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Autobiography of George C. Schatz

I was born in Watertown, New York, April 14, 1949. My parents, George and Dorothy, had been the children of immigrants who came to the US in the late 1800s and early 1900s. My father was from an ethnic German ("Saxon") family that lived in Transylvania (now part of Romania, but Hungary at the time they left) and my mother's family came from England and Ireland, stopping in Canada before coming to the U.S. Watertown is at the eastern end of Lake Ontario, in an area of great natural beauty but harsh winters, and my parents lived on a small farm near Sackets Harbor, a town of about 1000 people on the shore of Lake Ontario. Since I went to school in Sackets, I always consider this to be my home town, but in reality I grew up in the country (a place called East Hounsfield) in which my parents tried to raise cows, horses, sheep, chickens and vegetables while maintaining jobs a few miles away in Watertown. My father moved from working in a factory to being a hospital maintenance engineer during my childhood, while my mother was always an elementary school teacher. My father learned how to fix radios and televisions in the Navy during the war, and he kept this up as a part-time job during my youth, and occasionally got my brother and me to help out. My father's technical background was only practical, but he enjoyed doing amateur science, particularly astronomy. Meanwhile my mother was a truly dedicated elementary school teacher, having started teaching in a one-room school. She had gone to college (Oswego State), and she had a lot to do with my subsequent career, and the careers of my siblings, Bill, Helen and Dorothy, particularly with respect to teaching. Because I had a rural upbringing, I have fond memories of a number of activities that farm kids would be familiar with, including 4-H club meetings, county fair exhibits, milking the cows at 6 a.m., square dances, summer camp, skiing on the hills out back, community parties and baling hay. However, my parents gradually gave up farming as I got older, and indeed farming was a dying occupation throughout the region where I grew up and today most of the traditional farms are gone. As a result, my high school days focused on academics, sports (football, basketball, track, skiing), and music.

The public school in Sackets Harbor was very tiny by modern standards, with only around 40 students in my senior class, and all classes K−12 housed in a single building. Many of the students who graduated with me had been classmates since kindergarten, and one of my kindergarten classmates, Margaret Kimmett, became my wife. We were good friends throughout our school years, but we only dated once in high school, when we went to the prom in 10th grade. Our friendship continued, but it took until the end of college for the second date, and then the middle of graduate school for the third. However, after that things warmed up a lot, and we were married just before I finished graduate school. Margaret was always the literary expert when I was in high school, and later she completed a MLS degree at SUNY Albany. She has subsequently worked in public libraries.

Although I did not get much inspiration to pursue a scientific career in Sackets Harbor, there was plenty of opportunity to learn valuable life skills, and I learned a lot by participating in high school football, basketball, and music programs, particularly about the value of team work, and about perseverance and using people with a broad range of skills to perform important team functions. My interest in science was largely self-generated. I was always interested in chemicals that were in common use around the farm, and later I got a summer job managing a 4-H camp where I worked with water purification, sewage treatment, mosquito abatement, and even pyrotechnics for camp shows.

I did not know any scientists as a child, so it was with some trepidation that I decided as a high school junior to apply to college with the intention of becoming a scientist. I applied to several nearby universities, and I was particularly impressed by a visit to Clarkson University, which was about 75 miles away, so that is where I went. I was more familiar with chemistry than the other sciences due to my farm experiences, so chemistry became my major. In the end, both Clarkson and majoring in chemistry were good choices for me. The faculty at Clarkson were friendly, so I was able to get involved in a variety of research projects as an undergraduate, and once I saw how this worked I was hooked on a research career. My first research project (with a professor named Frank Goodrich) was concerned with developing a collocation-based method for solving the electronic Schrödinger equation. This did not work out very well as there were important aliasing errors that were not understood at the time; however, along the way I wrote a code for doing Hartree−Fock calculations for atoms, and this experience has guided my work in electronic structure theory. I also learned a lot about colloid chemistry at Clarkson (which was and still is a research specialty of the institution); however, I did not do research in this field. At the beginning of my senior year, I spent a semester at Argonne National Laboratory (thanks to encouragement from an organic chemistry professor, Richard Partch), where I did an experimental project (and wrote my first paper) with Juan McMillan concerned with EPR measurements on transition metal complexes. Argonne is also where I took my first course in scattering theory, and I started to learn about molecular quantum mechanics. While at Argonne, I attended a seminar given by Richard Feynman, the famous Caltech physicist. Although I was not particularly interested in the subject of his talk, particle physics, I was impressed by Feynman's fresh approach to science, and his infectious enthusiasm for discovery. This got me interested in graduate school at Caltech. So after returning to Clarkson, I filled out applications to several graduate schools, and without ever having visited the west coast I decided to go to Caltech.

One other important event occurred before I headed to Pasadena. In the summer after I graduated from Clarkson, I did research at Princeton with Myron Kaufman (an experimental physical chemist who is now at Emory University). There we studied several chemiluminescent reactions involving atomic fluorine. The experiments proved quite interesting, and ultimately a number of previously unknown reactions were identified, leading to my second paper. In addition, this research project got me thinking about the fundamentals of chemical reactions, thereby opening the door to what would become the subject of my Ph.D. work. In addition, I became familiar with Aron Kuppermann's research at Caltech, as this was somewhat related to my work with Kaufman.

In August 1971, I loaded up my car back at the farm in Sackets Harbor and drove to Pasadena to start graduate school. This was a huge transition for me in many ways, but over the course of the next four years I found a lot to like in Caltech. Among other things there were basically no required courses,

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so this freed me to take whatever I wanted. I loaded up heavily on physics courses, including full year courses in classical mechanics and classical electrodynamics that were enormously useful later on. In addition, I was able to take Feynman's graduate quantum course, which provided insights to the inner meaning of quantum mechanics, and I got to know Feynman himself, and occasionally discussed research projects with him. Courses with the chemistry faculty were also important, leading to long lasting friendships with Bill Goddard, Vince McKoy, Jack Beauchamp, Sunney Chan, and others. And I was able to take Aron Kuppermann's course in scattering theory, which provided a solid background for a lot of the research that I ultimately did, and which provided me with a way to get to know Kuppermann, and to appreciate his approach to science. Aron was warm and welcoming, with intense curiosity and a thorough approach to science. His research included both experimental and theoretical work at that time, which was attractive to me, for although I really wanted to do theory, I felt that the most useful theory would be done in close association with experiment. Thus I was happy to join Aron's group.

The field of reaction dynamics was very active in the early 1970s as a result of a number of advances in both theory and experiment. Of particular importance were molecular beam and laser chemistry experiments, for which major advances were regularly appearing, including the famous Y. T. Lee experiment on $F + D_2$, and Polanyi's infrared chemiluminescence experiments on $F + H_2/D_2$ and other reactions. This provided much stimulation to do theory, and Kuppermann had already made some important advances before I arrived based on the work of Don Truhlar (who was five years ahead of me) and others. In addition, I learned much from Joel Bowman (two years ahead of me) who was working on semiclassical methods for describing reactive scattering, and Michael Baer (postdoc) who was developing a coupled channel code for describing the planar $H + H_2$ (the next step beyond collinear $H + H_2$ which had already been studied by Truhlar and Kuppermann) When I arrived in this "supercharged" environment, Kuppermann and I decided that it was best for me to work on the collinear $F + H_2/D_2$ reaction, as this was the best that theory could hope to do for this system at that time, and there was much excitement about this system due to the experiments described above. In addition, Kuppermann had been himself working on a new coupled channel method that he thought might be able to handle this system. That code was not working when I initially became involved, but it turned out that by correcting a somewhat subtle bug, it was easy for me to get this going, and it quickly led not only to results for $F + H_2/D_2$ but also to a detailed study of resonance effects for collinear $H + H_2$ that proved important in defining the meaning of resonances for this system. This led to several papers, including some wonderful collaborations with Joel Bowman that provided important insight concerning the fundamentals of reaction dynamics.

Michael Baer returned to Israel toward the end of my second year, so I inherited his coplanar $H + H_2$ project. Michael had invested enormous energy in developing a computer code to describe this problem, but the calculated results did not satisfy the requirements of microscopic reversibility well enough, so there was a continuing search for a better method. This problem stumped me for quite a while, but eventually a new approach for expanding the wave function in the reactive region occurred to me, and this immediately led to much better results, culminating in our first paper on the converged coplanar $H + H_2$ reaction dynamics in 1974. This turned out to be a more important breakthrough than I initially realized, as converting the code to do $H + H_2$ in three dimensions was easier than I expected, and the first 3D results were reported in 1975. As a result of a unique arrangement that Aron made with Ambassador College, I was able to have enormous computational resources, so we generated a number of important results very quickly, including a study of quantum resonance effects that proved to be important to the field for a long time.

By the end of my fourth year in graduate school, I was ready to move on. Fortunately, I was able to get a postdoctoral position with John Ross at MIT. So shortly after marrying Margaret in 1975, we moved to Cambridge. I quickly learned that MIT was a completely different institution from Caltech, with different standards for what constituted important theory, and much less emphasis on calculations. This troubled me at first, but eventually I benefited greatly from the new insights I was able to extract not only from Ross but also from John Deutsch, Jim Kinsey, Bob Field, and others. In addition, I had some great office-suite mates during this period, including Peter Wolynes, David Yarkony, Horia Metiu, Shaul Mukamel, and others.

Ross wanted me to do gas-phase reaction dynamics projects, but because the computer facilities at MIT were not up to the task of doing calculations comparable to what I had done at Caltech, I decided to do mostly analytical theory with dynamical models. Fortunately, I had an extensive database of results to model based on my Caltech work, so this quickly led to several papers concerned with Franck−Condon models for determining rovibrational and angular distributions in $H + H_2$, $F + H_2$ and other reactions. In addition, I became interested in developing methods that described reaction dynamics using stochastic approximations that are often used in statistical mechanics to describe the evolution of systems out of equilibrium. This led to a Fokker−Planck approach to describing reactions within the framework of what is sometimes called the classical-path approximation for solving the Schrödinger equation.

While I was busy at MIT, I decided to start applying for academic jobs. Fortunately, Northwestern University was trying to fill a position that was being vacated by the retirement of a quantum chemist, Arthur Frost. Mark Ratner, who had been hired a couple years earlier, played a huge role in luring me to come, and he even visited me at MIT (unusual for the time). So although I had never lived in the Midwest other than a few months at Argonne while in college, in August 1976 Margaret and I moved to Evanston. We have lived in Evanston ever since, and our three sons, Paul, Albert, and Jonathan, were all born there and still live in the area.

Northwestern University has long had an excellent chemistry department, with particular strength in inorganic and materials chemistry. It occurred to me when I arrived that I should figure out how to connect with this expertise while still maintaining activity in the gas-phase reaction dynamics work that I felt still offered important challenges. Thus within a few months I became involved in two areas of research, one concerned with reaction dynamics, where I decided to continue some projects that had been suggested by my work with John Ross on stochastic reaction dynamics, and one concerned with something entirely new, theoretical studies of surface enhanced Raman spectroscopy, in collaboration with Richard Van Duyne. Van Duyne had already been at Northwestern for 5 years when I arrived, and although he initially was considered to be an analytical chemist because of his educational background, he was in my mind pursuing a program of research in physical chemistry that involved inorganic materials. So collaboration with him was an excellent way to be involved in topics that connected me to the Chemistry Department. Moreover, I was extremely fortunate to arrive at Northwestern just as Rick demonstrated the existence of what came to be called Surface Enhanced Raman Spectroscopy (SERS). This involved measurements of the Raman spectra of molecules like pyridine on roughened silver electrodes, where Rick discovered that the intensities were enhanced by a factor of 10^6 compared to pyridine in solution. I did not have much background relevant to this problem (except the colloid background at Clarkson and the one electrodynamics course at Caltech), but I quickly jumped at the opportunity to think about this, and fortunately my first postdoc, Fred King, was interested as well. Fred had a better background in optics theory that I did, so he helped me considerably. A consequence was that we soon wrote two papers that identified some of the fundamental issues (plasmon excitation, local field enhancement, charge transfer effects) involved. Subsequently, I was fortunate to get a joint grant from ONR with my colleagues Peter Stair, Eric Weitz, and Van Duyne, so this provided me the opportunity to develop early versions of theories that are very much alive today. In particular, I worked though electromagnetic theory (analytical theory for spheroids) and made estimates of size-dependent dielectric effects so as to determine the electromagnetic contributions to SERS. In addition, we did time dependent Hartree−Fock calculations for molecular hydrogen adsorbed onto lithium clusters as a model adsorbate/metal cluster system that could be studied with quantum mechanical methods. This theoretical work ultimately led to papers in the 1980−8 period in which we showed how the electromagnetic mechanism of SERS works for a variety of metal particle structures and compositions, we extended these calculations to nonlinear effects such as SHG and surface enhanced hyperRaman spectroscopy, and we examined the interaction of resonant Raman chromophores with SERS active surfaces. While none of this work could be quantitatively compared with experiment, there were a number of qualitative predictions that came out of these studies that have proven correct, and which have provided important guidance to the field. However the lack of quantitative connection to experiment ultimately convinced me to move away from SERS in the late 1980s and concentrate on simpler properties such as nanoparticle extinction spectroscopy and plasmon excitation on gratings. Ultimately, this direction of research transitioned (in the middle 1990s) to the modern era of nanoscience research concerning plasmonic particles that I will describe later.

My interest in reaction dynamics went through several stages, but ultimately two types of activities were pursued, both of which proved to be quite fruitful. One of these was concerned with collision processes involving triatomic and larger molecules using classical mechanics, and the other involved a return to research in quantum reactive scattering. My work on triatomics was started shortly after I arrived at Northwestern. It occurred to me that to describe collisional energy transfer and polyatomic molecule reaction dynamics for triatomic and larger molecules, it would be necessary to develop a quasi-classical theory for doing this in which the collision dynamics would be simulated using classical trajectories but taking the initial conditions from a semiclassical analysis of the vibrational motions. I had not worked on any trajectory-based calculations up to that point, but my Caltech office mate, Joel Bowman, had taught me a lot about this, and in addition there had been important advances in the mid to late 1970s concerning the semiclassical description of polyatomic molecule vibrational states. My first papers on this topic were concerned with collisional energy processes involving triatomic molecules where I was able to borrow from the earlier semiclassical work to define "good" action variables that could be used to define quantum states and energy transfer probabilities. Subsequently, I became interested in studying gas phase chemical reactions with this technology, and this led to a paper (in 1979) concerned with the reaction $O + CS_2 \rightarrow CS + SO$. This was a simple model, but it demonstrated a surprising result, which was that the symmetric stretch mode of the reactant molecule CS_2 could promote reactivity more efficiently than the antisymmetric stretch due to more direct coupling with reaction coordinate motions. Although there were no mode-selective experiments on this reaction (and still have not been), this principle has now been demonstrated experimentally, particularly in work done by Fleming Crim and Dick Zare. After this project I was able to get involved in studies of the $OH + H_2$ reaction which provided an extremely useful example for understanding state specific and mode-specific chemistry, and which has since become the benchmark for reaction dynamics involving four atoms and more. The primary activity in the early years of this work was to develop a quasiclassical trajectory code that could properly describe the vibration/rotation states of the water molecule product of this reaction. Later, we generalized this code so it could treat any atom−triatom or diatom−diatom reaction, and with this we studied the $OH + H_2/H + H_2O$ and $OH + CO/H + CO_2$ systems in great depth. These studies established the basic trends that govern mode specific chemistry, including which vibrational modes are likely to be most active in promoting reaction, and by how much. In addition, the $OH + CO$ reaction provided an excellent example of a reaction that involved intermediate complex formation, but which was not a completely statistical system. Fortunately, there was ongoing interest in experiments for these reactions, so my studies of these reactions proceeded for nearly 15 years, and there were followups with other reactions such as $CN + H_2$, $NH + NO$, $O_2 + O_2$, $Cl + HCN$, and others that continued until quite recently. In addition, we were able to extend this work to study transition state dynamics in the photodissociation of $HBr-CO_2$ complexes.

Generating potential energy surfaces for these reactions was a big challenge. One first needed to use high level electronic structure calculations (multireference CI) to determine potential surfaces with sufficient accuracy to determine meaningful reaction dynamics, and then it was necessary to fit the resulting data at discrete points to analytical functions that smoothly interpolated the results and also were realistic at geometries not contained in the fit. Fortunately, the first of these problems was mostly taken care of by some very talented collaborators who were electronic structure specialists, particularly Larry Harding at Argonne. The second problem was more in my territory, so many of my students and postdocs spent a good deal of time with surface fitting as a precursor to doing dynamics calculations. This was not especially fun, so we (and other research groups around the world) spent a good deal of effort coming up with effective ways to make fitting more accurate and more automated. The problems associated with this remain, but there has been good progress recently with using "direct dynamics" methods to circumvent fitting, as relatively low level electronic structure methods have now been improved to the point where they can be used directly without fitting.

My interest in potential surface development, and more generally in modeling reaction dynamics using high quality methods motivated me to collaborate more and more with the Theoretical Chemistry Group (TCG) at Argonne. Thom Dunning was the TCG group leader in the 1980s, and he helped me obtain a formal appointment at Argonne in 1986 which lasted, with

Al Wagner's help, until 2004. During this time I collaborated not only with Larry Harding but with almost everyone in the TCG. Another visitor at Argonne during the late 70s and early 80s was Joel Bowman, who was at IIT. We had a lot in common, and many interesting collaborations evolved at Argonne until Joel left to take a faculty job at Emory in the middle 1980s.

Northwestern does not have a sabbatical leave policy, but over the years I've managed to take leaves of absence on two occasions, first in 1982 when I spent four months at Berkeley, primarily visiting Bill Miller's group, and second in 1988−9 when I spent seven months at JILA (University of Colorado) as a JILA Fellow (mostly with Steve Leone). Both of these were wonderful experiences for my interest in gas phase chemistry, and both served to play an important role in getting me interested in doing quantum reactive scattering, thus returning to field I had worked in as a graduate student.

The calculations I did in the 1970s at Caltech involved enormous computations, which I knew would not be feasible during my first few years at Northwestern. However, a number of approximate methods were developed by Joel Bowman and others during the late 1970s and 1980s, thus making this type of calculation more feasible. In addition, my visit to Berkeley stimulated another direction of approximate theory with the development of the coupled channel distorted wave (CCDW) method. I subsequently did extensive studies with Jonathan Connor (Manchester) of the $Cl + HCl$ reaction based on this and other methods. In 1986 I decided that the time had come to develop a new code for doing fully converged quantum reactive scattering calculations; this led to the coupled channel hyperspherical (CCH) coordinate method, in which I adapted ideas from Aron Kuppermann, but including concepts for evaluating the scattering wave functions that came from the CCDW work. As fate would have it, this code was completed just as new experiments concerning the photodetachment spectra of transition state precursors in the $ClHCl^-$ and IHI^- molecules appeared from Dan Neumark's laboratory at Berkeley. This coincided with my visit to JILA, so I had time to adapt my codes to do these calculations, thus providing the first detailed analysis of these experiments, showing that the IHI measurements were sensitive to transition state resonances. The JILA period also stimulated me to learn about nonadiabatic processes, first for atom−atom inelastic collisions in collaboration with Leone, but eventually for reactive collisions. In the early 1990s I extended my CCH code to study $Cl + HCl$ and other reactions with multiple coupled potential surfaces, and including for spin−orbit interactions. In addition, I adapted my scattering codes to enable studies of photodissociation dynamics with my postdoc Hua Guo, and to study collisional energy transfer processes in collaboration with George Lendvay, a researcher from Hungary.

All of my scattering theory work to this point had been based on the time-independent scattering methods; however, in the 1990s, my interests in scattering theory also turned to wave packet methods. The most extensive work involved a collaboration with Stephen Gray who had joined the TCG group at Argonne in 1990. With Stephen's codes we managed to study a number of "hard" reactive scattering problems that had always stymied my time-independent CCH method. Thus we were able to study the $O(^1D) + H_2$ reaction, which involves the formation of a stable intermediate H_2O complex, including the coupling to multiple electronic states. This also led to studies of other insertion reactions, including $N(^2D) + H_2$ and $C(^3P) + H_2$. However, my interests in quantum scattering calculations eventually waned, as it seemed that the development of codes to describe reactions with more than three or four atoms was going to be exceedingly difficult. Instead, I became interested in studying larger reactions using quasiclassical methods, and fortunately for me, a project came along that was perfect for this application. Here I collaborated with Tim Minton and Diego Troya in studies of the reactions of hyperthermal atomic oxygen with hydrocarbons. We used direct dynamics methods for this work, initially with semiempirical electronic structure methods, but more recently with density functional theory and MP2 methods. The hyperthermal energies made for relatively short trajectories even for O + propane, and poor accuracy of the semiempirical potential surfaces was a secondary issue, so this approach worked well. More recently, this research has transitioned to studies of reactions at liquid and solid interfaces, where mixed QM/MM methods are needed, but still with the same QM electronic structure approach.

The mid-1990s led to a serious change in my research interests, with much stronger emphasis on nanoscience work. This started with renewed interest in silver and gold nanoparticles thanks largely to Rick Van Duyne, but it has since branched in many different directions, including self-assembly modeling, studies of dip-pen nanolithography, studies of nanomaterial fracture, and work on lipid structures, ion channels, and the properties of DNA. In many respects I owe this dramatic transformation to advice and encouragement from my colleagues Mark Ratner, Rick Van Duyne, and Chad Mirkin, all of whom have been great collaborators, but there have been many other faculty at Northwestern (and occasionally elsewhere) who have contributed, and of course it would not have happened had it not been for some talented students and postdocs.

The major change for me concerning nanoparticle optical properties was the implementation of computer codes for numerically solving Maxwell's equations, typically using methods taken from astrophysics and electrical engineering, for particles of arbitrary shape. This work started with a paper in 1995 in which we used a method known as the Discrete Dipole Approximation (DDA) to study the extinction properties of triangular particles made in the Van Duyne group. This was the first time that we were able to describe anisotropic particle structures made using top-down methods, and although it took four years after that to resolve a number of discrepancies between theory and experiment so that we really thought this approach was working, this was truly the beginning of a quantitative understanding of localized surface plasmon resonances (LSPRs), and of the interactions of LSPRs with molecules adsorbed on the particles. Fortunately for me, this development occurred when a wide variety of methods for using LSPRs for chemical and biological sensing were appearing, so this led to significant funding opportunities that in some cases still continue. In addition, the nanoplasmonics field proved to be extremely rich, with many different particle compositions, structures, dielectric environments, and nanoparticle arrangements that were amenable to experimental study, many different kinds of experimental observables, and from a theoretical perspective, many different types of computational methods that could be harnessed for this work. In addition to nanoparticles, there was interest in continuous metal nanostructures (e.g., hole arrays in metal films) where propagating plasmons (surface plasmon-polaritons) are important, as well as an extensive activity concerned with functionalization of the metal nanoparticles with molecules.

Although the initial experimental collaboration in this work was with Rick Van Duyne, other colleagues at Northwestern also became interested in metal nanostructures, including Chad Mirkin, Teri Odom, Bartosz Grzybowski, Tamar Seideman, and even Mark Ratner. This has created a community that continues to thrive based on the interplay of theory and experiment. I was especially fortunate to be involved with Chad Mirkin's work on the bottom-up synthesis of silver prisms, as this has provided a unique example of plasmon-directed synthesis of metal nanostructures that hints at exciting prospects for the future if we can learn how to take advantage of it. In addition, Chad's development of On-Wire-Lithography and Teri's soft lithography methods provide new platforms for plasmonics studies that have challenged theory in new ways, sometimes requiring new theory methods.

Although much of my early work was based on the DDA method, we became interested in the finite-difference time-domain (FDTD) method thanks to Stephen Gray (who also jumped into the nanoscience field and is now at the Center for Nanoscale Materials at Argonne), and more recently we have developed finite element codes. Most of the early work on nanoplasmonics was concerned with extinction and scattering spectra, but in the last several years there has been much interest in SERS, as well as some interest in nonlinear scattering phenomena and near-field measurements.

The work on metal nanoparticles has also turned to quantum mechanical methods, and in contrast to our primitive work on H_2/Li_n in the early 1980s, this time Lasse Jensen, Christine Aikens, and I were able to study pyridine adsorbed onto small silver and gold clusters using TDDFT methods. In addition, we were able to use TDDFT to study the dependence of the optical spectra of silver clusters on cluster size, providing for the first time a picture of how molecular-like excitations in small clusters evolve into plasmon-like excitations for larger clusters. We also studied the spectra of gold clusters, and in recent work we found that thiolated silver and gold clusters have remarkably rich spectra in which free-electron plasmonic excitations are only a small part of the story. Although my original dream of combining electronic structure theory with electrodynamics is still not fully realized, the work to date has suggested that electronic structure methods will play an important role in understanding many nanoparticle optical properties, and that coupled QM/electrodynamics methods will be useful.

My interest in using electronic structure methods to study the fracture properties of carbon nanotubes and other carbon-based materials started as a result of a conversation in 2001 with Ted Belytschko, who is one of my colleagues in Mechanical Engineering at Northwestern. I had known Ted for over 20 years, but his research was concerned with the mechanical properties of macroscopic objects up to that point so we had not collaborated. However, by 2001 he had progressed to the point where studying molecular scale materials constituted an important challenge to mechanical engineers, so I was pleased to work with him. Subsequently, we managed to get funding for this work, and this led to several fruitful collaborations in which we learned how to use electronic structure methods (along with empirical force field molecular mechanics and continuum mechanics) to evaluate the mechanical properties (stress/strain and fracture) of carbon nanostructures. This work produced a number of new concepts about how to mix quantum mechanics (QM) with molecular mechanics (MM) and continuum mechanics (CM) for strained structures. In addition to nanotubes, we also became interested in nanodiamonds, and in graphene-based materials that are used in polymer composites.

The late 1990s provided me with some very exciting opportunities for new funding concerning the properties of nanomaterials. Initially, this came about as a result of DoD funding through several MURI grants, but eventually Chad Mirkin headed NSEC and CCNE centers at Northwestern that are still in place, and I was also able to join the MRSEC center that has been in place at Northwestern since the 1950s. Some of this new support went into work on plasmonic materials that I described earlier, but in other cases, I was free to start new projects as I wished, but with the constraint that they had to involve collaborations with other people in the center. Some of this work involved collaborations with Chad Mirkin and Mark Ratner. One of these projects was concerned with characterizing the processes that happen in Dip Pen Nanolithgraphy (DPN), which is a scanning probe method developed by Mirkin for depositing molecules on surfaces to make nanoscale patterns. Here we developed lattice models of deposition/diffusion and later I also studied a related process known as nanografting in collaboration with Gang-yu Liu. In addition, we did grand canonical Monte Carlo (GCMC) calculations to characterize the properties of nanoscale water droplets. In another series of projects, Ratner and I collaborated on the development of GCMC methods for characterization of ion transport in cyclic peptide nanotube ion channels. And in another we studied the self-assembly of amphiphilic molecules that form cylindrical micelle structures, developing methods that take us from simple packing models, through coarse-grained (CG) models and to atomistic models. This activity got me interested in self-assembly modeling, which is an ongoing interest that has many facets.

Another direction of research that came out of collaborations with Mirkin was my interest in DNA. In the mid-1990s, Mirkin demonstrated that it was possible to link gold nanoparticles using DNA-linkers, and that the thermal melting properties of the resulting gel-like material were dramatically different from the melting that normally occurs in duplex DNA. This stimulated me to get involved in understanding the structures and thermal properties of DNA. Unfortunately, the time scale of melting in DNA is not compatible with studying this problem using standard molecular dynamics methods, so we spent quite a bit of time in the late 1990s developing a coarse-grained model for DNA. This model did describe melting, but unfortunately it could not describe the nanoparticle aggregates. However, it ultimately provided me with important ideas as to what were the essential physical elements in the DNA-linked materials. Subsequently, we started collaborating with another of my colleagues, SonBinh Nguyen, who learned how to get DNA to link polymers and even to link small molecules, thus providing new platforms for understanding melting. As a result, we developed mean-field theories and statistical mechanical models that include both the phase transition and cooperative melting mechanisms that play a role in the nanoparticle aggregates. In addition, we have learned how to use atomistic and coarse-grained methods for providing a more quantitative picture concerning DNA-linked aggregates of nanoparticles or polymers.

The work on DNA eventually got me into a new collaboration, this one involving another of my colleagues, Fred Lewis, who is an expert at synthesizing DNA hairpins and measuring spectra. Hairpins are in many respects the ideal platform for testing theoretical methods and ideas concerning the structures and optical properties of DNA, as the number of base pairs can be reduced to a small enough number so that high quality modeling with electronic structure theory and molecular dynamics is possible. In addition, hairpins have many of the same properties as larger pieces of DNA, including the presence of melting transitions and complex excited-state photophysics. Our work in this field is still at an early stage, but it has already proven useful for establishing the validity of force fields and molecular dynamics protocols that we use to describe DNA, and for establishing which electronic structure models capture the photophysics.

The soft-materials work also got us involved in a collaboration with another colleague, Tom O'Halloran, concerning the properties of lipids that form complexes with DNA, and using lipids to encapsulate drug molecules for chemotherapy applications. Here we have learned how to use coarse-grained models to make lipid bilayers, liposomes, and other lipid phases, thus extending our earlier work with CG models for DNA, but also borrowing from the work of others. This work is still in progress, but it seems like the CG models provide an excellent way to mimic the effect of lipid molecule structure on membrane structure and dynamics.

My association with *The Journal of Physical Chemistry* began in 1992 when Peter Rossky (Texas) who was then a Senior Editor asked me if I might be interested in replacing him as he had decided to step down. I was already quite familiar with *JPC* (my first publication in *JPC* was in 1972), and in fact I had been a supporter of Mostafa El-Sayed's efforts to rejuvenate the journal after his appointment as Editor-in-Chief in 1980. So I was happy to take over for Peter. Mostafa agreed with the switch, and I served as Senior Editor for the period 1993−2004. This occurred at a time when there were only five Senior Editors, and I was the only editor handling primarily theory papers, so I quickly became familiar with the many theory topics that the journal covers. Over the Mostafa years, I was able to influence the journal in a variety of ways, such as by recommending Special Issues and Feature Articles on theory-related topics, and through Mostafa's choices for additional Senior Editors. In 2004 when Mostafa stepped down, I was honored to be selected as his successor. In many respects my job as Editor-in-Chief has been easy, as Mostafa had already created the excellent journal that we know today, and its dedicated community of editors, authors, and reviewers. Although there have been many changes in the last four years, such as the splitting off of *JPC C* in 2007, the journal community and the overall scope of *JPC* remain unchanged.

As should be clear from this article, I owe many thanks my scientific teachers, collaborators, and especially my colleagues at Northwestern for the development of my research career over the years. In addition, I have had many dedicated students and postdocs, and I am proud that many of these have gone on to productive careers at other institutions. I also want to thank my journal staff (Davine, Jan, Julie, Barb and Vivian) and the Senior Editors and their offices for maintaining the high quality of the journal and its smooth operation. And I especially want to thank my wife Margaret for her love and companionship going all the way back to kindergarten in Sackets Harbor.

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