Experiences in Crystallography – 1924 to Date

A. L. Patterson

It is an almost impossible task to try to analyse the reasons why one becomes involved in any field of scientific endeavour. One can only describe the circumstances, significant or otherwise, and the actions taken, logically or otherwise, under these circumstances. In 1924 when I wrote to W. H. Bragg asking him to accept me as a Research Worker in the Davy-Faraday Laboratory, I was writing an M.Sc. thesis at McGill on the production of hard X-rays by the β-rays from radium active deposit. I had had a very sound general education at Tonbridge School in England. This covered all the languages and literature and the chemistry required of a specialist in mathematics and physics at McGill, and gave me a good start on the mathematics and physics as well.

A number of circumstances influenced things at this time. I had taken a Second Class Honours B.Sc. degree in 1923 and only one who has attended a British or Colonial University can realize the depth of ignominy attached to such a thing. This disgrace was correctly ascribed by my professors to too many friends in Montreal and an addiction to skiing, bridge, and dancing and other related activities. Some quite fantastic suggestions were made as to how I might *live this down* but knowing myself these suggestions were a little difficult to take.

The second point was that the department was haunted by Rutherford who had left McGill in 1907. Our courses were interlarded with references to work 'done in this laboratory'. This was all very well but the stomach of an undergraduate is limited in its capacity in this direction. Present and future activity were much more important. But the fact was that all the faculty had worked either with Rutherford at McGill or at Manchester or at the Cavendish. I had met Rutherford when I spent the summer of 1922 in Cambridge counting α-particles for a Canadian friend and taking Thirkill's famous laboratory course in optics. I was much more impressed by E. R. in person than I was by

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him as a haunt but I remember being very vehement at the time that I was more interested in waves than I was in particles. Little did I know that they were to get together so soon. Apart from all this the undergraduate training in physics at McGill was quite good. Perhaps stronger in classical theoretical physics than in other fields. There were some excellent lecturers in the department and they emphasized very heavily the training of students as lecturers. Physics students were required to carry a heavy load in mathematics. This was excellent and I learned a great deal particularly in the British tradition, but a great deal of 20th century continental mathematics had not penetrated.

And again I had become very interested in my thesis problem. There were two things involved: what happened to the β-rays as they collided successively with the atoms of the solid, and what was the nature of the radiation they produced. Good questions! I figured out two things I might do about it. I might learn how to analyse the spectrum of the X-radiation. This was preferable to measuring the total radiation in the classical tradition of my thesis experiments. The other thing was to learn something of the theory both of the statistics of the β-rays going through the matter and of the details of their encounters with the matter.

When in the spring of 1924, McGill announced the endowment of two Moyse Travelling fellowships, one in literature and another in science it was with this background that I made my application. In my first draft, I listed four possibilities in order: 1) W. H. Bragg at the Royal Institution; 2) M. Siegbahn in Stockholm; 3) N. Bohr in Copenhagen; 4) E. Rutherford in Cambridge. The order of this listing aroused dire consternation. Rutherford simply could not be last, and also for some reason I was allowed only three choices. I stuck to my guns on the first and finally wrote Bragg, Rutherford, and Bohr. Since there was no air mail and the timing for the application was close there was no question of being accepted anywhere before making the application. I was a little surprised and very pleased to be awarded the fellowship as I had been told that the Second Class would weigh heavily against me. It seemed that a very good friend on the mathematics staff had fought hard for me. I decided to go to England where my parents were anyway, and wrote a two-page letter to Bragg telling him why I wanted to work in his laboratory and asking him if I could see him when I got to London. Because I was sailing within a week or so I asked Bragg to write me care of my ship on arrival in Southampton. His reply was prompter than I could have expected and his letter had met the ship on its preceding trip. When I went aboard in Montreal, I found the following letter in my cabin on the paper of the Davy-Faraday Laboratory:

3rd July 1924

Dear Mr. Patterson,

I will expect you in London between the 23rd and the 26th, when I shall be glad to see you.

Yours sincerely,
W. H. Bragg

Fortunately before I left McGill, Dr. Eve's secretary had showed me his letter from Bragg which I quote only from memory:

Dear Eve,

Shall be very glad to have Patterson.

Yours sincerely,
W. H. Bragg

This with my own letter gave me a very happy Atlantic crossing. I arrived in time to see Bragg on the 23rd. He told me that Jenkinson was making two new cameras, one of which was assigned to me. It would have a single circle goniometer head! He suggested that I work on the phenyl aliphatic acids as a tie in between his work on naphthalene and anthracene, and the work Muller and Shearer were doing on long chain compounds. He then told me to go take a good long holiday, possibly to read the two volumes of Tutton, and follow them by Hilton so that I would be ready to read the new paper by Astbury and Yardley on space-groups when I came back in September. I did read Tutton, but Hilton and the paper took the best part of the next year.

When I returned in September I set out to build my own X-ray equipment. The tube was a gas tube designed by Shearer and was built in the shop. The pumps were of local design and I was told that when I had made one in soft glass, I would be given some of a very expensive new glass called Pyrex to make the two that I needed. The pumps were made with metal water jackets closed by split corks at the ends. They always leaked and my camping experience suggested wet asbestos cord as a means of conducting the drips down the drain. The high voltage source was an induction coil. The core was a heavy cardboard tube packed with soft iron wire. The primary was cotton covered heavy copper wire wound on the outside of the cardboard tube and taped in place. This primary and core was mounted vertically in the center of a square base-board on which three or four secondary coils were mounted, separated by small ebonite insulators. Fortunately for us, the secondary coils could be bought by the dozen and we did not have to wind them ourselves. The interrupter for the DC current was also a local product. An aluminium hot-water bottle was half-filled with the electrolyte and a platinum wire shielded by a pyrex tube was inserted through a vented cork. When operating, these interrupters wailed like banshees so that at least ten or twelve of them were set out in rows on the lab roof. Occasionally one would develop an excess of gas and blow up. The screams of the interrupters could be heard on the arc of the lantern in the lecture room and often a worried Davy-Faradayite would hastily leave a Tuesday or Thursday afternoon lecture because he had heard his own well-recognized note stop abruptly. I was very proud of myself when I first had X-rays some time in November.

There was a great deal of activity at the Royal Institution at this time. Astbury and Yardley had written their space-group paper and all of us were learning to apply it to practical examples. Bernal's paper on rotating crystal methods was already in manuscript. Astbury applied these methods to the acetyl acetones and I to phenyl propionic acid. He and I spent hours in working through the details of the paper sometimes getting different answers from one another and from Bernal. Bragg and Gibbs worked out the structure of α and β quartz, Müller and Shearer were making some sense out of long-chain compounds. But most of us were determining space groups and obtaining very little structural information. It was only when a molecule had some symmetry that some definite information could be given. This was the case with the work of Knaggs and of George. We were all hoping for some 'clue' such as a change in one axis between two related compounds as had given Bragg an idea for anthracene, or the 'enhancement principle' which suggested interleaving of molecules. Many hours of work and perhaps sometimes more of discussion were spent in trying to devise ways of analysing organic structures. Of course many of these hours did pay off in the work of the Royal Institution in the early thirties, when Lonsdale solved hexa-methyl- and hexachloro-benzene and Robertson, who arrived after I had left, got durene, naphthalene and anthracene, and then developed the heavy atom and isomorphous replacement methods. In addition to the people I have mentioned already, others of the very close group were Burgers, Jackson, Mathieu, Orelkin, Plummer and Ponte. In addition we all had wonderful chemical backing from Saville, Smith and Lawrence.

A very great impetus to our discussions was added with the appearance of Duane's paper in 1925 on the crystal as a three-dimensional Fourier series, reviving W. H. Bragg's suggestion from 1915. None of us seems to have known of the latter, and although George Shearer was doing beautiful work in using one-dimensional Fourier series in explaining intensity distributions in long-chain paraffins, acids, and ketones, no one had the notion of using two- or three-dimensional series.

It had been my plan to return to McGill after two years in Bragg's laboratory to complete the work for Ph.D. and I had applied for a Canadian National Research Council Fellowship to support me at McGill. In the meantime the Pulp and Paper Industry Research Laboratory at McGill had become interested in the work of Herzog and Jancke on the X-ray diffraction from cellulose, and suggested that I go to Dahlem to join the group headed by Hermann Mark which had been built up in Herzog's Institute to study such problems. Mark's group at the Kaiser-Wilhelm-Institut für Faserstoffchemie involved Bredig, Ehrenberg, Gottfried, Herrlinger, Kratky, Naray-Szabo, von Susich, Weissenberg, with Szilard and Kallmann in sort of orbital attachment. Another person in Herzog's Institute who was a great inspiration to me was Gerda Laski who headed an infra-red group. Down the street was Haber's Institut für Physikalische Chemie where I made many personal friends as I did in Eitel's Institut für Silikatforschung, next door. There I gave my first seminar in 'German' as I spoke it. The subject was W. L. Bragg's paper 'The Structure of Certain Silicates.' I can only quote Herzog's comment after my talk: 'Vorlesen können Sie vielleicht, aber Deutsch leider nicht.'

Of course the real excitement of each week was the Physical Colloquium with von Laue as chairman and Einstein, Planck, and Nernst sitting in the front row. Others that were around were Bothe, Hahn, Meitner, Pringsheim, Wigner and so many others that one cannot begin to list them all. Schrödinger had not yet come to Berlin, but his influence and Heisenberg's dominated the scene. The word was Quantenmechanik. But although I was excited about all this new physics I was still trying to understand von Laue's paper on the diffraction of X-rays by small particles. This had been suggested to me by Herzog and Mark as my project with the hope that I could make an accurate determination of the 'particle size' of cellulose. I was fascinated by the notion which von Laue introduced of the space surrounding a reciprocal lattice point and I could not see how to do anything about it except by the methods of approximation which he had used. Under the influence of my quantum mechanical friends I bought a copy of Courant-Hilbert and discovered that something called the Fourier Transform existed and at the same time learnt what a potent mathematical entity an orthogonal set of functions really was. These two concepts had not been developed at all in the courses I had taken. Each new set of functions had appeared independently of the others and the fact that they all had common properties was not mentioned. I had been taught about a Fourier Integral Theorem but always in its double integral form. The fact that it could be split into two, one the spectrum of the other was something very startling and illuminating. In a few weeks after reading the first few chapters of Courant-Hilbert I wrote a brief note for the *Zs. f. Physik* which was the basis for my later work on particle shape functions. After reading von Laue's paper I became very interested in trying to extend the theory. The work that I did made me very pessimistic as to the meaning of a particle size determination. As a result I did not have the courage to publish any interpretation of the experimental work which I had done on line-broadening in Dahlem. A theoretical paper did, however, come out of it all.

I never really got to know von Laue until Easter time in 1927. The copy of *Nature* which I received on Good Friday contained Davisson and Germer's paper on the diffraction of electrons by nickel. I rushed around to tell all my friends about it but found that everyone was away for the holiday. The only thing I could do was to read and re-read the paper, check all the calculations and do some of my own including an incorrect interpretation of why the diffraction angle did not check with the lattice spacing. I interpreted this a as change in lattice spacing near the surface of the crystal whereas Bethe correctly explained it a few weeks later in terms of a refractive index effect. When von Laue suspended the normal programme of the next colloquium and reported the paper, I disagreed with some of the points in his presentation and said as much. Most of the Professors in Germany valued their dignity very highly and to have disagreed with them in public would have been suicidal. Von Laue was not this way at all and said that I apparently knew more about the paper than he and asked me to review it. This I did, so scared that the first line I drew on the blackboard came out dotted. After the session, von Laue invited me to come out to his home a day or so later, when we had about three hours of discussion, first about electron diffraction, and then about the work on particle size and on the Fourier interpretation of the reciprocal lattice which I was trying to do in Berlin. Thereafter he was very friendly to me and I was able to see him frequently during the rest of my stay in Berlin. Discussions with him often began with a vaguely formulated problem and ended with something clear cut, even though a solution was perhaps not obvious.

At the same time as I arrived in Germany I had acquired my first and only McGill Ph.D. candidate. Thomas N. White, Jr. had taken his undergraduate degree in 1926 and it had been arranged for him to work with me on X-ray diffraction for M.Sc. and possibly Ph.D. When my plans were changed and my return to Montreal was delayed from 1926 to 1927 it was decided that he would set up some equipment and start X-ray work on his own. This he did to his great credit and obtained his M.Sc. in 1927 just before I returned from Germany to McGill.

During our collaboration at McGill, White and I became interested in the cyclohexane hexols, a remarkable group of compounds which occurs throughout nature. About seven or eight isomers are known and one and only one of these isomers or a related methoxy compound occurs in a given species. Closely related species may have different isomers, and widely separated species the same isomers. It was perhaps the background reading for these studies which made me so sure that X-ray diffraction had an important future in the support of biochemistry. However, all we could do then was to determine a lot of space groups.

In the spring of 1929 I visited G. H. Cameron at DuPont in Wilmington and on my way back stopped off in New York to see Ralph Wyckoff at the Rockefeller Institute. As a result of this trip two things happened. I started collaboration with Harvey Cameron which resulted in the monograph we published together in 1937 and in two theoretical papers which I wrote in 1939. Second, I was asked to join the staff of the Rockefeller Institute and Tom White was asked to come with me to hold a National Research Council post-doctoral fellowship at the Institute. While at the Rockefeller, White and I continued the work on the cyclohexane compounds and looked at a number of other substances.

But my obsession with the notion that something was to be learned about structural analysis from Fourier theory continued. Early in 1930, I looked through the tables of contents of all the mathematical journals in the New York Public Library. Whenever the title was promising I looked through the paper and whenever the paper was promising I read it carefully. A lot of the papers which I tried to read I didn't really understand and so missed the point of many papers (e.g., Toeplitz) which were later to be of importance for the 'phase problem'. I did pick up a number of ideas in the process and wrote two papers for *Zs. für Krist.* largely concerned with location of maxima of Fourier series and also on the 'enhancement principle'.

In the fall of 1931, I accepted a job at the Johnson Foundation for Medical Physics in Philadelphia where I hoped to do something with X-ray diffraction on biological materials. I did take some powder pictures of horse hemoglobin using a camera cooled by Prestone circulating between it and a barrel filled with ice and salt. Most of the work of the Foundation was concerned with nerve physiology and I was very interested in Bronk's work in this field and that of Hartline on the eye of limulus. I had a good time collaborating with Ray Zirkle in some experiments on the effect of X-rays on the growth of fern spores, but my other excursions into biology were not too happy and I decided in 1933 after two years in Philadelphia that somehow I had to get back into crystallography and Fourier series. The appearance of the early Fourier papers from the Royal Institution and West's beautiful paper on KH_2PO_4 only served to whet my appetite.

Fortunately I had saved some money during the years in New York and Philadelphia and thought I had enough to keep me for one year on my own. I didn't really understand about depressions and did not contemplate three years out of a job.

In this 'year' I hoped to do some structure work and to take another shot at the Fourier series. I knew that Bert Warren had an active and running X-ray lab at M.I.T. and that Norbert Wiener probably knew as much about Fourier integrals as anyone in the world. So I asked Bert to take me on as guest. No pay but no fees for use of the lab. The Physics Department of M.I.T. was in a big burst of activity under K. T. Compton as President of the Institute and John C. Slater as head of the Department. I cannot go into any detail about the rest of the Department but in Warren's group were Gingrich, Hultgren, Serduke, and G. G. Harvey. There were many bright seniors doing undergraduate theses and several very good Ph.D. candidates. When I was first there the main interest of the lab was tending toward liquids. I started again on particle-size work because of a seminar I had to prepare and had many opportunities to talk with Wiener. This latter was a laborious process but a very intriguing one. There was then and is now no subject which can be brought up on which Wiener does not have something interesting to say. And with him the subject is always changing. I estimate I got in about one question on Fourier theory per two or three hours conversation, but the answers were usually pay dirt.

I very soon learnt from him the fact that I had to work with the Faltung, but it took me more than a year to catch on to what it was all about. I spent most of my time looking at Faltungen of step-functions such as those which had been used by Shearer in the study of the aliphatic acids and ketones. And this was largely because I could make the Faltungen of step-functions by simple geometry and did not need to compute the series by the tiresome methods then available.

The understanding of the Faltung came, of course, from the work on liquids and their radial distributions. Warren with Gingrich and others had perfected the techniques used by Debye and Menke in the study of the X-ray scattering from liquids. These were of course based on the original suggestions of Zernike and Prins. Warren and Gingrich had already had the idea that these methods, applied to powders, would give the radial distribution in a crystal. While trying to learn about their work I noticed that the mathematical form of the theory given by Debye and Menke would be identical with that of the Faltung if the integrations over random orientation were left out and the randomness of choice of origin was left in. What was immediately apparent was that the crystal contained atoms and that the Faltung of a set of atoms was very special in that it would consist of a set of atom-like peaks whose centers were specified by the distances between the atoms in the crystal. It was fortunate that this was clear from the beginning and it was in this form that the interpretation of $|F|^2$-series was proposed. It is unfortunate that the notion arose later that the maxima of the Faltung were determined by the distances between the maxima of the Fourier series. This is clearly untrue in general and was never suggested by me. However, to go back to the story, all this happened on a Tuesday, and Friday was the deadline date for the Washington spring meeting (1934) of the American Physical Society. An abstract had to be prepared in a hurry to go in with that of Warren on the radial distribution in carbon black and that of Gingrich and Warren on the radial distribution in powders which was basic to my work. The only $|F(h)|^2$-series which I was able to compute in the month between the deadline and the meeting was the (hk0) of KH_2PO_4 and a one dimensional series for a simple layer structure. All three papers were very well received and had very full discussion with A. H. Compton in the chair and W. L. Bragg in the audience to ask the right questions.

Soon after the Washington meeting W. L. Bragg visited M.I.T. and told us about the copper sulfate structure of Lipson and Beevers and that they had a method for computing series by using 'strips'. We soon received the data for copper sulfate from them and computed the F^2-series and the sharpened F^2-series from their data. We had also computed the F^2-series of hexachlorobenzene from K. Lonsdale's paper.

All these series were computed by a method suggested to me by George Kimball which involved multiplication of every term in the series by its appropriate sine or cosine. We recognized the tremendous repetition involved. Since we had not yet heard the details of Beevers and Lipson's strips the method later developed in more detail by Tunell and myself was set up.

In retrospect it is a source of satisfaction to me that I did so much work on the F^2-series method before publishing the second paper. This made it possible for me to draw attention to many of the difficulties which were likely to and which did arise. I must say that I was very annoyed at myself for missing the beautiful extension of the method made by Harker. I guess that I really could not get out of the plane.

While I was looking for jobs during my second and third years at M.I.T., I was offered fellowships in physics at two quite well known universities on the condition that I change my field of research. It was therefore very gratifying in 1936 that I was offered an assistant professorship in physics at Bryn Mawr College, with the express purpose of developing X-ray analysis in parallel with the wider interests of Walter Michels in the solid state.

Bryn Mawr College throughout its history has been remarkable for the fact that with the small faculty usually associated with a liberal arts undergraduate college it runs a full program of graduate work for the master's and doctor's degrees. It is a most stimulating place with the tradition that the faculty run the academic end of the college without interference from deans or presidents. However, it is difficult for the faculty members (particularly in science) to get any research done themselves. Their only way of keeping research going is through the work of the graduate students. Of these there are not many, but they are of high quality and the X-ray work was kept going largely through the collaboration of two very good Ph.D. candidates and a number of M.A.'s.

The kind of work which a faculty member could do himself was typified by two particle size papers, started at M.I.T. but finished at Bryn Mawr. The work on homometric structures was ideal, since I was able to have the benefit of wonderful collaboration from mathematical colleagues. I also did some work on generalized transforms, and finally Walter Michels and I ended my stay at Bryn Mawr in the full glory of a book on elementary physics.

When in 1949 I was given the opportunity by Dr. Stanley Reimann to start an X-ray diffraction group at the Institute for Cancer Research, I realized that the urge to apply these methods to biological problems could now be satisfied. At McGill, when the interest first developed, the prospect of X-ray analysis, even for a crystal of a very small organic molecule, ended with a space-group determination. Now it is possible to say to a biochemist that almost any molecule containing a score or two of atoms that can be prepared in crystalline form can be analysed in a finite amount of time. And now too an attack on the largest molecules is succeeding with the development of special techniques.

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