Materials Design and Molecular Understanding

A Scientific Autobiography^{*}

by Arthur R. von Hippel

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Prehistory

Returning from two years of service in World War I -- and some additional months of military involvement in the time of turmoil that ensued -- I enrolled halfheartedly as a science student at the University of Göttingen in Germany. My father was a professor of criminal law and my grandfather a professor of ophthalmology. There had been admirals, generals and estate owners in the family but no scientists. My two brothers were studying law. It therefore seemed time to break free from tradition -- but with some misgivings.

The lectures of Professor Debye and Pohl respectively were full of theoretical clarity and experimental surprises and the friendships of old and new members of the youth movement provided a balance of human warmth and social engagement. After an escape to Munich for a term of art study and mountain climbing, my family's impoverishment by galloping inflation forced a speedy return and rapid progress toward a Ph.D. thesis.

The "Bohr Festspiele" (festival) in Göttingen in June 1922 -- organized by Hilbert, Franck, and Born -- proved a decisive event. There Niels Bohr presented his theory of the Periodic System.

We sat spellbound as Professor Bohr -- with inspired face and absentmindedness -- walked to and fro before the blackboard, on every passage hitting the ceiling lamp with his pointer. It began to sway in resonance with increasing amplitude -- and might have come down with a resounding crash -- had Hilbert not arisen, gently exclaiming "Great Master," and pried the pointer from Bohr's hand.

We had just began to breath again, when a voice came from the back: "I have a student who does not believe this!" and Sommerfeld marched forward, like a colonel of Hussars, with young Heisenberg in tow. A five-minute discussion arose. Then -- as Bohr turned back to the blackboard -- Born spoke up, "I also have a student who does not believe it!" and young Pauli arose, chewing his fingernails. When the lecture had ended, we normal students trooped out, in love with Bohr but deeply convinced that theoretical physics was a calling reserved for genius. I enlisted as a Ph.D. student in the "Institute for Applied Electricity."

Our money melted away as inflation ran its destructive course. Every morning at 10:00 AM, my father received his salary, with mother and four children lined up behind him. Then we raced into town to buy food, wherever it could be found because, at noon, a new, much lower value for the currency would be officially posted. I have still a 10-Reischsmark bill overprinted: "10 billion Reichsmark."

After these preliminaries, we children went to work, came home for supper, and frequently I then left again at 2 AM on my bicycle -- a candle in a paper bag as illumination in one hand -- to ride back to the institute for experimental work. My

thesis problem was the invention and theoretical description of a new, more inertiafree microphone for the broadcast age, which was just beginning.

In February 1924 the work was done, the thesis written and the Ph.D. exam passed "summa cum laude" -- fortunately, because my friends had gathered in a noisy crowd downstairs in the Aula [the assembly hall of Göttingen University] and awaited me with a tremendous cardboard top-hat with that inscription. As dictated by tradition, we marched to the marketplace with its "Goose-Girl" fountain and then climbed up to our fraternity home on top of the Johannis-church tower, where a final group singing above the town ended the proceedings.

The "thermo-microphone", described in two publications^{1,2} and patented, was a limited success. I listened anxiously as a soprano singer tried it out at the Berlin broadcasting station that year. At high C her voice vanished. Obviously my microphone was not the final word and it was soon superceded.

A friend from my father's East-Prussian childhood, Professor Max Wien in Jena, heard of my exam, liked the thesis and offered me an assistant position. He was the inventor of the "quenched spark-gap", which made transatlantic wireless communication possible. Experimenting still in his old age on electrolytic conduction at high field strengths, he discovered the two "Wien effects." In addition he was a wonderful gentleman, before whom we all trembled slightly.

Professor Wein wanted me to work on thin magnetic films made by cathodic sputtering. I asked how the mechanism of sputtering worked and, since nobody knew, set out to investigate it. It was a long and difficult spectroscopic study, measuring quantitatively the vapor pressure of the iron atoms in front of the receiving plate and comparing it with the build-up speed of the sputtered layer.³ The density of the iron vapor, calibrated spectroscopically against mercury vapor, established that the argon ions of the gas discharge released the iron from the cathode in the form of atoms.^{*}

The response to this *tour de force* were gratifying. I got my first Ph.D. student and the offer of a Rockefeller fellowship. Since I was of a somewhat adventurous bent -- having previously camped through Italy, Norway and England in youthmovement style, paid for by earning money through articles for an export journal --I chose Berkeley, California as the most distant possible place for my Rockefeller year. It was a wonderful experience, full of adventures and excitement in a still relatively empty West. Scientifically, the research problem I had chosen as my Habilitation^{*} paper for Germany was too difficult for the meager resources available but served its purpose.⁴ The greatest human gain was a strong friendship

^{*} To imagine the work involved in old-time experimentation, the reader must recall that we had to build all our equipment, including spectrographs and glass vacuum pumps.

^{*} The research paper required to qualify one for an academic career, starting as Privatdozent (equivalent to Assistant Professor).

with James Franck, who came to Berkeley as an honored lecturer during the winter semester. Little did I dream that he would become my father-in-law when I married a second time.

On returning to Germany, I married my beloved first wife --Marianne von Ritter -- and then lost her eight months later in the great flu epidemic of February 1929. Transferring to Göttingen to help her widowed mother bring up the younger children, I joined the Second Physics Institute, which was directed by James Franck.

Franck was one of the warmest and most inspiring personalities one could hope to meet. He did not calculate but visualized scientific phenomena with wonderful intuition. He provided, with decisive spectroscopic studies, the experimental basis for the quantum theory of atoms and molecules. His Institute, full of happy life, with its many students and foreign visitors, was counter-balanced by the Theoretical Physics Institute of Max Born, where men of genius like Pauli and Heisenberg and many other theorists of outstanding ability grew up. The First Physics Institute of Professor Pohl, an excellent experimenter and popularizer of science; the Mathematical Institute of Hilbert and Courant; the Geophysical Institute of Professor Goldschmidt; the Chemical Institute of Professor Windaus; and the Physical Chemistry Institute of Professor Tammann and his successor, Professor Eucken, -- all located near each other and in close intellectual contact -- made Göttingen a "capital of science."

It was a restless time, however. The impossible demands by the Allies for reparations forced Germany into bankruptcy and moral desperation. A scapegoat was sought and found by Hitler in the Jews. During my stay in America, I had dreamed and organized for an International Youth Movement Congress. Returning to Germany, I found the young people split up into Nazis and anti-Nazis amid growing unrest.

In the summer of 1930, I married Dagmar Franck, a close friend of my deceased wife Marianne, and thus choose my side in the coming struggle.^{*} There was still a breathing spell until 1933, during which I became deeply involved in the study of the mechanisms of ionization of gases, liquids, and solids by electron impact and of the electrical strength of materials.⁵ This work led to a deeper understanding of the development of sparks; the discovery of negative and positive sparks; an understanding of the role of electrons in the onset of breakdown in gases, liquids and solids; the determination of the true electrical strengths of alkali halide crystals and their changes as a result of added impurities; an understanding of the formation of color centers; and the extension of these ideas to thunderstorms and lightning strokes.⁶

Dagmar Franck was a Jew.

Electrical breakdown was a scientific analogue to the violent political breakdown that began to take place in Germany. Reichspresident Hindenburg, dotty with age, chose Hitler as his Chancellor in the hope of neutralizing the Nazis. Instead, the result was the burning of the Reichstag, the murder of the chief of the Army, the pogroms against the Jews, the start of concentration camps, the persecution of the Communists, and the dismissal of Jewish professors.

James Franck, decorated with the Iron Cross First Class in World War I, was exempt from dismissal but refused to accept this protection. His beautiful declaration of resignation, composed during a long night of debate and published in a Göttingen newspaper, resulted in we young assistant professors being responsible for the orderly closing-down of the institute's operations.

After refusing to swear the oath of allegiance to Hitler, I became an outcast, shunned by many old acquaintances like a carrier of the plague -- an initially upsetting but very educational but experience. Through Swiss intermediaries, I received an offer of a professorship at Istanbul, accepted and left in July 1933 for my new destination.

At that time, traveling to Turkey was a major undertaking: an eighteen hour railroad trip from Göttingen over the Alps to Trieste and then travel by steamship to Venice, across the Adriatic and through the canal of Corinth to Athens and finally across the Aegean Sea and through the Marmara Sea to Istanbul. Hardly anyone traveled for pleasure in those foreboding years. The eight-day trip was therefore an adventure of discovery and, when I learned that my new institute was to be set up in a section of the old Sultan's palace, it sounded like the beginning of a fairy tale.

In retrospect, our adventures were indeed fantastic. They included a trip with a German friend to Palestine (at that time still a British mandate) to find places to settle German-Jewish children. Our time in Turkey was also anxiety producing, hilarious and ultimately frustrating.

Therefore, after eighteen months in Turkey, I welcomed an offer from Niels Bohr to come to Copenhagen. In the wonderful atmosphere of Bohr's institute I was able to continue my studies on the electric phenomena at high field strength in gases, liquids, and solids.⁷ His friendship and that of his family became one of the great blessings of my life.

In the fall of 1936, at the invitation of M.I.T.'s President, Carl Taylor Compton, we moved from Copenhagen, to the U.S.A. and I started as "the" physicist in the M.I.T.'s Electrical Engineering Department.

The time had come to break away from classical engineering concepts: Vannevar Bush was experimenting with computers, Harold Edgerton with stroboscopic photography, Van der Graff built his tremendous electrostatic generator, and others pushed electric oscillators toward microwave frequencies. The time was ripe for the introduction of modern materials concepts into the field of electrical engineering. Therefore, after serving a short apprenticeship in teaching and supervising thesis students, I started, with a five thousand dollar grant from President Compton, the "Laboratory for Insulation Research."

I would like to insert here a short note about this unusual man. Compton was a very good physicist in his own right but simultaneously an ideal administrator. Full of human understanding, modesty, and scientific curiosity, he walked through M.I.T. in order to learn what was going on and where help or criticism was needed. He enjoyed new insights, was not afraid to ask "foolish questions," and tried to keep the faculty responsible for its own affairs. As a result, the administration of M.I.T. remained small, funds could be made available as seed money for new initiatives, and his door was always open for people in distress. When he died in 1954, we cried.

The name of our laboratory was chosen so as not to appear to infringe on the entrenched interests of M.I.T.'s science and engineering departments. Later, it proved a much too narrow designation but, since it had become widely known, we kept it.

A Breathing Spell Before World War II

Space at M.I.T. was at a premium. As a slightly suspect newcomer, I had to start my experimental work in the Institute's boiler room -- later advancing to the third floor, squeezed between graduate students. Slowly the pressure generated by a growing group of M.S. and Ph.D. students created a larger working area. Our research extended from crystal growing, and conduction and breakdown studies in gases, liquids, and single crystals to the formation of color centers and the strange behavior of semiconductors. Mr. Molnar, passed on to me as a Ph.D. student by Professor Harrison, found the M bands in the alkali halides. Fred Merrill and I cleared up the initial stages of the electric breakdown in gases using Lichtenberg figures.⁸ And, we began to understand the propagation of electrons in single crystals as a wave phenomenor;⁹ the importance of order vs disorder by comparing the properties of crystal with those of glasses;¹⁰ and advanced to studies of the effects on materials of temperature, frequency, and the transition from insulator to semiconductor and metal.

The outbreak of World War II in Europe began to draw us toward what was to become classified research. The International Telephone and Telegraph Company had lost its selenium rectifier facility and know-how in Germany and started a plant in New York but about 90 percent of the product had to be thrown away. The President of I.T.& T. therefore came to us for help and, after studying the phase transitions of selinium from the red insulating to the gray metallic form and the rectifying action of the boundary, we were able to design a method for producing good rectifiers rapidly by electroplating.¹¹ The production time was shortened from the two days taken by the thermal annealing process to twenty minutes with the electrolytic process. We also studied the making of Se photocells¹² and their spectral response and retained for M.I.T. the rights to the conversion of solar energy into electric power by this method.

At the same time, we extended our dielectric studies by measuring the dielectric and loss constants of materials from DC up to microwave frequencies.¹³ This work became important in World War II when the Laboratory for Insulation Research was made responsible for the development and measurement of Radar Dielectrics. The data from the measurements were classified "confidential" until the end of the War.

The War Breaks Out

In September 1941, I became a citizen of the U.S.A. and, on December 6, 1941, the Pearl Harbor attack pushed the U.S. into World War II. In contrast to my experience in Germany at the outbreak of World War I, the mood in the U.S. was somber without hysterical outbursts. As a result of an air raid alarm caused at M.I.T. by claims of an impending German attack everyone was ordered into the basement of Building 4. Brushing up my World War I anti-aircraft gunnery experience, I suggested to Dr. Compton that we set up a flak battery at M.I.T. and use gas grenades to cause misfiring of the airplane engines. Nothing came of the scheme besides the feeling that I could be trusted.

Some neighbors in Weston rumored spy stories about me for a short while but soon the community grew together in shared joys and sorrows.

Role of the Laboratory for Insulation Research During World War II

Our responsibility for the development and measurement of radar dielectrics -as well as for the initiation of their application and commercial manufacture and -saddled the Laboratory with a host of problems normally unknown in the academic world.

We had to create standard measurement techniques and equipment to determine the dielectric and loss constants of all kinds of materials as a function of temperature and of frequency from DC through the microwave region. The decimeter and centimeter wavelength range were practically unexplored territory. New types of standing wave equipment such as the M.I.T. Coax Instrument, had to be developed, and arrangements made for its manufacture and distribution to government and industry laboratories and to the Allies.

Polymers such as polystyrene and polyethylene -- previously only used as filler materials for rubber tires and for some household items -- had to be upgraded to low-loss radar dielectrics and, since the Navy persisted in pulling radar cables through the boiler rooms of battleships, their useful temperature range extended with additives. Many other materials -- plastics and rubber, ceramics and glasses, single crystals and polycrystalline materials -- were needed and had to be made in the Laboratory or by industry. Their characteristics had to be determined from as a function of frequency and temperature by our Dielectric Measurements Group and the results published in classified "Tables of Dielectric Materials."^{*}

In developing "High-Electric-Constant Ceramics," we discovered the ferroelectricity of $BaTiO_3$ and produced high-voltage condensers and ceramic delay lines. Our experience in making selenium photocells pulled us into war research on infrared photocells of the thallous sulfide type; and our dielectric studies led us to the application of dielectric heating for rapid wood curing. The classified reports of L.I.R. during World War II and subsequent publications give summarizing accounts of these activities.

In the course of these events, it became clear that a close liaison was required between our laboratory and the government agencies responsible for the procurement and proper application of dielectric materials. We therefore formed with the Army, Navy, and War Production Board, the "War Committee on Dielectrics." During the later war years we met in Washington once a week and -after mutual trust had been established -- a number of emergency situations were handled successfully. I remember especially two occasions where decisive action was required.

The war in New Guinea had bogged down, because mites and fungi ate up uniforms and equipment. We proposed to the Quartermaster Corps a changeover to halogenated compounds that could not be eaten. For a while, we met stubborn resistance because the Corps had made long-range contracts with its suppliers. To settle this argument, I proposed to set out to the battle zone with an experimental lab in a trailer and prove our point. Instead a colonel was dispatched to investigate and came back to say that he had only found mites and fungi in his trunk after returning to Boston. This made all of us so mad that finally action was taken. PVC and other halogenated materials were introduced and the mission was accomplished. That the later abuse of such materials as pesticides might lead to Miss Carson's "Silent Spring" could not have been foreseen.

^{*} Mr. W.B. Westphal, who joined us early in 1942 after receiving his Bachelor's degree, became the great expert in dielectric measurements and the group leader. He remained in the Laboratory as an old and trusted friend and continued publishing such "Tables" until 1980.

A still more hair-raising problem arose shortly before the Normandy invasion. The various Army, Navy, Air Force units involved each needed a separate frequency band for communication. Sharply tuned quartz-crystal filters had been prepared by the Signal Corps for this purpose but, unfortunately, these crystals had been housed in plastic containers. When they were taken out of storage, it was found that organic fumes emitted from the plastic had destroyed their silver-plated electrodes. A desperate last-ditch effort to rehouse the crystals in glass saved the day.

While our help as providers of information was needed in these and many other cases, the pressure to produce new materials and techniques forced L.I.R. to build a balcony system into our high-ceilinged main laboratory in Building 4 and to expand into a variety of other locations.

I was also a member of the Radiation Laboratory, the Central Organization for radar development at M.I.T. with its thousands of scientists and engineers -- but the L.I.R. kept its independence. A wonderful spirit of mutual confidence and friendship developed and lasted throughout life with many co-workers.

The daily living during the war was frequently difficult but sometimes also hilarious. A few reminiscenses may illustrate the situation:

- My salary was very low (ca. 3000 dollars per year) and remained frozen during the war. When three of our children were simultaneously hospitalized with ear infections, I had to sell my *Handbuch der Physik* to bail them out.
- Our working hours were very long. Once, driving home around midnight after an exhausting day, I turned into a deserted street and a policeman suddenly jumped out of hiding and claimed I had not properly stopped at the corner. I told him he should better enter the war effort instead of bothering people at that hour. Shortly thereafter I received a summons to appear before the Court in West Newton. Instructed by my lawyer I pleaded "*nole contendere*" -- but the judge did not accept that plea. Reluctantly, I pleaded "guilty," in order to avoid being dragged into a higher court. The judge was just starting to lecture me sternly when I heard the clerk whisper into his ear: "He is a professor at M.I.T." Changing his expression into a grin, the judge slammed down his gavel and declared, "Case dismissed!" Let us hope he knew about our war work and sympathized.
- As the gasoline shortage became critical, we formed a driver's club of five to six people and commuted together when our hours could coincide. Rationing was in full swing and people very hungry. While we were driving home one day, a pheasant flew into the windshield and was killed. Everybody looked longingly at the bird in anticipation of the wonderful meal his family might have, when our biologist spoke up in a cool voice: "The birds around here have a strange disease and can not be eaten; I will dispose of it". And this he did -- with his family.

When the war in Europe drew to a close, I volunteered to go to Germany and help with my knowledge of country and people. A military commission of some sort was required for that purpose. The Navy, bound by a rule that an officer had to have ten years of citizenship, could only offer the rank of "Seaman Second Class", while the Army proposed to make me a Colonel. This I accepted but Fate intervened. Completely overworked, I got a case of acute hyperthyroidism, losing a pound of weight a day and becoming terribly irritable. As a result, I found myself hospitalized for four weeks while my body slowed down for the operation -- and while my wife in a different hospital half-an-hour away was having our baby daughter. Our four sons, trouping from one hospital to the other, kept us in contact.

It takes years to rebalance one's system after a total thyroidectomy, but the Laboratory for Insulation Research had still its role to play in radar and infrared detection. I therefore carried on until the War ended -- and then through the transition that had to be made to peacetime operations.

This was an especially difficult period, because the Office of Scientific Research and Development (OSRD) under Vannevar Bush stopped and disbanded abruptly, and every scientist and engineer had to find a new home base. Most were dislocated and needed to be able to cite as many achievements as they could muster. As a consequence, much pressure was put on us to help complete experiments in which we had not been involved and to publish our scientific findings in coauthorship with others. We resisted these pressures because I was focused on a post-war challenge: transforming the whole field of materials research with a fundamentally new approach, "The Molecular Designing of Materials and Devices."

Any dedication to a new cause makes enemies and meets much resistance, but we had gained the trust of the armed services through our war-time performance. The L.I.R. therefore received the first three-service (Army-Navy-Air Force) contract to launch its post-war operations.

Aims and Approaches of our Post-War Research

When I first started my investigations of cathodic sputtering and electrical breakdown in gases, liquids and solids, the formation and properties of materials were generally described by thermodynamic - statistical approaches and by the phase diagrams of physical chemistry. The data were tabulated and accepted as prescribed by nature. Now, we asked more and more insistently how and why these phenomena happen and how they could be influenced by molecular channels.

During the war, we had been compelled to develop all kinds of outlandish instrumentations and materials. Now we wanted to gain fundamental understanding. In contrast to most university laboratories we did not respect departmental boundaries, but operated with a staff and students from a variety of departments. Simultaneously, we accepted numerous co-workers from abroad, including some from former enemy countries such as Japan, and therefore enjoyed a United Nations type of atmosphere.

Our progress was slowed by the fact that we had to design and build all our special equipment instead of assembling it from manufacturers' shelves as is done today. Also insight into the molecular events of trapping and transfer of electrons and ions and of transitions from insulators through semiconductors to metals, from crystals to glasses and polymers; from gases to liquids and solids; and from single dipole moments and spins to the coupled systems of ferroelectrics and ferromagnetics had to be acquired step by step. To grow prototype single crystals was a major research challenge and very expensive. For example, our first magnetite crystal cost us about \$20,000 in pre-1970s inflation dollars.

The details concerning the organization and activities of the L.I.R. between April 1947 and November 1965 are described in 34 unclassified semiannual Progress Reports and one Final Report. The population of the Laboratory grew from about forty to about eighty. About sixty Ph.D. theses, forty-eight Master theses and a very large number of Bachelor's theses were completed. A large number of post-doctoral co-workers from many countries who joined our research staff for various periods appear as authors or co-authors in our list of publications.

The goal of this pioneering effort "to unite science and engineering in the molecular designing of materials and devices" was revolutionary and seemed to endanger the entrenched interests of departments and schools. It was therefore hotly contested by "the powers that be" and patient demonstration efforts over many years were needed before the battle was won.

After writing a book *Dielectrics and Waves*¹⁴ for physicists, chemists and electrical engineers, I arranged a Summer Session Course at M.I.T. in September 1952 for scientists, engineers, manufacturers and users of dielectrics. The driving objective was that the participants should learn to speak each other's languages and share problems, failures, and advances. This goal was reached and a joint book, *Dielectric Materials and Applications*,¹⁵ was the result. It included in addition, the first open publication of the "Tables of Dielectric Materials" -- approximately 150 pages long.

Continuing this approach of mutual learning and understanding, we arranged a "Summer Session Course on Molecular Engineering" at M.I.T. in 1956 which ranged from the formation and structure of atoms and molecules through the designing of liquids and solids, including their electric and magnetic properties, explosions and breakdown to the air vehicles of the future. The book, *Molecular*

Science and Molecular Engineering,¹⁶ summarizing the outcome was a two-year effort that was finished in September 1958.

Still one more push was needed before the bandwagon began to roll. Asking, "What is the present status of our insights and capabilities?" the L.I.R. sponsored a summer-session course on "The Molecular Designing of Materials and Devices in 1963." The subjects ranged from "Prediction of *a-priori* Theory" and "Building from Atoms" all the way to the structure and properties of nonliving and living systems. The book, summarizing the insight of the scientists and engineers involved, was published in 1965 and marked the close of our effort.¹⁷

The battle had been won and I had reached retirement age. The Government wanted me to build a "Federal Center for Materials Science and Technology", but we insisted that such Centers belonged in the universities where young minds should be preconditioned for such integrating understanding. "Centers for Materials Science and Engineering" were therefore formed at various universities. They were still somewhat saddled with departmental allegiances, however, and also not quite ready yet to embrace the life sciences and medicine.

After a year of service in Washington as Science Adviser to the Naval Research Laboratory, I continued with a small research group under the name of the Laboratory for Insulation Research on my way "From Atoms Towards Living Systems,"¹⁸ until old age called a halt.^{*}

^{*} Of the many good scientific friends and associates I had the fortune to acquire, our books and reports provide documentation. Three of these associates remained my companions over several decades, however, and must be mentioned with especial gratitude: Bill Westphal, leader of the Dielectric Measurements Group; Aina Sils, secretary and librarian; and John Mara, illustrator of our publications.

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