

## Four great personalities of science: G. N. Lewis, J. Franck, R. S. Mulliken and A. Szent-Györgyi

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**Abstract** - The popular notion of a scientist, and the young student's conception of what abilities lead to a career of high achievement in science, can be quite at variance with the nature and creative abilities of real scientists. The four personalities described here through the author's extensive contacts exhibit extreme contrasts in scientific repertoire, yet each accomplished commanding advances in science with powerful influences on their fields of research. Gilbert Newton Lewis possessed the most powerful mathematical skills of the four, yet is known principally for simple but effective qualitative concepts of chemistry. James Franck established the groundwork for a half-dozen of the most rigorous principles of molecular physics, yet his chief tool was simple logic, using arithmetic comparisons as a tool. Robert S. Mulliken developed the most detailed understanding of the intricacies of molecular structure and spectra, using mathematics more as a descriptive framework rather than for a deductive analysis. Albert Szent-Györgyi possessed a very inexact knowledge of a broad range of the sciences, yet had a knack for making keynote discoveries based on intuition. The driving forces unifying the immensely creative careers of this group of scientists were an intense curiosity and an overpowering desire to understand, with their precise knowledge and working methods playing a secondary role. This suggests that the education of creative young scientists should strive to cultivate pursuit of curiosity and a strong drive for understanding as compared with emphasis on formal comprehensive knowledge.

### INTRODUCTION

When as students of Science, at any stage of our development, we contemplate the great figures who have shaped our discipline, we can be awed by thought of their intellectual achievement and rigor. We may even be led to think, as some laymen do, that there is such a thing as a unique scientist-mentality, and that a certain propensity in common for quantitative, directed, and organized abstract thinking marks the highly achieving scientist. But as soon as we get to know such scientists as working individuals we realize how varied are the personality types, working methods, and intellectual approaches of different accomplished scientists.

I have had the extremely good fortune to have some close association with Gilbert Lewis, Robert Mulliken, James Franck, and Albert Szent-Györgyi -- each a stellar contributor to his respective science. I have always been impressed by the striking contrasts between these four men, and how each, by his own personal style, succeeded in making permanent contributions to his fields of interest. In recounting some personal contacts with them I hope that I succeed in revealing the special characteristics of each of these men.

### GILBERT N. LEWIS – PHYSICAL CHEMIST

#### First contacts

When I came to Berkeley in February 1943, G. N. Lewis was 68 years old and had recently stepped aside as Dean of the College of Chemistry; I had just turned 22. I had come as a graduate student determined to work with Lewis, as he was certainly the best known physical chemistry professor then in the University of California in Berkeley. The Chemistry graduate students were required to select a professor before the end of their first semester and to commence research in that term. I went through the obligatory list of interviews in *pro forma* routine, saving Lewis for the last. My first hour with Lewis was impressive. I must have appeared nervous, for Lewis said: "Now don't think I'm going to embarrass you by asking you a lot of detailed questions about physical chemistry. I would just like to tell you about some of the interesting research which we have been thinking about recently."

Lewis then proceeded to talk with excitement about the phosphorescence studies done with David Lipkin and Theodore Magel, and the absorption and photochemical studies then just being completed with Jacob Bigeleisen, who overlapped with me for the month of May of 1943. Lewis and Calvin had developed earlier a semi-classical theory of light absorption in dye molecules, and Lewis and Bigeleisen had done some elegant polarization studies of x-band and y-band molecular coordinate resolution of the electronic transitions, and related photochemical studies.

#### Phosphorescence and the triplet state

Lewis held a daily research conference with me at 11:00 AM every morning, usually lasting an hour. Each session launched major plans to be carried out that very afternoon and evening. By the end of such an hour, a whole strategy for the day was mapped out. No apparatus was too complicated to be assembled that day, at least in preliminary form. Of course, I had an enormous advantage over other graduate students since Lewis could call on all and any shop men for instant service.

After a month's debacle with dye spectra, Lewis and I both wanted a refreshing change. I suggested phosphorescence had many puzzles to resolve. Lewis agreed, and said, "Have the shop build a phosphoroscope." Lewis then left for a month's vacation. It was just the first day of June 1943, and Bigeleisen had two or three days left in Berkeley. "Jake, what is a phosphoroscope?" I asked. He was furious that I had not used the library. "Look it up!" was his answer. I studied the idea of the Becquerel phosphoroscope, and designed one with parallel discs and had it built the next day. Bigeleisen's parting suggestion to my question, "What shall I study?" was: "Try carbazole, it seems to have a very long-lived blue-violet phosphorescence." This suggestion proved to be worth its weight in gold. In the Lewis inner laboratory, where the old brass-tube and mahogany-boxed Medium Hilger Quartz Spectrograph was housed, was a score of large dessicators containing boric acid and dextrose glass phosphor slabs made with various dyes as solutes (from the work of Lewis, Lipkin, and Magel, 1941). All of these dye "phosphors" exhibited broad, structureless phosphorescence. If I had also studied only dyes, our work may never have developed the direction and significance that it did subsequently.

The carbazole molecule spectrum was astonishing, even to me as a beginning spectroscopist. The spectrum consisted of numerous marvelously detailed "sharp" bands. I don't know how I contained my excitement. I probably showed my spectrographic plates to everyone in the building. Jacob Bigeleisen had left before I got a spectrum. Lewis was away for a month. I was the only graduate student in his laboratory. Having just finished Harvey White's Atomic Spectra and Structure course in the Physics Department, the excitement of spectroscopy was in me. That month I ran through several dozen molecular samples: naphthalene, benzophenone, anthraquinone, benzaldehyde, nitronaphthalene, *et cetera*. Every molecule showed characteristically "sharp" banded structure. All of these were studied in EPA glass at 90 K (we had only liquid air as a coolant). The bands observed were very numerous and highly structured compared with any dye spectra which had been obtained in the laboratory before.

Lewis was delighted and excited by the results when he returned. Our work now turned fully to a broad exploration of the phosphorescence of organic aromatic molecules in rigid glass solution at low temperature. My phosphoroscope, vacuum sublimers, and vacuum stills ran 24 hours a day for the year. At the end of that first summer I was allowed to give a research seminar on the preliminary observations. Admittedly, the presentation was somewhat repetitive in that I tried to show how virtually every molecule we studied showed a characteristic low-temperature phosphorescence -- especially if the molecules were non-fluorescent. The rest of that year was spent in an intensive study of the phosphorescence of over 100 different molecules.

In July of 1944 our work was ready to write up. The procedure used in writing this long paper is worth recording - as it at first startled me. Lewis sat me comfortably in the wide-armed, high-backed wicker chair which greeted all his guests, with pad and paper in my hand, and dictated the paper in perfected flowing English from beginning to end! I was allowed little interruptions here and there, with an occasional slight change of perspective, but on the whole that first paper (Lewis and Kasha, *J. Am. Chem. Soc.*, 66, 2100-2116, Dec. 1944) was already in Lewis' mind. How could he do it, I wondered? Then I realized, that everything had been discussed endlessly in the laboratory, and finally, as Lewis puffed his cigar and paced up and down Gilman Hall, and on the Campus, the perfect phrasing of each thought was developed. And when we sat down together, he was ready!


It is odd in retrospect that PHOSPHORESCENCE AND THE TRIPLET STATE should have to come as late as 1944, as a secure molecular correlation. In atomic spectroscopy and diatomic spectroscopy there was no difficulty with assigning multiplicities and observing forbidden transitions. I believe that one of the retardations on the understanding of polyatomic triplet states was the fact that the techniques used for observation of molecular phosphorescence, solid solutions in rigid glass matrices, impinged on the doped-organic impurity phosphors, and the suspicion carried over that the strange rigid-glass media those Berkeley chemists were using could involve analogous solid state phenomena. It is hard to believe today the general negativism and even hostility which greeted the presentation of this work. It took ten years to overcome early prejudices before the phosphorescence of molecules assigned as the lowest triplet  $\rightarrow$  singlet

transition became universally accepted as it is today. Certainly the Lewis and Kasha study showed that phosphorescence was an intrinsic molecular characteristic, independent of the medium or solvent used. Probably, aside from the novelty of the spectroscopic technique, the lack of comprehension of the rôle of radiationless processes in polyatomic molecules was the other source of diffidence.

In the second year of my work with Lewis, World War II was in its intensive final stages and I was permitted to remain in Berkeley only if I joined a war-time research project. I easily made the timely decision to take on the heavy extra burden in order to continue with Lewis. So in this second year, Lewis and I saw each other each evening and Saturdays, and frequently on Sundays. We averaged six hours a day together in our whole period of two and one-half years of contact. Our first paper was written in July and August 1944 in the evening hours of 7 - 11 PM. Meanwhile, our attention had turned to singlet-triplet absorption studies, in a paper published in 1945. In February 1945 I had completed my Ph.D. degree on the basis of these two researches.

### Gilbert Lewis' research style

At the blackboard during our conferences, at first in our morning sessions, and later in the evening sessions, Lewis' powerful analytical and intuitive scientific prowess shone through. There were two rather detailed mathematical problems that we worked on, one being the derivation of, from the Einstein A and B coefficient relations, the integrated absorption-lifetime equation used in our second paper (we did not know that R. C. Tolman had done it earlier). The other was laying the analytical groundwork for a detailed photomagnetism study which I completed with the overlooking and help of Melvin Calvin (published by Lewis, Calvin, and Kasha in *J. Chem. Phys.* in 1949, after Lewis' death). I was at the blackboard groping with a difficult step a couple of times, and Lewis seized the chalk and quickly showed in a few powerful steps a way to its resolution. Although Lewis had published some highly mathematical papers in his career (on relativity theory with Edwin Bidwell Wilson; statistical mechanics with Joseph Mayer), his later years were spent on rather qualitative experimental studies. But the old ability was very much in evidence, hiding just beneath the surface.

A special trick was used by Lewis when we got boxed into a logical impasse, and it proved to be very effective. It did happen pretty frequently that our series of logical steps in some argument of development carried us seemingly to a stone wall:  $A \rightarrow B \rightarrow C \rightarrow D \rightarrow \parallel$ . Lewis would realize this, and he would say, doing almost a little dance around a semi-circle  as if to look at an object from behind: "Let's look at it *backwards!*" So the argument was dissembled  $D \rightarrow C \rightarrow B \rightarrow A$ , and then suddenly it seemed clear that the steps should have been  $A \rightarrow B \rightarrow C \rightarrow D' \rightarrow E \rightarrow \text{Solution}$ , the blind alley becoming magically revealed by unravelling and reexamining the steps in the logic.

Lewis died of a heart attack working one Saturday afternoon in the Spring of 1946 at his favorite place in the laboratory, the Vacuum Bench. He had just passed his 70th birthday. Lewis loved to vacuum distill liquids from one flask to another, to sublime materials into reaction vessels, to watch color changes as reactions took place. The turning of the stopcocks, freezing samples with liquid-air dewars, warming up a solution with a water bath or even with a match which had just lit another cigar, these were all part of the visual physical chemistry that Lewis enjoyed most. He confessed to me that organic molecules had been his special joy in his later years, and how because he had failed a course in Organic Chemistry at Harvard as a student he had developed a life-time dislike of organic molecules - much to his later regret and chagrin.

Lewis had strong feelings against a researcher becoming too attached to a large instrument or heavy research machinery. Although he admired and made use of heavy instrumentation at times, he felt that someone who developed such instrumentation was inclined to become a slave to and an exponent of the instrument or machine. He preferred to be free to wander among available techniques and to wonder about ideas and not be tied to one technique. For somewhat analogous reasons he was prejudiced against complicated chemical syntheses as an aid to physical-chemical research. He admired researchers who could make molecules, but preferred to leave this to others, so that he could devote himself maximally to developing and testing physico-chemical theories. He believed that if a theory was general enough, there should be ample scope for the physical chemist to select a suitable sample from the chemical storerooms to test it.

### Gilbert Lewis' research previews

Gilbert Lewis' career, rich as it was in permanent contributions to the understanding of physical chemistry, also had a most unorthodox pattern, not noted especially by anyone until now, but one which was typical of Lewis and which had a profound effect on his intellectual fertility and freshness, throughout his whole scientific life. *Lewis previewed a research field in print using his own analysis, without an extensive literature review, before embarking upon research on it.* Most scientists think of reviewing a subject in print at the end of a long study and research period. Lewis told me he thought that approach could stifle originality. If a scientist got an absolutely thorough knowledge of the literature before doing research, he was likely to acquire many of the prejudices and mental blocks of his predecessors.

For example, in the beginning of his career he wrote around 1907 a (*Zeitschrift für Physikalische Chemie*)

paper, "Toward a New System of Chemical Thermodynamics," in which the physical thermodynamic laws were applied to physico-chemical problems. This was the origin of Lewis' concepts of *activity*, *activity coefficients*, *fugacity*, *partial molal free energy*, *et cetera*. Lewis told me that after completing his Ph.D. at Harvard he took a two-year appointment in Manila (as head of the Philippine Bureau of Weights and Measures) - a most unorthodox start for a scientific career - so that he could think for himself. He worked his way through Walther Nernst's *Physikalische Chemie* ("correcting all of the mistakes"), hiked his way around the volcanos and countryside of Luzon, and acquired a lifelong taste for Philippine cigars. Lewis then published his chemical thermodynamics overview paper, and followed this with about 15 years of research on chemical thermodynamics, culminating in the classic text, Lewis and Randall (1923). He never touched thermodynamic research again.

Lewis, indeed, changed research fields abruptly throughout his career, each time prefacing a research period with his characteristic penetrating research-proposal-overview paper.

Lewis' interest in valency theory began with a precocious Master's thesis ("The Electron and the Molecule," 1898). He later speculated on electron-pair bonds and electron octets, and his papers on the chemical bond appeared, with World War I interruptions about 1914, then 1916-18. At a symposium attended with Niels Bohr in 1923, he threw the challenge to the Bohr atom, showing that it could not explain molecular structure. He realized that the intrinsic planarity of the Bohr orbits precluded the explicit spatial geometries conditioned by the electrons in atoms in forming molecules and crystals. His chemical intuition set conditions on valency interactions for which only Quantum Mechanics was able to provide the basis. Other research papers by Lewis include his paper on "The Electronic Theory of Acids and Bases" (*J. Franklin Institute*), preceding experimental work with Glenn Seaborg on the subject. "The Color of Organic Substances" (with Melvin Calvin, 1939) preceded almost a decade of research on the color and photochemistry of dyes. Perhaps the atypical case was Lewis' work on heavy water. Lewis had discovered heavy water just before Harold Urey left Berkeley. Urey, after arduous effort, used the Rydberg isotope shift in the atomic spectrum of hydrogen to prove quickly that a new isotope of hydrogen (deuterium) existed. Lewis then worked furiously to establish his claim to the discovery of heavy water by doing every conceivable measurement. Finally all of this work was reviewed in a comprehensive paper, uncharacteristic of Lewis.

Gilbert Newton Lewis typified the physical chemist of great intuition who was able to conceive of beautifully simple models and concepts to explain complex physico-chemical phenomena. His conceptual contributions have made lasting additions to our knowledge, and his aptness for good nomenclature has enriched our scientific vocabulary, *viz.*, Lewis introduced the term *photon* for light quantum (in 1926!).

Gilbert Lewis once defined Physical Chemistry as encompassing "everything that is interesting." His own career touched virtually every aspect of science, and in each he left his mark. He is justly regarded as one of the key American scientists in (World) history. It would be a great omission not to record the warmth and intellectual curiosity radiated by Lewis' personality. He epitomized the scientist of unlimited imagination, and the joy of working with him was to experience the life of the mind unhindered by pedestrian concerns.

## JAMES FRANCK - ATOMIC AND MOLECULAR PHYSICIST

### Provocative meetings

James Franck was a physicist who thought in terms of very simple physical models and ideas, but who possessed also a capacity for very complex conceptual frameworks. One might suppose that Gilbert Lewis and James Franck would have made very compatible scientists, but the opposite proved to be true. I believe it was in late 1943 that Lewis told me he was expecting Franck for a short visit. Our work was only partially completed at that time, and in an early discussion stage. I knew the name Franck from the Franck and Hertz experiment on critical potentials, but otherwise the name did not have much meaning for me at that stage.

One afternoon a very dignified gentleman in a dark suit appeared at the door of my laboratory, asking in a soft voice with a German accent for Professor Lewis. He was imposing in appearance, his dark eyes and finely shaped facial features making a rather stern impression. I found the deep look in his eyes rather intimidating. I later found this first impression of severity to be wrong. I directed Franck to Lewis' large office at the end of the hall. My status as a beginning graduate student precluded my participation in this meeting of Franck and Lewis.

After about an hour and a half Lewis came into my laboratory and seemed to be in a negative mood. He indicated that Franck and he seemed to be on different wavelengths, and that this discussion had not been fruitful. Lewis shook his head and said, "Franck does not understand chemistry." Much of the influence on Franck at this time had come from the theoretical physicist, Edward Teller and the experimental physicist, Peter Pringsheim. Lewis surrounded himself by chemists with a strong physical-chemical orientation. It is conceivable that with such different origins the two men would not share many views.

On luminescence phenomena, the topic of the day, their backgrounds were quite in contrast.

### **Franck's atomic and molecular physics**

Franck's mode of thinking in terms of very simple physical models certainly led to his three greatest contributions to science. *The Franck and Hertz Experiment* (which led to the Nobel Prize in Physics) was in Franck's native milieu, thinking about elastic and inelastic atomic collisions. I have not read the original papers, but I understand from other reading that the critical potential work was *not* undertaken as it might seem in order to check Bohr's atomic theory of stationary states, but sprang up naturally from Franck's way of thinking about atomic collisions.

*The Franck Principle* for electronic transitions in diatomic molecules was another concept based on an idea from Classical Mechanics: the turning-points of a classical oscillator (pendulum) indicating the instant when an electronic transition would be most probable. This is the content of the Franck Principle, in which the ultra-fast electronic motion is separated from the relatively slower motion of the atomic nuclei. We now know this in quantum-mechanical format as the Franck-Condon Principle, from which we realize a contrasting deduction, that the most probable transitions correspond to the potential curve *minimum* of the initial state (maximum in the Gaussian vibrational eigenfunction), and that vibrational eigenfunction interference phenomena (Condon's "internal diffraction") make the electronic transitions most probable at non-classical configurations of the nuclei. Nevertheless, Franck initiated the thought process, and we call this the Franck-Condon Principle today.

*The Franck-Rabinowitch Solvent Cage* is another typically-Franck, quasi-mechanical idea suggested by models of encounter of atomic or molecular fragments diffusing from or into a cage.

In all of the above, details of chemical or molecular structure -- the exact purview of Lewis' thinking -- are supplanted by the abstract models of the physicist in which particles and forces are involved, apart from specifics of structure.

### **Controversy on the triplet state**

In December 1946, the Physical Chemistry Division of the American Chemical Society held a Symposium in honor of G. N. Lewis. As his last student, I was invited to present a paper on our research. The Lewis Symposium was held in Northwestern University near Chicago, and was published in *Chemical Reviews* in 1947.

As a young Ph.D. who had not yet attended a national meeting, I was thrilled and excited by the invitation. My elation was considerably dampened when a Berkeley student of Calvin's (Sam Aronoff), who was visiting from Franck's laboratory, brought back a personal report. "Franck has almost every line of his Lewis and Kasha reprint underlined in red, and to any passerby who will listen, he says, 'Look at this! Look at that! Ridiculous!'" Aronoff said. "And Mike, at the Lewis Symposium, Franck will speak against your paper, then Robert Livingston the photochemist; then Eugene Rabinowitch the photobiologist; and finally Edward Teller the physicist will deal the *coup de grâce!*"

I was greatly worried by this news and went to see Melvin Calvin for advice. He said, "Don't be too worried. Anyone who does his work well always will know much more than his critics about its nuances. But if you want to get some greater confidence, just read what Franck, Livingston, Rabinowitch, and Teller have been writing and you will find out what their thinking is." I did so, and quickly discovered that Spomer and Teller had written on the improbability of observing molecular singlet-triplet transitions from the radiative point of view, and that Franck and Livingston had written on tautomerism as a possible origin of phosphorescence. My readings generated a number of new ideas as responses to these writings, and were presented in my 1947 Lewis Symposium paper.

The evening before the big first day of the Symposium there was a social-mixer for the participants. Melvin Calvin introduced me to Franck, Livingston, and Rabinowitch. A group of us made a small circle for a couple of hours, with cautious sparring from all quarters. At the end of the evening, Franck said, "Well, it looks as if there is good ammunition on both sides!"

The next morning I was excited, when H. D. Smyth of Princeton introduced me as the first speaker. My permitted 20 minutes flew by. I then sat down next to Calvin, who said, "Well, that sounded very clean, very positive. Now, let's see what happens."

James Franck asked permission to speak. His presence was imposing. He started to speak with commanding authority. Soon I began to make a discovery about Franck's lecturing style. It was very difficult to follow his arguments. There was no audible punctuation or pause as his presentation developed in labyrinthian fashion. My head was swimming. I leaned over and whispered to Calvin, "I can't follow him; I won't be able to answer him." He said, "Don't worry, probably no one else can."

Franck continued for almost 15 minutes. When he was finished, he asked permission to call on Robert

Livingston to talk about evidence against the triplet state from the point of view of photochemistry. Livingston got up and in a series of slides showed that in the photochemical reactions he had studied there was no evidence for an intervening metastable state between the ground state and the first singlet excited state. (About 15 years later Livingston wrote me a note saying that he would now join other researchers in using triplet state nomenclature, but he personally still did not believe that a metastable state of a molecule could be a triplet state.)

Franck then asked permission to call upon Eugene Rabinowitch, who gave a discussion of photobiological processes including photosynthesis, indicating the absence of any evidence for triplet state excitation in bio-molecules. His book on photosynthesis published in 1956 bears a very strong critique of the Lewis and Kasha paper.

Finally, Franck called upon Edward Teller, saying, "Professor Teller will show why it is impossible to consider the triplet state as ever being of any importance in organic molecules in general."

Edward Teller proved to be a very imposing personality on the lecture platform. His booming voice resounded, his bushy black eyebrows quivered and rose and fell with emphasis as he spoke, his hand brandished the chalk as he attacked the board as with a stiletto. In ten minutes he had filled the board with quantum mechanical expressions of amazing complexity. *Q.E.D.* He was finished, triumphantly. "There could be no observable triplet states in molecules containing only C, H, N, and O atoms," he said.

Then, a rather ascetic, pale, thin man whom I didn't know got up from the center of the front row and asked in a mild voice with his finger pointing to the blackboard, "Edward, wasn't that atomic *mercury* for which you showed us your calculation?"

"Yes, Robert, it was atomic mercury," Teller replied, looking rather flustered. It was Robert Mulliken, saying "Dr. Kasha was talking about organic aromatic molecules. I think what Lewis and he are claiming sounds *perfectly plausible*."

Mulliken then asked Smyth as chairman (a rather submerged rôle that morning) for permission to speak. Robert Mulliken then ascended the platform, and drew several diagrams, slowly and carefully explaining how, stemming from several molecular origins, rather long-lived triplet states of organic molecules could arise, even with lifetimes of many seconds, as Lewis and Kasha had found. The argument carried the day (for some), in spite of the powerful challenge, and the morning of the conference ended on this note.

Franck and his colleagues were not convinced, and several years of challenges continued.

### Faraday Society discussion

Three years later I had been invited by W. C. Price to a Faraday Society Discussion on Spectroscopy in Cambridge in September 1950. At this Symposium James Franck was honored by a Faraday Society award at a gala evening banquet in Trinity College, Cambridge. By the time of the Symposium I had completed the measurement of the paramagnetism of a molecule in its lowest triplet state (the Lewis posthumous paper on photomagnetism: Lewis, Calvin, and Kasha, 1949); had learned about the quantum mechanics of spin-orbital interaction from David Bohm, and had established the atomic number ( $Z$ ) effect on radiationless transitions; had developed generalized ideas about  $n \rightarrow \pi^*$  transitions; and had understood the kinetics of *internal conversion* and *intersystem crossing* (the neglect of which had caused Teller's misconception on the observability of metastable molecular triplets). These topics were succinctly reviewed in my Faraday Discussion on Spectroscopy paper (1950).

Sir John Lennard-Jones was the chairman of the morning session and introduced me, reminding me of the rule of the Faraday Society Discussions, indicating that I was to present the highlights of my paper in five minutes, for which the manuscript had been circulated a month before to Faraday Society members. I presented my summary and returned to my seat.

James Franck asked for permission to speak and went to the platform. He embarked on a long presentation, again seemingly without punctuation, developing a labyrinthian argument against the general idea of a phosphorescence of a molecule as being emission from the lowest triplet state. His "remark" or question seemed to go on for many minutes. By this year my background had deepened considerably, with a year in Robert Mulliken's laboratory just completed, a year full of spectroscopic discussion with John Platt, and great theoretical guidance from Clemens Roothaan and Klaus Ruedenberg. But Franck's "remark" was so complex and undecipherable in its totality that I did not know what I would say when Lennard-Jones called me back to the platform. With a bemused smile on his face he asked, "Would Dr. Kasha like to give an answer to Professor Franck's remark?"

I stated very frankly, "I am afraid that by the time Professor Franck got to the end of his remark, I could no longer remember what he said at the beginning!"

Franck then said, "You know what I mean." He and I had had a series of long conferences at Chicago and on the occasion of a long visit to Duke University (where Hertha Sponer, his wife, was living). These were always pleasant discussions, but there were profoundly complex thoughts in Franck's mind which always led to his rejection of our triplet state interpretation.

At this Symposium I gave a quick reply about his concern for vibrational-electronic interaction in spin-orbital coupling. This reply was published as is the rule with the Symposium. The following week in Manchester where I was spending the year, I discussed this topic with Christopher Longuet-Higgins, reminding him of Franck's remark and showing him my reply. He said that he would have been inclined to give an analogous reply to the one I had given. I added a further "Added in Proof" clarification. Many years later, however, I learned about higher order effects, and realized in retrospect that Franck, judging by his complex phraseology, probably had second-order vibronic perturbation in spin-orbital interaction in mind, long before it became a textbook subject!

Another strong challenge to my Faraday Society Discussion paper came from Alexander Burawoy. Both the triplet-singlet and the  $n \rightarrow \pi^*$ ,  $\pi \rightarrow \pi^*$  assignments constituted refutations of his K- and R-band designations, he felt.

His attack was long and even bitter, and Lennard-Jones had to cut him off. I was almost afraid to pass Burawoy in the aisle as I reapproached the platform. I answered him spontaneously as well as I could.

Charles Coulson, possibly in reaction to the strongly negative reception my paper had received from the floor, wrote a warmly appreciative appraisal of my presentation in a review of the Faraday Society Discussion, published in *Nature* in 1950. Coulson then invited me to give several lectures at King's College, University of London, which he had just joined. We developed a fine special friendship. Strangely enough, I did not feel in the least bit rebuffed by the Faraday Discussion, as I was confident that the ideas I had presented would survive. This has proved to be the case.

#### **Franck and Teller**

Franck was greatly influenced in his thinking in his later years by the theoretical ideas of Edward Teller. The subtlety and elegance of Teller's thinking in molecular physics is best exemplified in the great Herzberg-Teller papers on vibronic interaction in polyatomic molecules. Franck understood these ideas in a deeply intuitive, qualitative way, but never used mathematics in his thinking. This subject, complex and subtle in its mathematical treatment, was rendered almost undecipherable in Franck's verbal portrayal. One can easily understand Lewis' discomfiture with such a discussion: Franck undoubtedly used Teller's mathematical concepts as the background of his thinking, and Lewis thought in terms of specific molecular skeletons and electrical charge distribution models (without quantum mechanical conceptual bases).

Franck and Teller published a paper on molecular excitons in *J. Chem. Phys.* which epitomizes the problem of verbalizing mathematical ideas. This paper presaged much later development of the exciton weak and strong coupling cases, as phenomenologically laid out by William T. Simpson and his students, and later workers. But the Franck and Teller paper, richly brocaded as it is in verbal concepts, is decipherable mostly in retrospect. I sometimes have my students read this paper as an illustration of the complexity of written language compared with the clarity of mathematics in the expression of complex scientific thought.

#### **Franck's later years**

I got to know Franck in a much more personal way in the course of a series of long conversations with him over the years in the interval from about 1952 to 1964. Most of these were in Falmouth, Massachusetts where Hertha Sponer had a summer house, and some were on visits to Sponer's spectroscopy laboratory in the Physics Department at Duke University and at Sponer's home in Durham, N. C. when Franck visited from Chicago. In these long discussions I found Franck to be very deeply informed on classical physics and especially in his knowledge of electromagnetic and optical phenomena. We gradually came into concurrence on our spectroscopic thinking. I also found that Franck, when finally convinced of the validity of a new idea, was then flexible enough to adopt it. So after all the hesitancy about lowest triplet states of molecules, Franck wholeheartedly accepted all of our previous work and attempted to adopt it to photochemical problems, especially photosynthesis.

Franck at a rather early age had had a heart attack, which led him to be very cautious about physical exertion. Also in his childhood he had stammered badly, and slowly overcame this speech difficulty. He told me his teachers regarded him frequently as being stupid because of his inability to give quick answers on account of his speech impediment. After he overcame this, he had learned to be very patient with people who spoke slowly or who responded slowly. He believed that there was little correlation between the deep scientific thinker and the rapid scientific talker. So the personal relation one had with Franck was to hear a very soft, deep, gentle voice, speaking most thoughtfully; and to have him listen deeply with the greatest attention and patience. His counsel was golden.

One day, in about 1954, Franck's physician told him that his heart had completely healed, that he was one of those rare cases where he was no longer a heart patient in spite of his attack some twenty years before.

By chance, I was in Philadelphia for a week of lectures at Haverford College, and he came to lecture at Bryn Mawr College where one of his former students was a faculty member. The new James Franck was remarkable. How vigorous was his lecture! How joyously he moved about, drew diagrams, and waved his arms in demonstration. This was a Franck we had not seen before. His life from then on took on a new vigor.

A few years later he made a visit to the Florida State University where he gave lectures in physics and in chemistry. He acted like a young scientist. He had a new mastery of current literature, and spoke with a new forcefulness. It was good to see Franck like this.

#### **Franck the humanitarian**

No account of James Franck could be complete without mention of his profound humanitarian vision and efforts. He authored the Franck Report during World War II, co-signed by his colleagues at Chicago, which can be regarded as the Magna Carta of the Atomic Age. Not widely circulated, it is nevertheless destined to be regarded as a prototype example of scientists' stepping out of their laboratories to exercise a societal humanitarian and technical advisory influence. Franck made numerous efforts in this direction.

When Franck was still in Göttingen before World War II he was one of a select group of some of the world's most distinguished scholars. Official word came to him that he, although being a Jew, need not think that Hitler would trouble him, for after all, he was a Nobel Laureate. Franck then left Göttingen at once. Once in Chicago, colleagues began to notice that his clothing began to look more threadworn and shabby with each passing month. Finally, he seemed to be in desperate financial straits. Then it was discovered that he was using practically the whole of his salary and resources to save as many scholars threatened by Hitler's rampage as possible. I do not know the precise list of these, but I believe that Franck had sponsored or assisted about 12 notable scientists in their escape from Europe on the threshold of war. The University of Chicago then came to the fore and obtained for Franck and his growing laboratory more adequate support from the Fels Fund. This worthy sponsorship sustained Franck's research efforts until the end of his life, and later continued the support of Hans Gaffron, Franck's long-time colleague in photosynthesis.

James Franck died on a visit to Germany in 1964 at the age of 82. He is justly remembered as one of the most influential scientists of his time, and one of the finest examples of the human race.

### **ROBERT S. MULLIKEN – MOLECULAR SPECTROSCOPIST AND QUANTUM CHEMIST**

#### **First meetings**

After the first morning session of the Lewis Symposium in December 1946 Robert Mulliken and his students and former associates invited me to join them for lunch. At this luncheon John Magee of Notre Dame and Henry McMurry of Phillips Petroleum Research Laboratories were present, together with Robert Mulliken and John Platt, the latter having just joined the Physics Department of the University of Chicago. Also present were some of Mulliken's new associates, notably Klaus Ruedenberg, and Clemens Roothaan, the latter just completing his doctorate research. The luncheon lasted almost two hours and had an atmosphere of excitement and discovery, as we sat around a large round table and exchanged questions and answers. The powerful circle of spectroscopists and molecular theorists was the first I had ever met, and it was a joy to me to satisfy part of their hunger for more knowledge on the previously elusive triplet states of organic molecules.

Robert Mulliken then invited me to come to Chicago and offered to sponsor my applications for an Atomic Energy Commission post-doctorate fellowship in physics at Chicago, as well as a Guggenheim Fellowship.

In May of 1949 Robert Mulliken conducted a Spectroscopy Symposium at the National Bureau of Standards under the auspices of the American Physical Society. The titles of the six invited lectures all began, "Where do we stand in ...." e.g., "Where Do We Stand in Atomic Spectroscopy," *et cetera*. I was invited to speak on "Where Do We Stand in Polyatomic Molecule Spectroscopy." This was my first public lecture on the spectra of pyridine and diazines, and it intruded on sacred territory. Sponer and Stücklen had assigned pyridine vapor absorption spectra at high resolution as a  $\pi \rightarrow \pi^*$  transition, following Mulliken's discussion of electronic homology between pyridine and benzene. Hertha Sponer had years of experience on high resolution spectroscopy, and the team of Hertha Sponer, Gertrud Nordheim, Alfred Sklar, and Edward Teller was as potent as one could assemble. This methodology seemed ironclad, as they worked their way through complex problems of benzene spectroscopy. Hertha Sponer was upset by my presentation, on solid experience. Robert Mulliken made no comment, but Christopher Longuet-Higgins was in his first post-doctorate visit with Mulliken in Chicago, and he quickly and strongly supported the possibility of  $n \rightarrow \pi^*$  transitions in pyridine and diazines.

In September 1949 I was able to come to Chicago under the AEC Post-Doctorate Program in Physics.

It was stimulating in the extreme to be surrounded by the deeply competent people working with Robert



Mulliken but my big surprise, especially in my first interview, was the reticent response Mulliken seemed to give to any discussion of new ideas - very frustrating to an enthusiastic beginner.

Shortly afterward I was invited to visit Duke University, and Gertrud Nordheim, Hertha Sponer, James Franck, and I had very long discussions about all of our electronic conflicts. This continued periodically during my subsequent year at Chicago, and afterwards.

#### Mulliken the scholar

If there ever was a scientist who could sit down with a blank piece of paper and pencil and think new science, it was Robert Mulliken. His thought processes seemed to be so deeply engraved and so intensely personal, that a conversation became unidirectional input for Mulliken, unless he had already developed many thoughts on the matter. I found that other visitors to Chicago had experienced interview visits similar to my own first one.

Much later Robert Mulliken would always come forth with very sound and very fundamental criticism of ideas presented earlier. For example, I found that Mulliken did not like my orbital designations for electronic transitions, e.g.,  $n \rightarrow \pi^*$ , but the first detailed criticism I received on this was at a Quantum Chemistry Conference sponsored by Charles Coulson in Oxford in 1958. I recognized that  $n$ -orbitals were not truly "non-binding" (I suppose that initially I had inherited Lewis' ideas on "lone-pair" electrons). Nevertheless, I also recognized that spectroscopic nomenclature was based on *zeroth order* descriptions and that the designation of molecular electronic transitions by orbital origin had proved enormously useful. Today ESCA research results indicate how strongly delocalized " $n$ -orbitals" are in the  $\sigma$ -bonding skeleton. Robert Mulliken perceived this two decades earlier, and in this as in many other details his earlier thinking usually has been vindicated by later results.

Mathematics was one of the chief tools of Robert Mulliken's research, but I believe that he used this tool schematically for conceptualization rather than deductively or for calculation. Robert Mulliken was a formulator of conceptual frameworks, rather than being a scientist who used these frameworks generally for computation. This attribute may have arisen from his deep preoccupation with *classification* of molecular electronic states, and especially with the endless detail of their electronic-vibrational-rotational substates. In his early work on molecular oxygen, he referred to E. Hückel for the quantum-mechanical details of energy calculations, while Mulliken himself at the time was mainly concerned with spectral assignments.

Robert Mulliken's writings do get too complex for facile reading quite often, especially if he embarks upon a comprehensive treatment. His monumental paper in *Journal de Chimie Physique* on molecular orbital theory is probably not the ideal way to get a working introduction to the subject. It requires a translation (aside from French) to bring it within the scope of an average good scientific reader. His great comprehensive surveys on the spectra of diatomic molecules are of the same character. Gold is undoubtedly awaiting to be mined from the Mulliken papers, but it takes a very patient, indefatigable, and skillful miner to do so.

#### Mulliken the lecturer

Robert Mulliken had a unique reputation as a lecturer, but his was not a style easily followed, understood, or even appreciated. When I first came to Chicago, I wanted especially to attend Mulliken's course on Molecular Orbital Theory, which was to be given in the Winter quarter. Noel Bayliss was in Chicago for several months during his sabbatical leave from Perth, and we sat together in the second row among a handful of graduate students.

I had a new thick notebook ready to be filled with riches from the Master. I think that Noel and I both felt: At last, here we have a chance to get a real insight into molecular orbital theory.

Robert Mulliken entered the lecture room carrying a thick folder of notes. In his soft, hesitant voice he began: "Well .... I shall be lecturing .... on molecular orbitals .... So .... I shall need .... a suitable symbol .... eh .... with suitably designated .... constraints ...." Mulliken thought for a few minutes, then he wrote a lower case chi on the board. Then he thought for a few more minutes. Then hurriedly he erased the symbol, saying, "No, that won't do, we shall use  $\chi$  for something else. I shall use  $\psi$ .... prime .... sub-one ....." The lecture developed slowly. Noel Bayliss and I blinked at each other. Our anticipation was fading. As the course developed, we *did* derive some special insight, and there were nuggets of ideas to be collected, but the process was very slow.

Much, much later, I got a clue to Mulliken's lectures, and I record this as faithfully as I experienced the incident. After Robert Mulliken's Nobel Prize had been announced -- by chance while he was at the Florida State University in Tallahassee -- his wife Mary-Helen got worried about his lecturing: "Michael! You are a good lecturer. You simply must teach Robert to give better lectures!" A somewhat embarrassing situation. How does one approach a Nobel Prize winner on giving better lectures? I saw Mulliken the next day, and entered his office down the hall from mine with some diffidence. Mulliken said, "Mary-Helen told me she had asked you to talk with me about lecturing." That broke the ice, but how to begin?

So I asked, "Well, Robert, tell me, how do you prepare a lecture?" Mulliken replied: "Does one *prepare* a lecture?" That, to me, told the whole story. Robert Mulliken was a chain thinker. When he had a lecture to give, he would get up and *think* his lecture out loud, as a chapter from his file of thoughts on the subject. Well, at least that may have been his lifelong method. But his Nobel Prize lectures in Tallahassee, Chicago, and Stockholm were highly organized, and having heard the first two, I can say that these were delivered with an unaccustomed directness and even vigor. However, his habitual early style did, I believe, inhibit the general understanding and utilization of Molecular Orbital Theory by others until a later day.

### Mulliken and Pauling

Beginning in about 1930 the development of the Valence Bond Theory by Heitler-London-Slater-Pauling and the Molecular Orbital Theory by Hund-Mulliken-Hückel seemed to be on an equal footing. But Robert Mulliken as the quiet scholar simply could not match the exuberant Linus Pauling as a protagonist of the molecular orbital theory.

I was too young to be a witness, but older scientists have told me that Linus Pauling was the most flamboyant, dashing, dramatic chemical theorist at large in the world of chemistry in the 1930-1945 period. Blond wavy hair flying, blue eyes sparkling, arms waving in demonstration, Pauling hypnotized more than a generation of chemists. The Valence Bond Theory ruled supreme. Pauling's *Nature of the Chemical Bond* (Cornell, 1939) was everybody's handbook to the glories of molecular quantum ideas, or at least some derived results.

Robert Mulliken's personality prevented his presentation of his knowledge of Molecular Orbital Theory, either in writing or in speech, in any manner comparable with that of Linus Pauling. The contrast between these two men may be the best example we have of the major rôle personality may play in the development of a science.

Linus Pauling was always careful to distinguish sharply between the nature of the formal Valence Bond Theory and his own Resonance Structure Theory, abstracted from it qualitatively and intuitively -- a distinction lost on the average chemist-passerby. William Simpson later established a formal connection between the two through what could be called a Valence Bond *Structor Theory* (*J. Am. Chem. Soc.*, ~ 1950). The Valence Bond Method has proved to be too cumbersome in comparison with the Molecular Orbital Method in numerical calculations of polyatomic molecules. So today it finds very limited usage.

It was the school of English theoretical chemistry which brought Molecular Orbital Theory into its proper perspective in dealing with polyatomic molecules. First, Sir John Lennard-Jones, then Charles Coulson, and several generations of their brilliant students, were able to demonstrate in a continuous stream of researches the value of the Molecular Orbital Method in dealing with polyatomic molecule computations. Charles Coulson's book, *Valence* (Oxford, 1951), was powerfully influential, as were Coulson's and Lennard-Jones' research writings and those of their generations of brilliant students (S. F. Boys, A. D. Buckingham, G. G. Hall, H. C. Longuet-Higgins, R. McWeeny, W. Moffitt, J. N. Murrell, J. A. Pople, G. S. Rushbrooke).

### Mulliken in Florida

In 1960 the Institute of Molecular Biophysics was established at the Florida State University under my direction, with generous and extended funding by the U. S. Atomic Energy Commission - later becoming the U. S. Department of Energy. This Institute had a catalytic effect on science development in the University. Under this sponsorship various notable scientists were invited for extended visiting research periods. The list included John van der Waals, Theodor Förster, Alberte and Bernard Pullman, John Murrell, and Noel Bayliss among those in the field of Chemical Physics.

In 1962 Robert Mulliken stopped in Tallahassee on his way back to Chicago from the Sanibel Symposium and raised the provocative question: "Michael, do you know what they're about to do to me at Chicago?" I guessed immediately: "Robert, you're just past 65, and they're getting ready to terminate your appointment," - just as they had done with Harold Urey. I said to Mulliken: "We shall have to do something immediately to give you a place to work." Mulliken had come to recognize that the Florida State University had indeed proved to be a well-developing academic center, and he was keen on the possibility of migrating to Tallahassee if the opportunity could be created. Three hours later, Gordon Blackwell, the President of the Florida State University, had been able to make a tentative promise that a new position for Mulliken could be created. A month later an offer was made to Robert Mulliken to join the faculty as a Distinguished Professor of Chemistry and Physics, with the option of spending all or any part of each year in Tallahassee on this appointment.

Upon returning to Chicago Robert Mulliken indicated that he was not worried about his termination, because of the Florida State University developing appointment. George Beadle from Cal Tech, the new President of the University of Chicago, looked into the matter, and made an equivalent offer to Robert Mulliken to stay at Chicago, breaking their historical precedent. This somewhat confused the Mullikens, so for seven years they spent four to six months in Tallahassee in winters, and returned to Chicago each Spring Quarter. Robert Mulliken thus became the third accomplished Chicagoan to join the Florida State

University. Hans Gaffron had come to Tallahassee at Franck's suggestion, and Robert Strozier came as President, preceding Mulliken by a few years.

Quantum Mechanics was now represented very strongly in the state of Florida. John Slater and Per-Olov Löwdin were at the University of Florida in Gainesville. In Tallahassee, the Florida State University had Robert Mulliken in Chemistry (housed in the Institute of Molecular Biophysics), and P. A. M. Dirac in the Physics Department. Dirac had migrated to Florida after leaving Cambridge University and worked in the Florida State University until his death in 1984 at the age of 82. Having a seminar in our Institute by John Slater, with Paul Dirac and Robert Mulliken in the audience, was a thrilling experience.

In 1966 near the end of October the Mullikens had arrived for their fourth session in Tallahassee. The day after their arrival, it was announced in Stockholm that Robert Mulliken had been selected for the award in Chemistry for his work on the Molecular Orbital Theory. We knew that this award was for work done at the University of Chicago, but there was a tinge of poetic justice in his being in Tallahassee for the announcement. The Chicago people were very gracious, and celebrations were held on both campuses in due course, and I had the pleasure of being present at both. At the first news conference, Robert Mulliken was asked, "Are you excited?" He replied, "No, I do not feel particularly excited. However, I might add that I do not have direct personal evidence about the Nobel Prize; only the news that it has been announced. Probably it is true."

The Mullikens liked Tallahassee and their life there. They bought a very beautiful section of land in an exceptional residential area, and had intentions of building a house. This would be a great change from the apartment life they had experienced so long in Chicago. The plant life and wild life interested Robert especially (he was a scholarly amateur botanist), and their life was relaxing. As Robert Mulliken grew older he became more accessible to conversation, and his witty humor became more apparent.

At the laboratory Robert worked in his quiet office, or in the library, and was always available for informal chats. He gave an occasional invited lecture in undergraduate or graduate classes each year, and an occasional seminar. On two occasions, a research associate (Michiya Itoh) accompanied him from Chicago to carry out experimental research in spectroscopy.

After completing their seventh winter in Tallahassee, personal disaster struck the Mullikens upon their return to Chicago. Mary-Helen Mulliken had a stroke and was partially paralyzed. To get proper care she was confined to a convalescent hospital. Robert Mulliken stayed in Chicago after his wife died a couple of years after her stroke.

#### **Robert Mulliken the elder scholar**

Robert Mulliken enjoyed his life in Chicago among his colleagues and old friends, his books, and his unusual room-full collection of annotated reprints.

As Professor Emeritus of the University of Chicago he worked regularly and continued to do new research. As an elder statesman of molecular spectroscopy he was in a special position to take a leisurely backward glance at old fields of research, either neglected or confused by later workers. His restudies of such topics as the iodine and the ethylene molecules are in this category. He developed new theoretical expositions in this period, including a fresh discussion of Ruedenberg's analysis of the Virial Theorem applied to the chemical bond; and also an elucidation of the role of kinetic energy terms in the Franck-Condon Principle.

In 1975 the University of Chicago Press published a special volume entitled, "*Selected Papers of Robert S. Mulliken*," edited by D. A. Ramsay and J. Hinze. This volume of 1127 pages in large page format displays the intellectual range of Robert Mulliken in his long career as a molecular theoretician. The Appendix includes a four-page list of colleagues and associates of Mulliken through the years at Chicago, and includes a Bibliography of Mulliken's research papers and writings, 237 items (until 1974). The list grew steadily.

It is fascinating to take note of Robert Mulliken's two main interests outside of science. He had a life-long interest in botany, especially the nomenclature of trees, at which he became quite expert. Botanical classification, with its meticulous attention to intricate detail, seems to be intellectually parallel to his earliest interest in the detailed classification of the intricate details of molecular spectra. But botany gave Robert Mulliken a refreshing exposure to the living world of nature. Later in his life he became absorbed in the beauty and complexity of oriental rug design. The richness of the matrix art these represented fitted in perfectly with Robert Mulliken's love of and capacity for assimilating and integrating complex patterns. His campus fame in Florida upon winning the Nobel Prize led to an invitation to speak at the English Department's informal weekly *soirée*. He easily accepted this invitation, but immersed himself in an intensive week of preparation with giant volumes on oriental rugs obtained from the library. Unlike his unperturbable state at the announcement of the Nobel Prize, and in all of the interviews and press exposure, the week of his oriental rug discourse brought him to a very high emotional state. His anxiety the day of the lecture rose with each hour. By lunchtime he was flushed with color and was almost breathless. My concern was so great that I secretly arranged for a doctor to be present, ready for any foreseeable emergency. As it happened, the minute Robert Mulliken started talking about his latterly favorite avocation,

all anxiety faded away, and a rich exposure to a man's love for a subject charmed everyone present.

Robert Mulliken started an autobiography in his middle eighties, a work of great detail and interesting and curious observations, including very revealing ones about himself. Robert Mulliken died in 1986 at the age of 90. His autobiography (*Life of a Scientist*) was edited by Bernard J. Ransil, one of his last students, and was published by Springer-Verlag in 1989.

Robert Mulliken personifies the highly disciplined scholar who has been able to exploit the full range of the human intellect in the exploration of the complexity of molecular electronic structure. His rôle was not to show the simplicity of nature in its skeletal elements, but to reveal its intricacy of detail. He would be sympathetic to J. J. Thomson's observation: "Surely an atom must be as complicated as a Grand Piano." Robert Mulliken would be able to add: "And a molecule must be as complex as a Symphony Orchestra."

## ALBERT SZENT-GYÖRGYI – BIOCHEMIST

### Szent-Györgyi the experimenter

Albert Szent-Györgyi (Note a) had been searching for electronic phenomena to bring some new input into biochemistry, when he made a quite novel discovery on the phosphorescence of dyes. He sought to consult me for an explanation. After an exchange of letters, he decided to come to Tallahassee in November 1956 to lecture on and to demonstrate his phenomenon. We had never met, but I had no trouble in identifying him in the line of people disembarking from the airplane which had just landed at our small airport in Tallahassee. His energetic presence gave an immediate impression of a most vibrant personality. I said, "Szent-Györgyi, welcome!"

He looked puzzled. I said, "I am Michael Kasha." He looked at me questioningly, and then smiled saying, "Oh, did your father send you?" He seemed to be expecting an old man, presumably with a long white beard. He got over his shock quickly, and we became very good friends. His enthusiastic personality warmed every exchange.

In his lecture he described vividly why he thought biochemistry and biology needed input from molecular and especially from electronic theories in order to pave the way to a real understanding of biological phenomena on a molecular basis. Thus, he turned to triplet states of molecules in the hope of finding some new clues. But, being a biochemist, interested in the mysteries of life, he said he used "the solvent of life: water; and not those liquids you physicists use: alcohol, isopentane, ether, *et cetera*." He then showed how different the behavior of various dyes was in "frozen water solutions: at low temperature compared with frozen alcohol solution." The experiments were impressive. He had brought sealed sample tubes with him from his Wood's Hole laboratory, and showed us in a Chemistry Department Seminar the luminescence of various samples at room temperature, then in frozen samples at liquid nitrogen temperature. A particularly impressive case was that of acridine orange, which showed a green fluorescence at room temperature and at 77 K in ethanol solution; but in water solution the room temperature green fluorescence changed to a strong red phosphorescence at 77 K. This experiment is illustrated in a color plate published about that time in one of the series of small monographs which Albert Szent-Györgyi got into the habit of publishing periodically (this one titled, *Submolecular Biology*).

The challenge was thrown to us: how could the crystal field effect in water, specifically in its ice structure, give such a powerful new spin-orbital perturbation as to induce a specially high probability of intersystem crossing, uncharacteristic of dyes? The seminar baffled me and my capable students, the group then including Seán McGlynn, Eion McRae, and others. The next morning we had Szent-Györgyi in our laboratory. "Do the experiment from the beginning, in front of us," I said. We gave Albert all the needed ingredients and watched. He literally shoveled dye into test tubes with a spatula, and added water. I was aghast. We would usually work with  $10^{-6}$  molar dye solutions to study their luminescence behavior.

I said, "Why, Szent-Györgyi, you are studying dimers, polymers, aggregates, and microcrystals!" Albert Szent-Györgyi seemed surprised, and still thought that "water forces" were important. He was correct, but in an indirect way. We had a very heart-warming friendly visit with him for about three days, and he returned to Wood's Hole.

The month of December 1945 was a month spent untangling the various possible explanations for this phenomenon of Szent-Györgyi's. Eion McRae was in his last month before submitting his very independent doctoral study on quantum-mechanical theory of solvent effects on spectra. He put the discussions on the right track by seeking to understand dipole-dipole interactions in dye-molecule dimers. But how to explain enhanced spin-orbital coupling and enhanced intersystem crossing? Every day for a week or two, another diagram would be presented on the board, and someone would see its flaws. The give-and-take between McRae and myself intensified, and finally we had it: there was *no* enhanced spin-orbital coupling. There

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Note a: Hungarian spelling of St. George.

was simply a kinetic effect on intersystem crossing competition with the singlet state fluorescence, arising from molecular exciton splitting in the dimer. The rôle of water was simply to reduce cationic repulsion, permitting dimerization of the dye molecules. McRae and I published a note on this "non-spin-orbital-perturbation phosphorescence enhancement in dimers" in a *J. Chem. Phys.* communication in 1957. Simultaneously, by a different route, William Simpson found the same phenomenon and explanation. This contact with Szent-Györgyi launched my laboratory and me into a decade of research on molecular exciton effects.

Albert Szent-Györgyi's rôle as a scientist in the second half of his career has been generally to make curious new discoveries, and to stimulate others to embark on wholly new directions of research. When Szent-Györgyi's little book came out declaring that triplet state excitation is enhanced by the forces in frozen water, another major research channel was opened: the Beukers and Behrend thymine photodimer experiment. Albert Szent-Györgyi had almost a religious following among many biochemists. Szent-Györgyi said one must freeze molecules in ice to enhance triplet state formation. So Beukers and Behrend froze thymine in ice, irradiated with UV, and produced their famous photodimer. This initiated a whole era of research on thymine photodimers and their rôle in DNA photoinactivation, mutations, repair mechanisms, *et cetera*. It turned out that the crystal structure of thymine hydrate simply gave the molecular juxtaposition favoring thymine *covalent*-dimer formation.

The two cases described above dramatically illustrate the stimulating rôle Albert Szent-Györgyi had repeatedly in the last three decades of his research life. Even though he himself made no major discoveries in the period to match those of his earlier career, his influence was widely felt.

Working with Albert Szent-Györgyi in his own laboratory a few days each time I was in Wood's Hole vividly showed Szent-Györgyi's research style. He began with a notion based on some physical idea, gleaned intuitively from reading, or a discussion. He approached the laboratory bench with enthusiasm. He had an idea to test. His store of chemical experience set his choices. He filled test tubes, measured out portions by eye, mixed solutions, thumb over test tube. Colors appeared. Solutions were frozen. Observations were made. The laboratory was not neat. In fact, the neat bench frequently became a mess. But new things were seen, experiments no one had thought of were done, and the curiosity was intense. No one had ever worked at the laboratory bench with more joy or more curiosity toward an intuitive goal.

#### Szent-Györgyi's discoveries

Albert Szent-Györgyi had been driven in his career by the intense desire to put meaning into biochemical research on the ultimate subjects of biology. What is the living state? What is the nature of muscular contraction? What is the difference between the normal cell and the abnormal, cancer cell?

In the first phase of his career Albert Szent-Györgyi's success came about from a supersensitive power of observation for the minutest detail, the minutest difference. What to other researchers might be a nuisance observation, or no observation at all, was to Albert Szent-Györgyi the pivot point to a major discovery.

When using a blue copper ion reagent in testing for sugars in biological extracts, Szent-Györgyi happened (naturally) to have a Hungarian white-paprika extract on hand, and noticed that this particular extract gave a slight time delay in the color reaction. Why the time delay? Where another research might have passed this over for the serious work at hand, Albert Szent-Györgyi stopped to investigate. Presently from paprika he extracted a substance which proved upon testing to have anti-scorbutic properties. Now called ascorbic acid, it caused a sensation: it was the first case of a "vitamine" isolated as a crystalline substance, Vitamin C. Albert Szent-Györgyi won the Nobel Prize for this discovery and instant world fame (in spite of some early controversy on identification and priority).

Muscular contraction fascinated Albert Szent-Györgyi, partly because it is so fundamental a biological property of the living animal, and partly because, in its interaction with the neuronal system, it suggested to him biological action promulgated over distance. In some routine extractions from muscle protein, Szent-Györgyi happened to notice that one solution was a little more viscous than the others. He stopped his preparatory work to investigate. What caused this extra viscosity in one particular solution? He isolated a new muscle protein, *actomyosin*, one of his great discoveries. This launched him as a pioneer in key research on muscular contraction.

By 1940 Albert Szent-Györgyi was feeling uneasy with conventional biochemistry and biology. Many biologists are tempted to become biological mystics, so overpoweringly mysterious do the ways of living organisms seem, and so inadequate the simple laboratory methods and concepts. So Szent-Györgyi began sending intellectual antennae into the realms of theoretical physics and chemistry. He was convinced especially that nerve conduction and muscular contraction may involve phenomena known to the solid state physicist and untapped by the conventional biologist.

So to a very strong degree, Albert Szent-Györgyi began to abandon his thinking based on conventional biochemical methods and to convert his thinking and approaches to electronic and quantum concepts. This

conversion of Szent-Györgyi marked the transition from his period of singular biochemical discoveries stamping his earlier biochemical career, to his rôle as a biological *provocateur* and as a searcher for a new quantum-electronic basis of biological action.

### Quantum biology – the elusive ingredient

Albert Szent-Györgyi's first recorded contact with quantum physics is published in his Koranyi Lecture. This was published in 1941 simultaneously in *Nature* and in *Science*, an obvious indication of the importance which Szent-Györgyi attached to his thesis. The thesis of this paper is that since obviously (*sic.*) nerve conduction and muscular contraction are not explicable by classical models, we must seek answers in quantum theory. The *Conduction Band Theory* of the metallic state provides just such a model as needed for action at a distance (Szent-Györgyi said), so obviously that is how things are in biology. This theme was elaborated upon in one of Szent-Györgyi's little monographs, *The Chemistry of Muscular Contraction*.

Albert Szent-Györgyi had approached his colleague and friend, Zoltan Bay, the professor of theoretical physics at the University of Szeged on this thesis of biological action at a distance. Zoltan Bay, as a theoretical physicist steeped in quantum ideas, eagerly described the Conduction Band Theory of metals to Szent-Györgyi. Zoltan Bay accepted Szent-Györgyi's biological thesis at face value, and Szent-Györgyi accepted Zoltan Bay's quantum physics at face value. I believe this thesis to be mistaken, and an example of uncritical application of one field to another. The power of Szent-Györgyi's influence however, has been very strong and aspects of this thesis are still abroad today.

The trouble with metallic Conduction Band Theory is that it represents *strong interaction* theory for atomic particles. The atomic identity is lost in the formation of the solid state, wholly new energy states and properties arising in the metal. In molecular solids and aggregates, *very weak interaction* forces apply, and molecular identity is preserved: the peptide group, and the peptide chain side groups are still recognizable by their individuality in the complete protein; in nucleic acids, the spectral properties are little changed from those of their component bases. Thus, it seems simply meaningless to apply strong interaction theories to biological macromolecules when obviously only very weak interaction theories could be involved.

In 1948 I published an article in *Science* as a critique of Albert Szent-Györgyi's thesis on the "Continuum Model" of proteins. Again in 1958 I published a careful schematic analysis of molecular interaction in solids and aggregates (Boulder, Colorado Symposium, 1958, published as the book, *Biophysical Sciences Study Program*, Wiley; and in *Reviews of Modern Physics*), criticizing Albert Szent-Györgyi's thesis. One of Szent-Györgyi's students, John Gergeley, had gone to Manchester, and with M. G. Evans published a calculation on molecular orbitals applied to  $\beta$ -pleated-sheet H-bonded protein structures. This primitive calculation sought to show that a protein could be a semiconductor. The calculation failed to do so, even with its arbitrary adjustable parameters. But Evans and Gergeley in their *J. Chem. Phys.* paper (1949) gave an enthusiastic encouragement on their thesis of semiconductivity. This topic was straightened out by Alberte Pullman in a careful *a-priori* SCF MO calculation (in *Modern Quantum Chemistry*, ed. by Sinanoglu, 1964). This calculation shows that a pleated-sheet protein has such a large gap between the filled and unfilled orbitals, that the protein cannot be a semiconductor, only an insulator. The hydrogen-bonds allow too little electronic interaction.

### Wood's Hole conferences

After leaving Hungary and a lifetime of adventure behind him, Albert Szent-Györgyi found his permanent home on Cape Cod, Massachusetts. At Wood's Hole, a beautiful peninsula covered with picturesque bays and hills, lies the New England marine landscape at its finest. There is long established a series of government and private laboratories in oceanography, fishery, and marine biology. It was to the Rockefeller Foundation-initiated Wood's Hole Marine Biology Laboratory that Albert Szent-Györgyi came, establishing his Institute for Muscle Research. In the summer research period it would be difficult to be anywhere in Wood's Hole: at the beaches, docks, streets, restaurants, and not meet with a scientist or group of scientists, as hundreds make their annual pilgrimage there.

Albert Szent-Györgyi invited me to be a house guest at his rambling, picturesque old house on Penzance Point in June 1957. My wife Lilli and I enjoyed Albert's colorful tales of his life experiences, and his robust sense of humor. For a couple of weeks I lectured spontaneously for two to three hours every morning to his research group of a half-dozen co-workers, which included the physicist Irvin Isenberg, then converting to biophysics. Life with the Szent-Györgyis was colorful and gay. Swimming in the cold ocean, fishing for flounder, daring motor boat rides through narrow channels in the nearby islands, night fishing for striped bass in the swift tidal currents around the peninsula, volley ball - all these were part of the informal life of Albert Szent-Györgyi. Mornings of intensive science and late afternoons of intensive recreation became the delicious summer diet at Wood's Hole. Our enchantment with this lasted ten summers, and our memory of those good days calls us back.

In the summer of 1958 Albert Szent-Györgyi sponsored a giant summer conference with government support. Among the conferees were Hugo Theorell, the biochemist; Theodor Förster, spectroscopist and

physical chemist; John Platt, molecular spectroscopist; William Arnold, photobiologist; Zoltan Bay, theoretical physicist; Alberte and Bernard Pullman, quantum chemists; William McElroy, bioluminescence biochemist; Irvin Isenberg, physicist and biophysicist; Grigorio Weber, biochemist and luminescence expert; Sydney Vellick, biochemist; Richard Steele, biochemist, Irving Klotz, physical biochemist; Henry Linschitz, photochemist; myself as molecular spectroscopist and physical chemist; and, of course, Albert Szent-Györgyi as our enthusiastic host and venerable group leader. The theme of this conference was: What are the ideas of quantum electronic phenomena? What are the problems of biochemistry and biology? What bridges of understanding and interpretation could be built between them?

The conference was one of the major educational and stimulatory events in the lives of the participants. Most of us had not met before, and these meetings not only had a profound effect on the direction of our research interests, but led also to new lifetime friendships.

I developed a wicked reputation, at least in Albert Szent-Györgyi's mind, through our daily many-hour sessions each morning and each afternoon. My style of exposition and my extensive use of diagrams made everything complicated seem very clear and simple. But Albert Szent-Györgyi soon learned to his dismay, that he could not remember or reproduce any of the ideas and interpretations later. And I learned that unless one teaches and learns the underlying fundamentals, the easy interpretation may not seem so easy in a repetition.

### **82nd birthday symposium (Note b)**

No one who knew Albert Szent-Györgyi would have expected the symposium celebrating his 82nd birthday to be a conventional experience. But with all the great expectations generated by the program plans, the symposium climaxed with an event which even the strongest imagination would have been taxed to forecast.

The build-up of warmth of personal response toward Albert Szent-Györgyi was steady, with each lecturer's recounting of his career experiences. The build-up of esteem for Szent-Györgyi grew hourly as every research specialist got an appreciative and learned glimpse of Szent-Györgyi's influence and discoveries in his other great areas of biochemical research. This impression was the more deeply engraved as various lecturers displayed Albert Szent-Györgyi's own words from his publications of the era of discovery. The lucidity, the prescience, the drive toward a new understanding, all came through with thrilling impact.

The invited lecturers had finished their two days of warm appraisal in the symposium, and now the stage was set for Albert Szent-Györgyi. There was a certain tension and excitement. What would our great friend do? Would he, too, reminisce? Would he falter? The only person not looking backward, not even casting a glance over his shoulder that day, was Albert Szent-Györgyi.

He started a little hesitantly, in a slightly hushed voice. He later confessed privately that he felt an enormous pressure to make (even for him!) an unusually dynamic presentation. He started to build up his case with physico-chemical caution and solidity. It went well, and a physical chemist's ears heard music.

Then the experiments demonstrating his thesis began. Holding giant test tubes in his hands, first one above his head, then the other, gesturing with each to make his point, his face vividly animated, a brilliant red color sparkled as the liquids mixed. A dramatic sequence began, more experiments, color slides were projected, Szent-Györgyi danced about the platform, alternating his attentions on the audience and on his test tubes. It was a ballet, the "Ballet of Albert Szent-Györgyi," flushed with energy and scientific drama. The physical chemistry and biochemistry were too tightly knit, too convincingly demonstrated, to be taken lightly. It was an hour of triumph.

Theoretical physicists teach us that the magnetic monopole, if captured, would represent a field strength, an energy, unmatched in the physical world. That afternoon in Boston, October 17, 1975, at 5 PM the magnetic monopole seemed captured within the heart and soul of Albert Szent-Györgyi. The intense magnetism, the pulsating energy, the sheer radiance, were felt by all.

### **Albert Szent-Györgyi – the man**

Knowing Albert Szent-Györgyi was to know a man with an unequalled zest for active physical life coupled with a deep love of intellectual pursuits.

Szent-Györgyi's physical activities belied his age. He learned to water ski at the age of 80. He bicycled or motor-scooted to the laboratory daily. He invented an ocean swimming course around the peninsula where he lived, in which one swims along with the icy current at 7 knots, then must reverse at the Point, and must back-swim vigorously against the current in order to enter a side channel. Albert has led many groups of visitors on this swim. The one lady who did not succeed was later rescued by a Coast Guard boat far from the Point, on her way out to sea!

In a real sense Albert Szent-Györgyi was the complete man. His deep tastes for music and literature were satisfied by a steady diet. But in this indulgence he was forever seeking the fresh intellectual and emotional experience. He always seemed to find a new piece of music about which he was excited. A latter-day acquisition in his seaview house was a replica of a Greek sculptured head of a horse. It appealed to Szent-Györgyi as a vibrant work of art because of the vitality captured by the sculptor through a just-credible anatomical distortion.

Albert Szent-Györgyi was so energetic intellectually that he gave the impression of being undisciplined as a scientist. This impression I believe was correct; he was impatient with rigorous detail. But he was quickly perceptive and imaginative, and this and his philosophical bent helped to make up for the unstructured nature of his knowledge.

As a European, Szent-Györgyi was polylingual in a pragmatic fashion. In his use of English however, he had mastered an inspired phraseology, and his thoughts expressed in his dynamic lectures and in writing have excited many a young scientist to higher planes of scientific endeavor.

#### **Albert Szent-Györgyi in his 80's**

Research support for Szent-Györgyi in his middle eighties was sparse. After a difficult period of diminishing research financing from government sources, a new private research foundation was formed by an admirer, a Washington attorney, who sought to raise research funds for the Nobel biochemist whom the government agencies would not fund. This effort was successful, although not without controversy. Two aspects of an elder scientist at work deserve some elaboration.

Society in general is unprepared for the Albert Szent-Györgyis of Science. When a research panel of young scientists receives a proposal for review, it is judged against other proposals by strictly commensurate standards and criteria. The *avante garde* proposal by an adventurous scientist is easily rejected for the safer conventional one. One would think that a scientist of Albert Szent-Györgyi's record and influence would have received automatically some moderate level of support by a simple procedure. He had uncovered new science before, and there was always a good chance that he would do it again. Nature is unpredictable in a high degree, and track records do count in Science.

Unfortunately, there is another aspect to this problem, and this may be a main difficulty for the older scientist. A compatriot of Szent-Györgyi's was the biophysicist Georg von Békésy, who won the Nobel Prize for his deduction of the inner-ear mechanism. Among von Békésy's many fine observations on the conduct of research ("Albert Einstein never had a research contract -- nor a personal secretary.") was his remark: "Everyone needs a good friend; even a scientist does. But what a scientist needs more than a friend is a very good enemy. Usually a good enemy later becomes a very good friend. Then is it the *obligation* of the scientist to find other good enemies." Substantial science lives on good criticism, and if one loses contact with this, one's Science may suffer.

Szent-Györgyi did not follow von Békésy's advice. In his later years Szent-Györgyi began to shun the scientific criticism of his colleagues. He published his researches under privileged academy auspices and in a series of small monographs - losing contact with the sharp critiques of his scientific peers. The consequence was certainly that in his last decades he provoked controversy, but made no discoveries comparable to those of his early career and in the more conventional publication modes.

Albert Szent-Györgyi died in October 1986 at the age of 93, and up to his last years continued laboratory research in his quest for new biological ideas.

Albert Szent-Györgyi stands as a unique scientist of his time. His keen powers of observation, his intense curiosity, and his energetic probing of the mysteries of biology had led him to a career of keynote discoveries and a career of inspiring influence in biochemistry.

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