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FREDERIK W. H ZACHARIASEN

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A Biographical Memoir by

MARK G. INGRAHAM

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Photo by Anne Plettinger

W H Zachariasen

FREDERIK WILLIAM HOLDER ZACHARIASEN

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BY MARK G. INGHRAM

FREDERIK WILLIAM HOLDER ZACHARIASEN'S contributions to science have been rich and varied. He was a leader in the determination of the crystal structure of inorganic crystals using x-ray diffraction. Though primarily an experimentalist, he contributed to theory whenever he found the theory inadequate. In over 200 published papers he included experiments on the crystal structure of minerals, on the structure of inorganic crystals, on the structure of anionic groups, on atomic and ionic radii, on the glassy state, on the liquid state, on actinide crystal chemistry, on high-pressure phases, on crystal structure and superconductivity, on the melting process, and on the variation of bond strengths with bond lengths. His contributions to theory include papers on temperature diffuse scattering of x-rays, on stacking disorder, on the phase problem, and on extinction including the Borrmann effect. Each of these theoretical efforts was followed by careful experimental investigations to establish the correctness of the theory he had developed.

Linus C. Pauling of the University of California at Berkeley, who also concentrated on the determination of crystal structure during his early years, said of Zachariasen's work (1975), "I feel that he is to be classed among the outstanding scientists of the twentieth century, and at the top in the field of inorganic crystal structures." Robert Penneman of the Los Alamos Scientific Laboratory said (1975), "The breadth of his contributions is enormous; there is no major advance in crystalography in one half century that does not bear his mark." Bernd T. Matthias of the University of California, San Diego, said (1975), "His was a monumental achievement in understanding the detailed nature of the whole periodic system." Such accolades abound.

THE MAN

Zachariasen had absolutely no use for pretense or titles. His friends and associates always called him by one of his two nicknames, "Willie" or "Zach." It would give completely the wrong impression of "The Man" if I were to refer to him as "Zachariasen" in this memoir. I will therefore use the name he preferred: "Willie." He would want it no other way.

Willie was born in Langesund, at the mouth of the Langesundfjord, in southeastern Norway. Willie's father was a sea captain. Langesund is about 15 kilometers from Brevick, which is at the center of the nepheline-synetic pegmatite veins which have yielded over thirty new species of minerals. The islands of the Langesundfjord are also rich in deposits of rare silicates and other well-crystallized minerals. Raymond Pepinsky, one of Willie's early Ph.D. students, relates a story in which Willie, decades after his youth, correctly identified on sight a Langesund eudidymite specimen which had been mislabeled by the Krantz firm in Germany. X-ray examination proved Willie correct. "Willie, how could you have known?" he was asked. "I played on the Langesund islands when I was a boy," he replied with his usual warm grin, "and I remember those crystals." Such mineral crystals so interested Willie that when he went to

Oslo University in 1923, he studied in the Mineralogical Institute under the guidance of the great geochemist Victor Moritz Goldschmidt.

Goldschmidt (1888–1947) was one of the first to recognize that crystal structure data as determined using x-rays could reveal the distribution of elements within crystalline minerals. Among the crystals which interested Goldschmidt, and which Willie studied while still in Oslo, were some rare earth crystals. Crystals containing these elements abound on the Langesundfjord islands. As Pepinsky tells us, "In order to protect the richest of such deposits for his own leisurely study of its mineral species, Goldschmidt purchased one of the islands Willie knew best."

Willie published his first paper, "Über die Kristallstrucktur von BeO," in 1925 at the age of nineteen. He published nineteen more papers before publishing his Ph.D. thesis in 1928 at the age of twenty-two. He was awarded the Ph.D. that same year. He was then appointed assistant professor at the University of Oslo, but was granted a leave for 1928–29 to accept a Rockefeller Foundation Fellowship to study with Sir Lawrence Bragg at Manchester University in England.

After his postdoctoral at Manchester, Willie returned to Oslo. Early in 1930 Willie received an offer from Arthur Holly Compton, Nobel Prize winner for the discovery of the Compton effect, to join the faculty of the Department of Physics of the University of Chicago as an assistant professor. Willie had been recommended to Compton by Bragg. Willie accepted that appointment and stayed at the University of Chicago until he retired in 1974.

One week before Willie sailed for New York he married Ragni (Mosse) Durban-Hansen, granddaughter of the pioneer Norwegian geochemist W. C. Brøgger (1851–1949). Brøgger, among other things, had discovered and first described the minerals of the Langesund region. In many of Willie's publications on the structure of minerals he refers to "Brøgger's well-known monograph on the pegmatite minerals of the Langesundfjord." Clearly Willie loved and was immersed in the study of minerals and crystallography while young and in Norway.

The invitation to the University of Chicago in 1930 was an effort on Compton's part to build up x-ray studies at Chicago. It is of interest to note that, at that time, Compton was not chairman of the Department of Physics. He built up the group in x-rays almost on his own. Willie arrived on campus on the same day as another new assistant professor active in x-ray studies: Samuel King Allison. Allison had also been invited to join the faculty by Compton. Allison already knew the campus since he had grown up in the University area and had received his Ph.D. from the University seven years earlier. The Zachs and the Allisons became the closest of friends—a friendship that lasted the rest of their lives. Also added to the faculty in 1930 as an instructor was Ernest O. Wollan. In addition to these four faculty members active in x-ray studies there were three postdoctorals: Marcel Schein from 1929–31 as a Rockefeller Foundation fellow, Elmar Dershem from 1929–42 as a research associate, and John H. Williams from 1931–33 as a National Research Council fellow. Williams had earlier worked with Allison at the University of California, Berkeley. In addition to the very close friendship with the Allisons, the Zachs also became close lifelong friends with the Scheins and John Williams. Willie had little respect for Compton as a person. Three of these seven—Allison, Compton, and Williams—worked on the physics of production of x-rays and on their interaction with matter. Dershem and Schein worked in similar areas, except with very soft x-rays. Wollan worked on the scattering of x-rays by gasses. Willie was alone in crystal structure determination. During the few years this group was together, almost all of their research was published independently. There were three exceptions, two papers by Dershem and Schein and one by Wollan and Compton. Compton and Allison did publish jointly an extensive and excellent reference book, *X-Rays in Theory and Experiment* (1934). In spite of the extensiveness of that book, it contained nothing on Willie's love, crystal structure. Shortly after that book was written, all except Willie changed their fields of research: Compton, Dershem, and Schein to cosmic rays; Allison, Williams, and Wollan to nuclear physics. Willie continued, single-mindedly, to determine what he could about the structure of matter using x-rays as a tool.

Willie had very little financial support for his work during the thirties. Aside from funds to support Compton's work (supported in part from the outside) and operating A. A. Michelson's two grating ruling engines (maintained during that period by the chairman of the department, Henry Gordon Gale), there was very little money left for others. Willie had to use homemade x-ray tubes built by his associate Dershem. When in 1938 his kenotron rectifier burned out, he had to shut down his research for six months until the department could find the $75 necessary to buy a new rectifier. All trips to meetings of the American Physical Society were at participants' expense. Willie had to make his own slides. A typical procedure for such trips was for Willie and Sam Allison to take one car, add as many of their students as possible and head for the meeting. If the meeting was to be in Washington, D.C., for example, they would stop overnight at the Gamma Alpha house (scientific fraternity) at the University of Ohio, and then at a cheap tourist house in Washington. This lack of support from the department of which he was a member continued until 1943.

In 1943 Willie joined the Manhattan Project. During the next two years he helped to unravel the chemistry, and to determine the crystal structure, of the transuranic elements and compounds. Many who were involved in that project feel that it would have taken much more time to do these jobs if it had not been for Willie's insight and genius in crystal structure determination.

Late in 1945 Willie first accepted administrative duties. His influence and effectiveness in these positions has positively affected many lives. In 1928, just two years before Willie went to the University of Chicago, a national survey had rated the Department of Physics of that university number one in the country. This was due in large part to the presence at that time of Michelson, Milliken, and Compton, three Nobel Prize winners. During the thirties, under the guidance of Gale and Compton, that rank slipped badly. This, according to Willie, was due primarily to the autocratic rule within the department, and to the hiring by the department of its own students as junior faculty, largely to assist the faculty member under whom that student had received the Ph.D. degree.

The changes Willie made were momentous and lasting. He immediately ended the domination of the department by Michelson's grating ruling engines by givng them away, one to Bausch and Lomb, and one to the Massachusetts Institute of Technology. He immediately turned the department from autocratic to democratic. The then tenured staff of the department met for the first time in many years to consider departmental affairs. Without much delay they voted to terminate the appointments of ten nontenured staff members. They also voted to reject as a member of the Department of Physics a new professor whom Compton had just hired, as usual without consulting anyone else in the department—a ticklish situation which Willie had to handle. A position for this professor was finally found in another part of the university.

Next, the staff voted to appoint Enrico Fermi and Edward Teller as professors, Robert F. Christy and Walter H. Zinn as associate professors, and John H. Simpson as instructor. It was Willie's job to invite these men to join the staff and to persuade them to accept. Willie did invite them, and all accepted. With this success, the Chemistry Department, partly due to Willie's needling, invited Willard F. Libby, Joseph E. Mayer, and Harold Urey. This enabled Willie, with the support of the physics faculty, to invite Maria G. Mayer to become a volunteer professor of physics, since the university's nepotism rules at that time forbade two members of one family holding faculty positions. She accepted. A bit later, with the support of the physics faculty, Willie invited Gregor Wentzel and others. The fact that all these outstanding physicists accepted positions at the University of Chicago is testimony to Willie's persuasiveness and the confidence which people put in his work and leadership. With this staff there was no trouble in attracting the most oustanding students in the country. By 1949 the department was once again rated tops in the nation, and among the Ph.D.'s graduated between 1945 and 1950 were five who were later awarded Nobel Prizes in physics.

As soon as Willie took over, with the unanimous support of the faculty, Willie introduced bylaws by which the department was to operate. According to these bylaws the department was no longer to be administered by a chairman who acted as a head, but by a true chairman. To Willie "chairman" meant that the person administering the department could do only those things that the faculty voted while the chairman occupied the chair at faculty meetings. All faculty were to vote on new appointments;

all faculty of higher rank on promotions. A policy committee, a budget committee, a curriculum committee, a services committee, etc., were established made up of faculty, by vote of the faculty. With this reorganization the old autocratic procedures of Gale and Compton were gone. As far as graduate students were concerned, Willie persuaded the faculty to accept the rule that all Ph.D. theses had to be published under the students' names alone. He felt that if a piece of research had not been done independently enough to justify publication by the student alone, it was not acceptable as establishing that the student could do independent research. Willie's standards were always very high. This rule held until the late 1960s when exceptions were made for students in high-energy physics, where a staff member's participation was required before that student could get access to a large accelerator.

Willie had a heart attack in January 1949 and a second attack four months later. With the second heart attack Willie resigned the chairmanship of the Department of Physics and slowed down a bit. He had published twenty-seven papers in 1948–49. Willie, however, was not one to walk on eggs. In 1954–55 he published fifteen papers. With the department once again going downhill, Willie was drafted once again to take over the chairmanship in 1956. Again he worked his magic. In 1959 the faculty of the Division of Physical Sciences persuaded him to take over as dean of the division. Again he did his magic, and the caliber and support of the faculty throughout the division improved. Having accomplished what he thought he could, he resigned as dean in 1962, two years before his term was up, and returned to his research. In 1970 Willie had a bad case of phlebitis. His best friend Sam Allison had died of complications from phlebitis a few years earlier. Two heart attacks and phlebitis were enough to cause him to rethink his future. He was just at retirement age. He was offered a special postretirement appointment but decided to accept that appointment only part time, so that he could spend some time "living." Since Willie and his wife, Mossa, had many friends from the Manhattan Project living in Los Alamos and its neighbor Santa Fe, New Mexico, they decided to move to Santa Fe. There they purchased the first home they had ever owned. Willie did as he had agreed; he and Mossa returned to the University of Chicago for one quarter of each year, for three years, to give one course. He kept quite active in research through his contacts with associates in the Los Alamos scientific laboratories, mainly his friends Finley H. Ellinger and Robert A. Penneman. He also worked with his friend and associate Bernd Matthias at the University of California, San Diego. He published several papers with each. He also spent time enjoying food, music, art, mystery stories, Indian lore, his home, and "living."

Willie was a superb teacher both at the graduate and undergraduate levels. I well recall one course I took from him in graduate-level classical mechanics in the late thirties. This course was considered one of the very best in the department at that time. Willie would enter the lecture room, place his notebook on the desk, and proceed to give a beautiful, understandable, and rigorous lecture. Then, after answering questions he would pick up his still-closed notebook and leave. Only once in that particular eleven-week course did he open that notebook to check on a formula he had derived. He decided that it was correct, though in a different form than his notes. He then closed his notebook and finished the lecture. He just never made mistakes. It was not until years later that I learned that such lectures were the result of careful preparation on his part. Some years later, when I was chairman of the Department of Physics, I asked Willie to give an introductory course in physics (physical science) for nonscience students at the undergraduate level. He agreed and did a superb job in this difficult assignment. Student evaluations were enthusiastic. He developed a close personal relationship with the roughly 150 students in his class. The course was given during the period of student protests in the late 1960s. The University of Chicago students were no different from any other college students, and students, including students from his class, took over the administration building of the university. Willie was one of a few faculty whom the students would let into that building. They obviously enjoyed conferring with him. He helped calm troubled waters.

Willie sponsored a number of students for their Ph.D. degrees, all of these in the period 1930–40. His Ph.D. students were John Albright, Donald A. Edwards, Ssu Mien Fang, Jane (Hall) Hamilton, Dorothy Heyworth, Richard C. Keen, Raymond Pepinsky, Stanley Siegel, Rose (Mooney) Slater, and G. E. Ziegler. He had two assistants while he was working for the Manhattan Project: Wallace Koehler and Ann Plettinger. Koehler stayed with Willie for a few years after World War II working toward a Ph.D., but he did not finish. Plettinger stayed with Willie as an assistant until he left Chicago. She was coauthor with Willie on nine of his post-World War II papers. Willie accepted no students seeking advanced degrees during or after World War II. He felt that the work he was doing was chemistry, not physics, and that it was not a suitable field of research for physicists. He was in a physics department. Willie did accept several postdoctorals who came to him with outside named fellowships. He used the criterion of outside support as one indication of the independence, competence, drive, and real interest of these people in the work he was doing. He felt that if he provided the support, he would get people who simply wanted a job and weren't good enough to get their own support, or a faculty position elsewhere.

During Willie's first twenty years at the University of Chicago his favorite vacation was a stay with his friend Sam Allison at Three Lakes in Wisconsin or, more frequently, a fishing-canoeing trip into the Lake of the Woods area in northern Minneosta with his friends from his early years in Chicago, Sam Allison and John Williams. Williams had gone to the Department of Physics at the University of Minnesota. The fourth person on these trips was generally Buddy Thorness, also from the University of Minnesota. Willie loved the woods, the water, the portages, the fishing, the cooking, and most of all the repartee with these close friends.

After Willie's two heart attacks in 1949 the frequency of these canoeing trips dropped off. For relaxation Willie then became a devotee of the billiard room of the faculty club of the University of Chicago for a short game after lunch. The favorite game soon became a frustrating game called "Cowboy Pool." It was the game of choice for Willie because it is a game that cannot be played by a person without a sense of humor. As his ofttimes partner in these games Julian Goldsmith has said:

> Willie was the leader of the group, made up of people of diverse and unrelated interests. He set the tone and developed refined rules, designed to eliminate the trivial and make the game more challenging—typical of Willie. He set the standard for gamesmanship, repartee, razzing, hexing, friendly insult, amateurism, and comradery that made winning or losing of little importance. His influence made the game a subtle Rorschach test, and the real nature of the players became quickly evident. Willie's humor and strength of personality pervaded the room. He had a rapier-like wit. He added to what may sound like a sterile activity. With Willie it wasn't.

THE SCIENCE

Willie was always interested in the structure of matter that x-rays could reveal. He was not a developer of new x-ray instrumentation or techniques. The techniques he learned while a student of Goldschmidt at Oslo University were the Laue (single crystal), the Debye-Scherer (powder), and the rotating crystal techniques, all of which used photographic recording. During his postdoctoral period with Sir Lawrence Bragg in Manchester University, Willie learned the Bragg technique of measuring the intensities of reflections from single crystals by means of ionization chambers, and the use of those measurements to derive Fourier diagrams (two-dimensional representations) of electron distributions within crystals. After World War II Willie adopted, and contributed to the development and use of, the single fixed crystal spectrometer using proportional counters for measurement of spatial intensity measurement. He did on a few occasions write papers in which he used neutron diffraction to determine the position of light elements within crystals, e.g., the rare earth hydroxides (1955) and MgH (1963). In these few cases Willie did the interpretation of the data, and his associates determined the neutron diffraction patterns.

Willie was also a very good chemist. His mentor Goldschmidt had prepared a number of the compounds and crystals that Willie used while a student in Oslo. Willie later prepared a number of compounds and crystals for his own use. After World War II, Willie had his own small chemistry laboratory. In that facility, among other things, he prepared a number of fluoride compounds and metaborates. He also grew crystals, e.g., of the metaborates, of sufficient size and perfection to do single crystal studies.

Mineralogical Crystals

As detailed earlier, Willie was born and grew up in Langesund, Norway, an area rich in well-crystalized minerals. He was intrigued from the very beginning with the structure of these minerals. His first published paper was on the crystal structure of BeO, which as a mineral is called Bromellite, named for the Swedish mineralogist who discovered it. His second paper was on Wurtzite (ZnS) and the related crystals α and β CdS. Most of Willie's first thirty-four papers concerned minerals or compounds of interest to mineralogists. Specifically, and in order, the minerals he studied after Wurtzite included Phenacite (Be_2SiO_4), Willemite (Zn_2SiO_4), Montroydite (HgO), GeO_2 isolated from Argyrodite, Bixbyite ($[Fe,Mn]_2O_3$), Titanite ($CaTiSiO_5$), Eudidymite and Epididymite ($NaBeSi_3O_7[OH]$), Thortveitite ($Sc_2Si_2O_7$), Benitoite ($BaTiSi_3O_9$), Hambergite ($Be_2BO_3[OH]$), and Colusite ($[Cu,Fe,Mo,Sn]_4[S,As,Te]_{3\text{-}4}$). Along with these structure determinations, Willie discussed the structure of a number of minerals having analogous structures and formulas. As a survey of these minerals shows, as time went on, Willie determined the structure of more and more complex minerals.

Inorganic Crystals

Willie's interest in the structure of inorganic crystals in general and the reasons for variations in those structures become apparent in his years in Oslo. He did not just study the mineral Wurtzite (ZnS), he also investigated the chemically similar crystals ZnSe and ZnTe; not just BeO but also BeS, BeSe, BeTe, and MgTe; not just CdS but also CdSe and CdTe; not just HgO but also HgS, HgSe, and HgTe. He also made systematic studies of sesquioxides (X_2O_3) and crystals of the form AXO_3 (1928,3). Such studies enabled him to make correlations and generalizations. One important piece of work of this type was his publication of tables of atomic radii. His first publication of a paper specifically on this topic was in 1931.

Some of Willie's extensive general and systematic studies of inorganic crystal structure are sufficiently distinct to be listed separately.

Groups in Crystals

As Willie said in the introduction to a number of his papers, "I have been interested in the determination of the shape and accurate dimensions of inorganic groups in crystals." His interest was not only in the shape and dimensions, but variations in those parameters for the same group, in different crystals, and the reasons for those variations. This interest first appears in print in his thesis where, among other things, he studied the shape of XO_3 groups. The interest continued throughout his career. His first paper specifically on the subject, "The Structure of Groups XO_3 in Crystals," was published in 1931. He published thirty-three more papers, which in large part were studies of groups. He distinguished groups from radicals in the sense that an XO_3 group in a long string of XO_3 groups in which O's are shared between all adjacent groups is a group, it is not a radical. He correlated the structure of these groups with the number of valence electrons in the group. In his 1931 paper on XO_3 groups he showed that XO_3 groups having twenty-four valence electrons, e.g., $(NO_3)^{-1}$, $(CO_3)^{-2}$, and $(BO_3)^{-3}$, have coplanar structure, while those having 26 valence electrons, e.g., $(PO_3)^{-3}$, $(SO_3)^{-2}$, $(ClO_3)^{-1}$, $(AsO_3)^{-3}$, $(ScO_3)^{-2}$, $(BrO_3)^{-1}$, and $(SbO_3)^{-3}$, are pyramidal. In a 1932 paper entitled "Note on a Relation Between the Atomic Arrangement in Certain Compounds, Groups and Molecules and the Number of Valence Electrons," he refined these ideas. He made predictions based on valency for XO_2 groups and, for example, predicted that the $(NO_2)^{-1}$ group in $NaNO_2$ would not be linear. He put his student G. E. Ziegler to the task of checking this prediction. It proved correct. Willie also pointed out that crystals whose chemical formulas are AXO_3

do not necessarily have XO_3 groups. He demonstrated this to be the case in $LiIO_3$, $NaIO_3$, and $CsIO_3$, where iodine occurs in IO_3 octahedra which share corners with one another.

In another series of systematic investigations Willie studied the structure of S_nO_m groups. Specifically, he determined the structure of sulphite $(SO_3)^{-2}$, pyrosulphite $(S_2O_5)^{-2}$, persulphate $(S_2O_8)^{-2}$, and trithionate $(S_3O_6)^{-2}$ during the period 1931–34. The structure of sulphate $(SO_4)^{-2}$ had been determined earlier by others, but controversy still existed about its structure in Na_2SO_4 and $AgSO_4$. Willie redetermined these structures. The dithionate group $(S_2O_6)^{-2}$ had been determined by others earlier. Based on such studies Willie showed that the pyrosulphite group should be written as SO_3SO_2 linked by a covalent bond between the two sulphurs, not as SO_2SSO_2, as had been previously assumed. Willie also showed that the persulphate group could be described as two SO_4 groups linked together by a covalent bond between two of the oxygen atoms SO_3OOSO_3 and that the trithionate group could be described as two SO_3 groups bonded to a common sulphur atom SO_3SSO_3. Such generalizations had obvious implications for later investigators who investigated other compounds containing these groups.

Borate Groups

The most extensive series of investigations of groups that Willie undertook was the determination of the structure of borate groups and of borates. This series of investigations continued through sixteen papers extending over thirty-three years. He introduced some of these papers in the later part of the series with the phrase, "This investigation is part of a systematic study of borate structures being carried on in this laboratory." As one might guess from Willie's history, the first borate Willie investigated was of a mineral, Hambergite, $Be_2(BO_3)(OH)$ (1931,1), in which he found the borate group to exist as an almost perfect O_3 triangle with the boron atom at its center. He reexamined this crystal in more detail in 1963. Willie also investigated boric acid H_3BO_3. Again he found an identifiable BO_3 triangle to exist, this time with a hydrogen bonded to each oxygen. The structure is thus better written as $B(OH)_3$. He then investigated a series of metaborates, i.e., crystals in which chemically the borate group is BO_2. The metaborates he studied included in order $Ca(BO_2)_2$, $KH_2(H_3O)_2(BO_2)_5$, $K(BO_2$, $\beta H(BO_2)$, $\beta H(BO_2)$, $Na(BO_2)$, and $Li(BO_2)$. Again the breadth and depth of Willie's interest in each problem are obvious. Willie had suggested quite early, i.e., after his study of $Ca(BO_2)_2$, that BO_2 groups do not exist in crystals. It surprised many to learn that no identifiable $(BO_2)^{-1}$ groups were found. The structures Willie did find in these crystals were varied and beautiful, and they clearly intrigued Willie. In $Ca(BO_2)_2$ he found the structure to consist of an endless chain of almost perfect BO_3 triangles with two oxygen atoms in each triangle shared with an adjacent triangle. He later showed how these chains were bound together by calcium atoms. He showed that the same structure existed in $Li(BO_2)$. In $K(BO_2)$, which Willie studied in 1931 and later in $Na(BO_2)$, he found the borate to again exist as a BO_3 triangle, but this time three triangles were formed into a six-membered ring of B_3O_3 with a third oxygen bound to each boron to complete the three BO_3 triangles making up the ring. In 1940 H. Tazaki found metaboric acid β-$H(BO_2)$, orthorhombic, to have the same borate structure Willie had found for $K(BO_2)$. Willie later studied the other two forms of metaboric acid. He found $\beta H(BO_2)$, monoclinic, to consist of chains of borate groups, two-thirds BO_3 triangles, and one-third BO_4 tetrahedra. In $\beta H(BO_2)$ cubic, he found all borons to be in tetrahedral configuration. Willie then went on to still more complicated metaborates. In $KH_2(H_3O)_2(BO_2)_5$

he found the borate structure to be a chain made up of pentaborate groups consisting of one BO_4 tetrahedron bound to four BO_3 groups, the groups sharing oxygens. He found $K_2B_4O_7(H_2O)$ to consist of $[B_4O_5(OH_4)]^{-2}$ radicals made up of two BO_4 tetrahedra and two BO_3 triangles having only corners in common. In Willie's last paper on metaborates, $LiBO_2$ (1964,2), he gave preliminary results on $LiBO_2 8H_2O$ and promised to give details later. Given more time he certainly would have done so. This example of one of Willie's areas of interest well illustrates the depth and breadth of each of Willie's investigators.

Atomic and Ionic Radii

In 1932 Willie published a paper entitled "A Set of Empirical Crystal Radii for Ions with Inert Gas Configuration," in which he compared empirically calculated values for these parameters with x-ray measurements. This paper was a needed improvement of work done by Goldschmidt and by Pauling. In the paper Willie takes into account coordination number (number of nearest neighbors in the lattice), valence, and radius ratio. Starting with this paper, Willie returned again and again in later papers to refine his self-consistent table of atomic ionic radii. Such data are important in correlating the behavior of differential chemical elements and in making predictions of the properties of substances whose crystal structure has not yet been determined. Willie's last paper specifically on the subject was published in 1973, "Metallic Radii and Electron Configuration of the 5f-6d Metals."

Glass

In 1932, i.e., during Willie's first years at the University of Chicago, he departed from his study of crystals to give the first correct description of the structure of glass. He never considered this work as very important and, on at least one occasion, had to be reminded of the work before he recalled that at one time he had written about the subject. Those working on glass, however, consider his 1932 paper, which he expanded somewhat in a German version in 1933, to be the starting point of the real understanding of glass. Charles H. Green in a 1961 article on glass in *Scientific American* said, "The present day understanding of glass rest heavily on a single lucid paper, only twelve pages long, written in 1932 by William H. Zachariasen." Alfred R. Cooper in his introductory paper to the 1980 Borate Glass Conference said, "We dedicate this session on glass structure to Frederik William Holder Zachariasen because his single contribution to glass literature, 'The Atomic Arrangement of Glass,' may be the most influential paper on glass structure in this century."

Before Willie's paper on glass it was said that glass consists of crystalline materials. More precisely, the main features of the x-ray patterns of glass could be explained on the basis that vitreous silica consists of cristobalite crystallites, having dimensions of about 15Å and a lattice constant about 6.6 percent greater than in crystalline silica. Willie argued that this description could not explain the properties of glass. He then proposed a very different structure using oxide glasses as an example. Specifically, he suggested that glasses are made up of oxide groups AO_n satisfying the following four rules: (1) An oxygen atom is linked to not more than two atoms A; (2) The number of an oxygen atoms surrounding atoms A must be small (refined in the next paragraph to triangular or tetrahedral configuration for known glasses); (3) The oxygen polyhedra share corners with each other, not faces (this leads to random orientation of adjacent groups and hence no long-range order, i.e., no crystalline structure); (4) At least three corners in each oxygen polyhedron must be shared. Based on these rules, Willie was able to predict compounds which would produce a glass as well as explain many of the properties of glass.

Willie's paper on glass has appeared to many who have written about it to have come out of the blue. However, Willie had discussed crystals of the rutile type XO_2 in his 1926 paper with Goldschmidt, and some silicates in his 1930 paper with Bragg. From this work he had some concept of how SiO_4 groups associate. As I have detailed in the previous section, he had also studied how borate groups fit together in a variety of ways. Thus Willie's paper on the structure of glass was simply an insightful extension of his earlier studies of groups.

Liquid Structure

Willie published one paper on "The Liquid 'Structure' of Methyl Alcohol" one year after his classic paper on the vitreous state (glass). Methyl alcohol was particularly interesting to Willie since alcohols have a tendency to form a glass. The results were consistent both with Willie's papers on the structure of glass and with Bertrum Warren's x-ray studies of the liquid state. Warren had been a postdoctoral with Sir Lawrence Bragg at the same time as Willie.

The 5f Elements

Perhaps Willie's most celebrated work was the elucidation of the nature and the chemistry of the transuranic elements. His work in this area, started in 1943 when he joined the Manhattan Project, was extremely important to the Manhattan Project. He continued to do some work in this area throughout the rest of his life. Robert Penneman of the Los Alamos Scientific Laboratory has said of this work, "No other crystalographer has done so much to expand our knowledge of heavy element chemistry, or had such a central role in the early development of atomic energy." The initial problem faced in the understanding of the chemistry and structure of the transuranics becomes clear when one recalls that during the early stages of the Manhattan Project, only microgram quantities of these artificially produced elements were available. This meant that the chemistry of these elements, including the important separation processes for plutonium from its host materials, had to be determined using only these amounts. It is indeed difficult to determine the chemical composition of a sample using ordinary chemical techniques when only microgram amounts of that chemical are present. The procedure adopted involved the chemists doing microchemistry on these samples, and then sending them in capillaries to Willie to find out what they had produced. As it turned out during the early stages of the Manhattan Project, in many cases the compounds produced were a complete surprise. As these studies continued, and more detailed information was obtained, the concept of these elements being a 5f series of elements, analogous to the 4f series of elements in the rare earths, grew. The experimental evidence for this concept rested in large part on Willie's work. He found from his x-ray studies that the radii of successive transthorium elements in isostructural compounds decreased slowly, i.e., by about 0.03Å per successive element, much as the radii of the rare earth elements decrease slowly, by about 0.03Å per successive element. This was the first strong evidence for the 5f character of these elements. Willie's studies were crucial in the development of the metallurgy of the transuranium elements, particularly in the important case of plutonium. Within a few months of the preparation of the first milligrams of metal, Willie recognized that the metal had several phases stable at near-normal temperature and pressure (now a total of six). He solved the extremely complicated structure of α-plutonium with only slide rule and insight. The x-ray pattern is complicated since the first seventeen x-ray reflections are absent. Others had failed to solve this structure using the largest computers then available.

In 1952 Willie put forward one suggestion concerning these elements which has proved to be incorrect. He suggested that the 5f elements should be called thorides rather than actinides. The argument is in essence, are the elements predominantly trivalent, or are they tetravalent? In a 1961 paper that discussed the question, he said, "It is frankly admitted that the conclusions presented in this paper are somewhat speculative, and that the results as to electronic configuration ought to be based on physical properties which depend more directly on electronic interactions." His original reasoning in making this suggestion was based on consideration of the atomic radii exhibited by these metals. He had determined these radii, and they were just the ones one would expect for tetravalent metals with five or six electrons *if* the 5f electrons do not contribute to bonding, an assumption generally held at the time. Later theoretical work using large computers showed that this assumption was not generally valid. That work and later experimental results on superconductivity of these elements appear to have convinced Willie shortly before his death that this one suggestion was incorrect.

One of the most beautiful verifications of the similarity in electronic structure of the 4f and 5f elements was the discovery in 1974 by Willie and Penneman that above 56 kbars, cerium has the same crystal structure as α-uranium. Before their insightful discovery, α-uranium had a structure that was unique.

Diffraction Theory

Willie first felt the lack of adequate theory when in 1940 he wrote a paper on "A Theoretical Study of the Diffuse Scattering of X-rays by Crystals." He followed this paper with five other papers on the subject, including careful experimental checks. He assigned his student Stanley Siegel the job of completing these checks. Siegal's work was published in 1941. Partly as a result of this work, Willie took on the task of going over the then extant theory. This effort resulted in his classic 1945 monograph, *Theory of X-ray Diffraction in Crystals.* The succinctness of this book, as with many of Willie's publications, is illustrated by the story related by Wallace C. Koehler, Willie's student at the time. According to Koehler, after spending considerable time trying to get from one equation to the next, separated by the phrase "from this it follows," he asked Willie how he did it. Willie replied, "Don't worry, it took me two days to make that step." As this story illustrates, the book is used mainly by experts in the area. Many more recent papers are little more than direct expansions of paragraphs from Willie's book.

One problem that for many years made precise crystal structure determinations difficult was the lack of a method for determining the phase of structure factors. In 1952 Willie developed a method for determining the phases directly from the measured intensities. He immediately tested his method experimentally by applying it to metaboric acid. From that time his technique was refined until by 1975 over half of the structures being determined by x-ray techniques were solved by the "direct method" traceable to Willie's work.

Another problem that long plagued precise structure determination was a discrepancy between the calculated and measured intensities of diffracted x-ray beams. In 1963 Willie started looking at this problem in more detail. As one might guess from his history, his careful look at the problem began with a mineral, Hambergite, in which he found his carefully measured intensities to be incompatible with theory. In a reconsideration of the theory, he showed that C. G. Darwin's formula for the secondary extinction correction, which had been universally accepted and extensively used, contained an appreciable error when applied to x-rays. The error was in the treatment of the polarization of the x-ray beam. In 1963 Willie published a first-order approximation for the extinction correction for a mosaic crystal of arbitrary shape. In 1965 he published two more theoretical papers in which he derived more precise formulae for extinction and multiple diffraction. Shortly thereafter he published an experimental test of his new theory using a small quartz sphere. These papers also took into account corrections necessary in highly absorbing crystals for the Borrmann effect. Extinction becomes more serious as the scattered intensity increases. Scattering close to the incident beam is generally the most intense. It is just these low-angle beams which are important in the determination of valence electron distribution. Without a precise method for taking extinction into account, determination of outer electron distributions are unreliable. As Pepinsky said in 1975, these papers on extinction and the Borrmann effect are "a landmark in diffraction theory and broke the dam which had held back structure determination. The pathway is now open to new attacks on the problem of bonding electron distribution in simple structures, to far more accurate complex structure analyses, and eventually to bonding electron structure in these complex structures."

Melting Points

Willie published one paper in 1967, jointly with his friend Matthias and two of Matthias's associates, on "Melting-Point

Anomalies." As these authors said in their paper, "At present there is not even a satisfactory beginning of a macroscopic theory of melting. Speculative discussions, such as those given in this paper, are hence justified because they may suggest directions for fruitful theoretical exploration." Their suggestions involved the "partial f character of the hybridized wave function of the valence band," in essence the effective number of free electrons. Willie and his friend Matthias continued to think about this problem until Willie's death, but published nothing more.

Superconductivity

With his friend Bernd Matthias and some of Matthias's associates, Willie was coauthor of seven papers on superconductivity. During their years together at Chicago (1946-48), while Matthias was an assistant professor and Willie chairman, they had many fruitful discussions, some of which contributed to Matthias's later correlations of the superconductivity critical temperature to the effective number of valence electrons per atom. In later collaborations, among other things, Willie "interpreted correctly the nonstoichiometric phases present in superconducting multicomponent mixtures." As these authors conclude, "The position of the elements in the periodic table, the valence electron concentration and the crystallographic structure exhibit a strong influence on the superconducting behavior." Willie's influence is clear.

SUMMARY

Willie's contributions to science, and to the scientific community, have been rich and varied.

IN PREPARING THIS MEMOIR I have drawn freely from memorials and accolades written by S. Chandraesekhar, A. R. Cooper, J. R. Goldsmith, M. Marezio, B. T. Matthias, P. B. Moore, Linus Pauling, R. A. Penneman, Raymond Pepinsky, D. H. Templeton, and Anthony Turkevitch, and from a recorded conversation between Willie and Edward Wolowiec in which Willie recalls some of his history while in Chicago.

SELECTED BIBLIOGRAPHY

1925

Üeber die kristallstruktur von BeO. *Nor. Geol. Tidsskr.* 8:189–200.

With F. Ulrich. Ueber die kristallstruktur des α- and β-CdS, sowie des wurtzits. *Z. Kristallogr.* 62:260–73.

Die kristallstruktur der telluride von zink, cadmium und quecksilber. *Nor. Geol. Tidsskr.* 8:302–6.

1926

Die kristallstruktur von berylliumoxyd und berylliumsulfid. *Z. Phys. Chem.* 119:201–13.

Notiz ueber die kristallstruktur von phenakit, willemit und verwandten verbindungen. *Nor. Geol. Tidsskr.* 9:65–73.

Die kristallstruktur der A-modifikation von den sesquioxygen der seltenen erdmetalle. (La_2O_3, Ce_2O_3, Pr_2O_3, Nd_2O_3). *Z. Phys. Chem.* 123:134–50.

Üeber die kristallstruktur der telluride von beryllium, zink, cadmium und quecksilber. Mit prazisionsbestimmungen der gitterkonstanten. *Z. Phys. Chem.* 124:277–84.

Beitrag zur frage nach dem ionisationszustand der atome im raumgitter des berylliumoxyds. *Z. Phys. Chem.* 40:637–41.

Üeber die kristallstruktur der selenide von beryllium, zink, cadmium und quecksilber. Mit prazisionsbestimmungen der gitterkonstanten. *Z. Phys. Chem.* 124:436–48.

With V. M. Goldschmidt, T. Barth, D. Holmsen, and G. Lund. Geochemical distribution law on the elements. VI. Crystal structure of the rutile type with remarks of the geochemistry of the bivalent and quadrivalent elements. *Nor. Vidensk. Akad.–Oslo Arbok.* 1:1–21.

With V. M. Goldschmidt, T. Barth, and G. Lund. Geochemical distribution law of the elements. VII. Summary of the chemistry of crystals. *Nor. Vidensk. Akad.–Oslo Arbok.* 2:1–117.

1927

The crystal structure of the modification C of the sesquioxides of the rare earth metals, and of indium and thalium. *Nor. Geol. Tidsskr.* 9:310–16.

The crystal structure of $MoSi_2$ and WSi_2. *Nor. Geol. Tidsskr.* 9:337–42.

Die kristallstruktur des ammoniumfluorids. *Z. Phys. Chem.* 127:218–24.
Ueber die kristallstruktur von $MoSi_2$ und WSi_2. *Z. Phys. Chem.* 128:39–48.
Ueber die kristallstruktur des palladiumoxyds (Pd0). *Z. Phys. Chem.* 128:412–16.
Ueber die kristallstruktur des magnesiumtellurids. *Z. Phys. Chem.* 128:417–20.
Ueber die kristallstruktur des quecksilberoxyds. *Z. Phys. Chem.* 128:421–29.

1928

The crystal structure of tetramethylammonium iodide. *Nor. Geol. Tidsskr.* 10:14–22.
Ueber die kristallstruktur des wasserloslichen modifikation des germaniumdioxyds. *Z. Kristallogr.* 67:226–34.
Untersuchungen ueber die kristallstrukturen von sesquioxygen and verbindungen ABO_3. *Nor. Vidensk. Akad.–Oslo Arbok.* 4:1–165. (Ph.D. diss.)
Ueber die kristallstruktur von bixbyit, sowie vom küntlichen Mn_2O_3. *Z. Kristallogr.* 67:455–64.

1929

Bemerkungen zu der arbeit von L. Pauling: The crystal structure of the A-modification of the rare earth sesquioxides. *Z. Kristallogr.* 70:187–89.
Notiz üeber die kristallstruktur von titanit. *Nor. Geol. Tidsskr.* 10:209–12.
The crystal structure of potassium chlorate. *Z. Kristallogr.* 71:501–16.
The crystal structure of sodium chloride. *Z. Kristallogr.* 71:517–29.
Die feinbäuliche relation zwischen eudidymit und epididymit. *Nor. Geol. Tidsskr.* 10:449–53.

1930

With W. L. Bragg. The crystalline structure of phenacite, Be_2SiO_4 and willemite, Zn_2SiO_4. *Z. Kristallogr.* 72:518–28.
The structure of thortveitite, $Sc_2Si_2O_7$. *Z. Kristallogr.* 73:1–6.
The crystal structure of titanite. *Z. Kristallogr.* 73:7–16.
The crystal structure of sodium perchlorate, $NaClO_4$. *Z. Kristallogr.* 73:141–46.
The chemical formula of the 'zircon pyroxenes' and the 'zircon pectolite.' *Nor. Geol. Tidsskr.* 11:1–3.

Bemerkung zu der arbeit B. Goszner und F. Muszgnug: Üeber die strukturelle und moleculare einheit von eudialyt. *Centr. Bl. Mineral* 7:315–17.

The crystal structure of benitoite, $BaTiSi_3O_9$. *Z. Kristallogr.* 74:139–46.

On meliphanite and leucophanite. *Z. Kristallogr.* 74:226–29.

1931

The crystalline structure of hambergite, $Be_2BO_3(OH)$. *Z. Kristallogr.* 76:289–302.

Note on the structure of groups in crystals. *Phys. Rev.* 37:775–76.

The structure of groups XO_3 in crystals. *J. Am. Chem. Soc.* 53:2123–30.

With H. E. Buckley. The crystal lattice of anhydrous sodium sulphite, Na_2SO_3. *Phys Rev.* 37:1295–1305.

With F. A. Barta. Crystal structure of lithium iodate. *Phys. Rev.* 37:1626–30.

With G. E. Ziegler. The crystal structure of potassium chromate, K_2CrO_4. *Z. Kristallogr.* 80:164–73.

A set of empirical crystal radii for ions with inert gas configuration. *Z. Kristallogr.* 80:137–53.

Meliphanite, leucophanite and their relation to melitite. *Nor. Geol. Tidsskr.* 12:577–82.

On the interpretation of the selective photoelectric effect from two component cathodes. *Phys. Rev.* 38:2290.

The crystal lattice of calcium metaborate, CaB_2O_4. *Proc. Natl. Acad. Sci. USA* 17:617–19.

1932

The crystal lattice of potassium pyrosulphite, $K_2S_2O_5$, and the structure of the pyrosulphite group. *Phys. Rev.* 40:113–14.

The atomic arrangement in glass. *J. Am. Chem. Soc.* 54:3841–51.

With G. E. Ziegler. The crystal structure of anhydrous sodium sulphate, Na_2SO_4. *Z. Kristallogr.* 81:92–101.

Note on a relation between the atomic arrangement in certain compounds, groups and molecules and the number of valence electrons. *Phys. Rev.* 40:914–16.

The crystal lattice of germano sulphide, GeS. *Phy. Rev.* 40:917–22.

The crystal lattice of potassium pyrosulphite, $K_2S_2O_5$, and the structure of the pyrosulphite group. *Phys. Rev.* 40:923–35.

Note on the crystal structure of silver sulphate, Ag_2SO_4. *Z. Kristallogr.* 82:161–62.

With G. E. Ziegler. The crystal structure of calcium metaborate, CaB_2O_4. *Z. Kristallogr.* 83:354–61.

Die struktur der glasser. Gortschr. *Mineral. Kristallogr. Petrog.* 17:451–52.

1933

The crystal lattice of sodium bicarbonate, $NaHCO_3$. *J. Chem. Phys.* 1:634–39.

Calculation of the refractive indices of sodium bicarbonate from the atomic arrangement. *J. Chem. Phys.* 1:640–42.

Die strukture der glaser. *Glastech. Ber.* 11:120–23.

X-ray examination of colusite, $(Cu, Fe, Mo, Sn)_4$ $(S, As, Te)_{3-4}$ *Am. Mineral.* 18:534–37.

1934

Note on the structure of the trithionate group, $(S_3O_6)^{-2}$. *J. Chem. Phys.* 2:109–11.

With R. C. L. Mooney. The structure of hypophosphite group as determined from the crystal lattice of ammonium hypophosphite. *J. Chem. Phys.* 2:34–37.

With R. C. L. Mooney. The atomic arrangement in ammonium and cesium persulpate, $(NH_4)_2S_2O_8$ and $Cs_2S_2O_8$, and the structure of the persulphate group. *Z. Kristallogr.* 88:63–81.

The crystal lattice of boric acid, BO_3H_3. *Z. Kristallogr.* 88:150–61.

The crystal lattice of oxalic acid dihydrate, $H_2C_2O_4 \cdot 2H_2O$ and the structure of the oxalate radical. *Z. Kristallogr.* 89:442–47.

The atomic arrangement in potassium trithionate crystals $K_2S_3O_6$ and the structure of the trithionate radical $(S_3O_6)^{-2}$. *Z. Kristallogr.* 89:529–37.

1935

The liquid "structure" of methyl alcohol. *J. Chem. Phys.* 3:158–61.

The vitreous state. *J. Chem. Phys.* 3:162–63.

Note on the scattering of x-rays from fluids containing polyatomic molecules. *Phys. Rev.* 47:277–78.

Note on the crystal lattice of samarium sulphate octohydrate. *J. Chem. Phys.* 3:197–98.

1936

The crystal structure of germanium disulphide. *J. Chem. Phys.* 4:618–19.

1937

The crystal structure of potassium acid dihydronium pentaborate $KH_2(H_3O)_2B_5O_{10}$ (potassium pentaborate tetrahydrate). *Z. Kristallogr.* 98:266–74.

The crystal structure of potassium metaborate, $K_3(B_3O_6)$. *J. Chem. Phys.* 5:919–22.

1938

Comments on the article by A. P. R. Wadlund: Radial lines in Laue spot photographs. *Phys. Rev.* 53:844.

1940

A theoretical study of the diffuse scattering of x-rays by crystals. *Phys. Rev.* 57:597–602.

The crystal structure of sodium formate, $NaHCO_2$. *J. Am. Chem. Soc.* 62:1011–13.

With S. Siegel. Preliminary experimental study of new diffraction maxima in x-ray photographs. *Phys. Rev.* 57:795–97.

Diffraction maxima in x-ray photographs. *Nature* 145:1019.

1941

On the diffuse x-ray diffraction maxima observed by C. V. Raman and P. Nilakantan. *Phys. Rev.* 59:207–8.

On the theory of the temperature diffuse scattering. *Phys. Rev.* 59:766.

Temperature diffuse scattering of a simple cubic lattice. *Phys. Rev.* 59:860–66.

The temperature diffuse scattering maxima for rocksalt. *Phys. Rev.* 59:909.

On the theory of temperature diffuse scattering. *Phys. Rev.* 60:691.

1945

Theory of X-ray Diffraction in Crystals. New York: John Wiley and Sons. 252 pp.

1947

Direct determination of stacking disorder in layer structures. *Phys. Rev.* 71:715–17.

1948

The UCl_3 type of crystal structure. *J. Chem. Phys.* 16:254.

The crystal structure of U_2F_9. *J. Chem. Phys.* 16:425.
Crystal radii of the heavy elements. *Phys. Rev.* 73:1104–5.
Double fluorides of potassium or sodium with uranium, thorium, or lanthanum. *J. Am. Chem. Soc.* 70:2147–51.
The crystal structure of the normal orthophosphates of barium and strontium. *Acta Crystallogr.* 1:263–65.
Crystal chemical studies of the 5f-series of elements. I. New structure types. *Acta Crystallogr.* 1:265–68.
Crystal chemical studies of the 5f-series of elements. II. The crystal structure of Cs_2PuCl_6. *Acta Crystallogr.* 1:268–69.
Crystal chemical studies of the 5f-series of elements. III. A study of the disorder in the crystal structure of anhydrous uranyl fluoride. *Acta Crystallogr.* 1:277–81.
Crystal chemical studies of the 5f-series of elements. IV. The crystal structure of $Ca(UO_2)O_2$ and $Sr(UO_2)O_2$. *Acta Crystallogr.* 1:281–85.
Crystal chemical studies of the 5f-series of elements. V. The crystal structure of uranium hexachloride. *Acta Crystallogr.* 1:285–87.

1949

Crystal chemical studies of the 5f-series of elements. VI. The Ce_2S_3-Ce_3S_4 type of structure. *Acta Crystallogr.* 2:57–60.
Crystal chemical studies of the 5f-series of elements. VII. The crystal structure of Ce_2O_2S, La_2O_2S and Pu_2O_2S. *Acta Crystallogr.* 2:60–62.
Crystal chemical studies of the 5f-series of elements. VIII. Crystal structure studies of uranium silicides and of $CeSi_2$, $NpSi_2$ and $PuSi_2$. *Acta Crystallogr.* 2:94–99.
Crystal chemical studies of the 5f-series of elements. IX. The crystal structure of Th_7S_{12}. *Acta Crystallogr.* 2:228–91.
Crystal chemical studies of the 5f-series of elements. X. Sulfides and oxy-sulphides. *Acta Crystallogr.* 2:291–96.
Crystal chemical studies of the 5f-series of elements. XI. The crystal structure of α-UF_5 and of β-UF_5. *Acta Crystallogr.* 2:296–98.
Crystal chemical studies of the 5f-series of elements. XII. New compounds representing known structure types. *Acta Crystallogr.* 2: 388–90.
Crystal chemical studies of the 5f-series of elements. XIII. The crystal structure of U_2F_9 and $NaTh_2F_9$. *Acta Crystallogr.* 2:390–93.

Crystal chemical studies of the 5f-series of elements. *Rec. Chem. Prog.* (Spring):47–51.

With R. C. L. Mooney. Crystal structure studies of oxides of plutonium. In *The Transuranium Elements.* National Nuclear Energy Series, vol. 14B, pp. 1442–7. New York: McGraw-Hill.

The crystal structure of plutonium nitride and plutonium carbide. In *The Transuranium Elements.* National Nuclear Energy Series, vol. 14B, pp. 1448–51. New York: McGraw-Hill.

The crystal structure of $PuSi_2$. In *The Transuranium Elements.* National Nuclear Energy Series, vol. 14B, pp. 1451–53. New York: McGraw-Hill.

Crystal structure studies of sulfides of plutonium and neptunium. In *The Transuranium Elements.* National Nuclear Energy Series, vol. 14B, pp. 1454–61. New York: McGraw-Hill.

X-ray diffraction studies of the fluorides of plutonium and neptunium: Chemical identity and crystal structure. In *The Transuranium Elements.* National Nuclear Energy Series, vol. 14B, pp. 1462–72. New York: McGraw-Hill.

Crystal structure studies of chloride, bromides, and iodides of plutonium and neptunium. In *The Transuranium Elements.* National Nuclear Energy Series, vol. 14B, pp. 1473–85. New York: McGraw-Hill.

The crystal structure studies of sodium plutonyl and sodium neptunyl acetates. In *The Transuranium Elements.* National Nuclear Energy Series, vol. 14B, pp. 1486–88. New York: McGraw-Hill.

The crystal structure NpO_2 and NpO. In *The Transuranium Elements.* National Nuclear Energy Series, vol. 14B, pp. 1489–91. New York: McGraw-Hill.

1950

With S. Fried and F. Hagemann. The preparation and identification of some pure actinium compounds. *J. Am. Chem. Soc.* 72:771–75.

With R. Elson, S. Fried, and P. Sellers. The tetravalent and pentavalent states of protactinium. *J. Am. Chem. Soc.* 72:5791.

1951

Crystal chemical studies of the 5f-series of elements. XIV. Oxyfluorides, XOF. *Acta Crystallogr.* 4:231–36.

1952

Crystal chemical studies of the 5f-series of elements. XV. The crystal structure of plutonium sesquicarbide. *Acta Crystallogr.* 5: 17–19.

Crystal chemical studies of the 5f-series of elements. XVI. Identification and crystal structure of protactinium metal and of protactinium monoxide. *Acta Crystallogr.* 5:19–21.

Crystal chemical studies of the 5f-series of elements. XVII. The crystal structure of neptunium metal. *Acta Crystallogr.* 5: 660–64.

Crystal chemical studies of the 5f-series of elements. XVIII. Crystal structure studies of neptunium metal at elevated temperatures. *Acta Crystallogr.* 5:664–67.

A new analytical method for solving complex crystal structures. *Acta Crystallogr.* 5:68–73.

On the anomalous transparency of thick crystals to X-rays. *Proc. Natl. Acad. Sci. USA* 38:378–82.

Experimental crystallography. *Annu. Rev. Phys. Chem.* 3:359–74.

1953

Crystal chemical studies of the 5f-series of elements. XIX. The crystal structure of the higher thorium hydride, Th_4H_{15}. *Acta Crystallogr.* 6:393–95.

With F. H. Ellinger. The crystal structure of samarium metal and of samarium monoxide. *J. Am. Chem. Soc.* 75:5650–52.

1954

Crystal chemistry of the 5f-series elements. In *The Actinide Elements*, ed. Seaborg and Katz, National Nuclear Energy Series, vol. 14A, pp. 769–96. New York: McGraw-Hill.

With R. N. R. Mulford and F. H. Ellinger. A new form of uranium hydride. *J. Am. Chem. Soc.* 76:297–98.

With F. H. Ellinger. The crystal structure of $KPuO_2CO_3$, $NH_4PuO_2CO_3$, and $RbAmO_2CO_3$. *J. Phys. Chem.* 58:405–8.

The precise structure of orthoboric acid. *Acta Crystallogr.* 7:305–10.

With L. B. Asprey and F. H. Ellinger. Preparation, identification and crystal structure of a pentavalent americium compound, $KAmO_2F_2$. *J. Am. Chem. Soc.* 76:5235–37.

With P. A. Sellers, S. Fried, and R. E. Elson. The preparation of some protactinium compounds and the metal. *J. Am. Chem. Soc.* 76:5935–8.

Crystal chemical studies of the 5f-series of elements. XX. The crystal structure of tri-potassium uranyl fluoride. *Acta Crystallogr.* 7: 783–87.

Crystal chemical studies of the 5f-series of elements. XXI. The crystal structure of magnesium orthouranate. *Acta Crystallogr.* 7: 788–91.

Crystal chemical studies of the 5f-series of elements. XXII. The crystal structure of K_3UF_7. *Acta Crystallogr.* 7:792–94.

Crystal chemical studies of the 5f-series of elements. XXIII. On the crystal chemistry of uranyl compounds and of related compounds of transuranic elements. *Acta Crystallogr.* 7:795–99.

1955

With L. B. Asprey, and F. H. Ellinger, and S. Fried. Evidence for quadrivalent curium: X-ray data on curium oxides. *J. Am. Chem. Soc.* 77:1707.

With F. H. Ellinger, C. E. Holley, Jr., B. B. McInteer, D. Pavonee, R. M. Potter, and E. Staritzsky. The preparation and some properties of magnesium hydride. *J. Am. Chem. Soc.* 77:2647.

With F. H. Ellinger. Crystal chemical studies of the 5f-series of elements. XXIV. The crystal structure and thermal expansion of γ-plutonium. *Acta Crystallogr.* 8:431–33.

With C. E. Holley, Jr., R. N. R. Mulford, F. H. Ellinger, and W. C. Koehler. The crystal structure of some rare earth hydrides. *J. Phys. Chem.* 59:1226–28.

With S. Fried. The chemistry and crystal chemistry of heavy element compounds. International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland.

1957

With F. H. Ellinger. Crystal structure of alpha-plutonium metal. *J. Chem. Phys.* 27:811–12.

With L. B. Asprey, and F. H. Ellinger, and S. Fried. Evidence for quadrivalent curium. II. Curium tetrafluoride. *J. Am. Chem. Soc.* 79:5825.

1958

With B. T. Matthias. Superconductivity of rhenium nitride. *J. Phys. Chem. Solids* 7:98.

With B. T. Matthias and E. Corenzwit. Superconductivity and ferromagnetism in isomorphous compounds. *Phys. Rev.* 112:89.

1959

With F. H. Ellinger. Unit cell and thermal expansion of β-plutonium metal. *Acta Crystallogr.* 12:175–76.

With H. A. Plettinger. Crystal chemical studies of the 5f-series of elements. XXV. The crystal structure of sodium uranyl acetate. *Acta Crystallogr.* 12:526–30.

On the crystal structure of protactinium metal. *Acta Crystallogr.* 12:698–99.

1960

With D. B. McWhan, J. C. Wallman, B. B. Cunningham, L. B. Asprey, and F. H. Ellinger. Preparation and crystal structure of americium metal. *J. Inorg. Nucl. Chem.* 15:185–87.

1961

With H. A. Plettinger. The crystal structure of lithium tungstate. *Acta Crystallogr.* 14:229–30.

With M. Marezio and H. A. Plettinger. The crystal structure of gadolinium trichloride hexahydrate. *Acta Crystallogr.* 14:234–36.

The structure of plutonium metal. In *The Metal Plutonium*, eds. A. S. Coffinbery and W. N. Miner, pp. 99–107. Chicago: University of Chicago Press.

1963

With C. E. Holley, Jr., and J. F. Stamper, Jr. Neutron diffraction study of magnesium deuteride. *Acta Crystallogr.* 16:352–53.

With F. H. Ellinger. The crystal structure of beta plutonium metal. *Acta Crystallogr.* 16:369–75.

With H. A. Plettinger. Refinement of the structure of potassium pentaborate tetrahydrate. *Acta Crystallogr.* 16:376–79.

The crystal structure of cubic metaboric acid. *Acta Crystallogr.* 16:380–84.

The crystal structure of monoclinic metaboric acid. *Acta Crystallogr.* 16:385–89.

With M. Marezio and H. A. Plettinger. Refinement of the calcium metaborate structure. *Acta Crystallogr.* 16:390–92.

With M. Marezio and H. A. Plettinger. The bond lengths in the sodium metaborate structure. *Acta Crystallogr.* 16:594–95.

With F. H. Ellinger. The crystal structure of alpha plutonium metal. *Acta Crystallogr.* 16:777–83.

Interpretation of monoclinic powder X-ray diffraction patterns. *Acta Crystallogr.* 16:784–88.

With M. Marezio and H. A. Plettinger. The crystal structure of potassium tetraborate tetrahydrate. *Acta Crystallogr.* 16:975–80.

The secondary extinction correction. *Acta Crystallogr.* 16:1139–44.

With H. A. Plettinger and M. Marezio. The structure and birefringence of hambergite, $Be_2BO_3(OH)$. *Acta Crystallogr.* 16:1144–46.

The crystal structure of palladium diphosphide. *Acta Crystallogr.* 16:1253–55.

With Ch. J. Raub, T. H. Geballe, and B. T. Matthias. Superconductivity of some new Pt-metal compounds. *J. Phys. Chem. Solids* 24:1093–100.

1964

Plutonium metal. In *The Law of Mass-Action: A Centenary Volume*, pp. 185–94. Det Norske Videnskaps-Akakemi I Oslo. Oslo: Universitetsforlanget.

The crystal structure of lithium metaborate. *Acta Crystallogr.* 17:749–51.

1965

Experimental differentiation between primary and secondary extinction with application to radiation disorder in sodium chlorate. *Acta Crystallogr.* 18:703–5.

Multiple diffraction in imperfect crystals. *Acta Crystallogr.* 18:705–10.

With H. A. Plettinger. Extinction in quartz. *Acta Crystallogr.* 18:710–14.

Dispersion in quartz. *Acta Crystallogr.* 18:714–16.

With F. H. Ellinger. The crystal structures of $PuGa_4$ and $PuGa_6$. *Acta Crystallogr.* 19:281–83.

Extinction. *Trans. Am. Crystallogr. Assoc.* 1:33–41.

1966

The crystal structure of Rh_2Te_3. *Acta Crystallogr.* 20:334–36.

With T. H. Geballe, B. T. Matthias, K. Andres, E. S. Fisher, and T. F. Smith. Superconductivity of alpha-uranium and the role of 5f electrons. *Science* 152:755–57.

With Robert Benz. Th_3N_4 crystal structure and comparison with that of Th_2N_2O. *Acta Crystallogr.* 21:838–40.

1967

General theory of X-ray diffraction in real crystals. *Phys. Rev. Let.* 18:195–96.

With B. T. Matthias, G. W. Webb, and J. J. Engelhardt. Melting-point anomalies. *Phys. Rev. Lett.* 18:781–84.

Theory of X-ray diffraction in crystals with stacking faults. *Acta Crystallogr.* 23:44–49.

A general theory of X-ray diffraction in crystals. *Acta Crystallogr.* 23:558–64.

1968

Experimental tests of the general formula for the integrated intensity of a real crystal. *Acta Crystallogr.* A24:212–16.

Extinction in a lithium fluoride sphere. *Acta Crystallogr.* A24:324.

Extinction and Borrmann effect in mosaic crystals. *Acta Crystallogr.* A24:421–24.

Extinction and Borrmann effect in a calcium fluoride sphere. *Acta Crystallogr.* A24:425–27.

With G. Arrhenius, E. Corenzwit, R. Fitzgerald, G. W. Hull, Jr., H. L. Luo, and B. T. Matthias. Superconductivity of Nb_3(Al, Ge) above 20.5°K. *Proc. Natl. Acad. Sci. USA* 61:621–28.

1969

Theoretical corrections for extinction. *Acta Crystallogr.* A25:102.

Intensities and structure factors. Concluding remarks. *Acta Crystallogr.* A25:276.

With Robert Benz. Crystal structure of the compounds U_2N_2X and $Th_2(N,O)_2X$ with X = P, S, As, and Se. *Acta Crystallogr.* B25:294–96.

1970

With F. H. Ellinger. Unit cell of the Zeta phase of the plutonium-zirconium and the plutonium-hafnium systems. Los Alamos Scientific Laboratory of the University of California, report no. LA-4367:1–4.

With Robert Benz. Crystal structure of the compounds U_2N_2X and Th_2N_2X with X = Sb, Te and Bi. *Acta Crystallogr.* B26:823–27.

With A. S. Cooper, E. Corenzwit, L. D. Longinotti, and B. T. Matthias. Superconductivity: The transition temperature peak below four electons per atom. *Proc. Natl. Acad. Sci. USA* 67:313–19.

With Robert Benz. Crystal structure of Th_2CrN_3, Th_2MnN_3, U_2CrN_3 and U_2MnN_3. *J. Nucl. Mater.* 37:109–13.

1971

Precise crystal structure of phenakite, Be_2SiO_4. (In Russian.) *Kristallogr.* 16:1161.

With A. L. Bowman, G. P. Arnold, and N. H. Krikorian. The crystal structure of U_2IrC_2. *Acta Crystallogr.* B27:1067.

1972

With A. C. Lawson. Low temperature lattice transformation of HfV_2. *Phys. Lett.* 38A:1.

With R. Benz and G. P. Arnold. ThCN crystal structure. *Acta Crystallogr.* B28:1724.

With D. C. Johnston. High temperature superconductivity in Li-Ti-O ternary system. *Mater. Res. Bull.* 8:777–84.

Metallic radii and electron configurations of the 5f-6d metals. *J. Inorg. Nucl. Chem.* 35:3487–97.

1974

With F. H. Ellinger. Structure of cerium metal at high pressure. *Phys. Rev. Lett.* 32:773–74.

1975

On californium metal. *J. Inorg. Nucl. Chem.* 37:1441–42.

1976

Bond lengths and bond strengths in compounds of the 5f elements. In *Proceedings of the Fifth International Transplutonium Element Symposium,* Baden-Baden, Germany, pp. 13–17.

1977

With F. H. Ellinger. The crystal structures of cerium metal at high pressure. *Acta Crystallogr.* A33:155–60.

With J. P. Charvillat. Lattice parameters of the ternary compounds Cm_2O_2Sb, Cm_2O_2Bi, Am_2O_2Bi and $Pu_2(O,N)_2Sb$. *Inorg. Nucl. Chem. Lett.* 13:161–63.

On the crystal structure of α'-cerium. *J. Appl. Phys.* 48:1391–94.

1978

Crystal structure of the α''-cerium phases. *Proc. Natl. Acad. Sci. USA* 75:1066–67.

Bond lengths in oxygen and halogen compounds of d and f elements. *J. Less Common Met.* 62:1–7.

1980

With R. Penneman. Application of bond length-strength analysis to 5f element fluorides. *J. Less Common Met.* 69:369–77.