented me with a certificate "The Trash Can" dated 12/13/91, which I have proudly displayed in my office ever since.

Perhaps stimulated by my tales of the Klemperer group's "George R. Tomasevich Award for Experimental Elegance", my group created a similar award, the "Accidental Spectroscopist". Zygmunt Jakubek was cited for twice attempting to use an I_2 gas cell to calibrate the frequency of a laser beam prevented by a well-situated beam stop from entering the I_2 cell. I received a citation, the meaning of which remains unclear to me: "Bob is cited for his contribution to the battle for the introduction of gratuitous complexity to the spectrum of calcium monochloride. The details are too complex to explain in words but involve the 'Fano Effect'".

It is only a diatomic molecule. How evil can a diatomic molecule be? EuO (Steve McDonald) and ZnO (Jason Clevenger and Christina Hahn) have soundly defeated us. EuO is evil. ZnO is only mean. Often, the main way poor, helpless molecules take revenge on all-powerful *laser* spectroscopists is by bait and switch. A huge effort is invested in recording and assigning what can *only* be a spectrum of a Ph.D.-granting molecule only to discover the spectrum is a well-known transition of a less interesting molecule. Mike Dulick was victimized twice by CN, to Richard Barrow's delight, attempting to observe the spectrum of CeO by vaporizing Ce from a carbon crucible. Jeff Gray and John Gawlik discovered that every line of their new spectrum of NiO exactly coincided with a line from the First Positive band system of N_2. Mike McCarthy was chagrined to discover that many new spectra of transition metal monohydrides were actually atomic lines. Foss Hill's act of alchemy was to see his spectrum of PdO become CuO. David Jonas, a charter member of the *not-diatomics* subgroup, suffered the ultimate humiliation when he discovered that his spectrum of HCN was actually NO. Adam Steeves and Hans Bechtel, two of the greatest experimentalists I have known, spent several months recording beautiful C_2 spectra, convinced that they had finally found the S_1-S_0 electronic transition of HNC.

Susan Geller. In early 1983 Kate Kirby, at the instigation of Hélène Lefebvre-Brion, arranged for me to have a blind date with Susan Geller. On our first date we went to an American Repertory Theater performance of "Waiting for Godot". What a strange choice! At dinner before the play we clicked our water classes together and said, in unison, "to beginnings". We continue to repeat this toast at all of our birthday and anniversary dinners.

When I met Susan, I had been living alone in a seventh floor apartment at 60 Brattle Street in Harvard Square for 10 years (previously occupied for 13 years by Irwin Oppenheim). I loved living in Cambridge and thought I would never move out of that apartment, move out of Cambridge, or move into a house with a yard and garden. It is amazing to me how quickly Susan and I ended up in a house in Belmont. I love weeding (it fits the compulsive nature of an order-seeking spectroscopist), growing tomatoes, and watching dogwoods bloom in the spring. One of my favorite things is cooking dinners for Susan.

Susan and I go to a movie almost every week with her best friend from Radcliffe, Amy Hiatt. We continue to go to plays at the ART, the Shakespeare Festival in Ashland, Oregon with my brother Jay and his wife Debbie Dwyer, and an eclectic selection of concerts (always including the maximum number of string quartets) in the Boston Celebrity Series. It is amazing to me that Susan and I seem to have diverse yet similar tastes in music. And we like to eat, especially in France!

Peter M. Giunta. Peter has been my Administrative Assistant for 20 years! He decided soon after he began working for me that it would be a good idea to type all of my MIT course lecture notes. Anyone who has seen my handwriting knows how appropriate this is. Now all of the MIT Chemistry faculty hand out typed lecture notes. Then, out of the blue, Peter informed me that he wanted to learn LaTeX and would use my then out of print book "Perturbations in the Spectra of Diatomic Molecules" as a training vehicle. This led me to suggest to Hélène Lefebvre-Brion, at her retirement celebration, that we begin work on a corrected and expanded version of our book, which eventually became "The Spectra and Dynamics of Diatomic Molecules". That book would never have existed without Peter. Peter has catalyzed many of the good things that have occurred in my research group at MIT for the last 20 years.

Ever since that fateful Halloween almost 60 years ago when I wore the professor costume designed by my mother, I never had a "Plan B". I could not have guessed how much fun being educated by my students, colleagues, and collaborators would be. 35 wonderful years at MIT have gone by far too fast. How do I begin to thank all of the people (and small molecules) that have given me so much pleasure and excitement? I owe Pat Vaccaro and David Jonas special thanks for putting together this amazing special issue of *The Journal of Physical Chemistry*!

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JP909444E