My Time with X-rays and Crystals

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A great German poet once said, 'Where kings are building, carters find work.' Most of history is written about kings, and that is as it should be, but the work of carters has its own history too, and that too is important. For great discoverers would achieve little unless followed by enterprising settlers. I shall recollect here a brief phase of the early colonisation of the immense domain of knowledge on which Max von Laue first set foot 50 years ago, followed by W. H. and W. L. Bragg, whose discoveries first revealed its major treasures.

The example of great scientists is the light which guides all workers in science, but we must guard against being blinded by it. There has been too much talk about the flash of discovery and this has tended to obscure the fact that discoveries, however great, can only give effect to some intrinsic potentiality of the intellectual situation in which scientists find themselves. It is easier to see this for the kind of work that I have done than it is for major discoveries, and this may justify my telling this story.

At the time when I started this work, a chain of research institutes existed in Germany supported by the Kaiser Wilhelm Gesellschaft. One of these was the newly founded Institute of Fibre Chemistry in Berlin-Dahlem, to which I was appointed in the autumn of 1920. My first contacts revealed the peculiar character of the company I had joined. Following German custom, I called on the Directors of the other institutes in Dahlem, and first of all on the great Fritz Haber, Director of the Institute of Physical Chemistry. Haber, who had seen my speculative papers on reaction kinetics, referred to them with a stern admonishment: 'Reaction velocity' said he 'is a world problem. You should cook a piece of meat.' He meant that first of all, I should prove my capacity as a craftsman; the rest would follow. Rather different was the impression conveyed by Carl Neuberg, Director of the Institute of Biochemistry. 'Don't stay here, dear Colleague,' he

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said. 'Accept the first offer of a Chair at a university. If you don't make a discovery for a couple of years here, you are just an old ass; at a university you have always your academic laurels to rest upon.'

Discovery requires in fact something beyond craftsmanship, namely the gift of recognizing a problem that is ripe for solution by your own powers, large enough to engage your powers to the full, and worth the expenditure of this effort. Haber had admonished me, because he thought I had taken on a problem that was not yet ripe, and in any case too large for me. Perhaps he was right; but at any rate, my new job with the Institute of Fibre Chemistry led me in a different direction. On my arrival at the institute its Director, Reginald Oliver Herzog—whom I remember warmly for his kindness and wide intellectual perceptiveness—immediately gave me what Haber said I needed, a piece of meat to cook. Following his discovery of the crystalline nature of cellulose (paralleled by Scherrer) Herzog—aided by his assistant Jancke—had just found that a bundle of ramie fibres irradiated by an X-ray beam at right angles produced a diffraction pattern composed of sets of four equivalent dots symmetrical to two mirror-planes, one passing through the primary beam and the axe of the fibres, and the other normal to the former. There was excitement about this diagram and I was asked to solve the mystery. In the next few days I made my first acquaintance with the theory of X-ray diffraction, of which, owing to wars and revolutions and my exclusive interest in thermodynamics and kinetics, I had heard little before. However, I soon returned the result that the four-point diagram was caused by a group of parallel crystals arranged at random around one axis, and this interpretation was included in a joint publication with the work of Herzog and Jancke. So I had cooked a piece of meat—and this transformed my position. Herzog, with kindly enthusiasm, showered me with every facility for experimental work, most precious of which were funds for employing assistants and financing research students. In this I was incredibly lucky. I was joined by Herrmann Mark, Erich Schmid, Karl Weissenberg, all three from Vienna, by Erwin von Gomperz and some others; the place was soon humming. It was the time of runaway inflation and poor Herzog found it difficult to pay all these people. Protest meetings were held, resolutions passed, Weissenberg in the lead; the Institute earned the name of an 'Assistenten-Republik'. We had a glorious time.

I shall now try to give an account from memory—without access even to my own papers—of the way some of my further contributions were born in those days. I found that all the dots forming the fibre diagram lay on a series of hyperbolae, each hyperbola comprising dots reflected by planes having identical indices with respect to the crystal axis parallel to the fibre. I established the formula determining the series of these hyperbolae, as a function of the identity period parallel to the axis of the fibre. Thus equipped, I evaluated the elementary cell of cellulose and drew the conclusion that the structure of cellulose was either one straight giant molecule composed of a single file of linked hexoses, or else an aggregate of hexobiose-anhydrids; both structures were compatible with the symmetry and size of the elementary cell—but unfortunately I lacked the chemical sense for eliminating the second alternative.

This foolishness had also an amusing consequence. When I first stated my conclusions in the Colloquium presided over by Haber, there was a storm of protest from all sides. The assertion that the elementary cell of cellulose contained only four hexoses appeared scandalous, the more so, since I said that it was compatible both with an infinitely large molecular weight or an absurdly small one. I was gleefully witnessing the chemists at cross-purposes with a conceptual reform when I should have been better occupied in definitely establishing the chain structure as the only one compatible with the known chemical and physical properties of cellulose. I failed to see the importance of the problem.

A failure of the same kind was my treatment of sero-fibroin. Herzog had discovered its fibre diagram and handed it to me for evaluation. I determined its elementary cell and observed that there was only room for glycine and alanine in it. But I could not make up my mind what to think of the other observed decomposition products. I did not recognize the immense importance of the question, and passed it on half-baked to Brill, for the doctoral thesis he was doing under my supervision.

Such failures are worth recording, in order to correct the current theories of the scientific method, based altogether on success stories. It is interesting to recall in this connection, that the weakness of my initiative was due in part to the fact that my confidence in this line of inference was impaired by a slight deviation of the position of two dots on the equator of the cellulose diagram from the theoretical values predicted by my analysis. These loose ends were debilitating; but I am glad that at least I did not obey the current Sunday school precepts of the scientific method, which would command you to reject a theory if a single piece of evidence contradicts it. (I still don't know what caused the discrepancy in those dots.)

Following my discovery of the hyperbolae in the fibre diagram of cellulose and my evaluation of its elementary cell on this basis, the principles thus established were transposed into the rotating crystal method in collaboration with Weissenberg and Mark. The former far surpassed me in mathematics, while the latter lent me his manipulative skill bordering on genius. To the best of my recollection, the project, including the suggestion of using an elongated Debye-camera for the purpose of including the higher layer lines, came from me. Weissenberg generalized the layer line relationship, which I had only established for the directions of the crystal axes and perpendicular incidence of the beam, to include identity periods in any direction and all angles of incidence; Mark carried out the first experiments with the new method. The first use of the rotating crystal apparatus for the determination of an unknown crystal structure was made, I think, by Mark and myself in 1923 on white tin. It was used in the first place by Mark, Schmid and myself for elucidating the plastic flow of zinc crystals.

The strength of solids had now become my principal interest. The technological purpose of the Institute had thrown this great problem into my lap. I saw that the characteristic feature of the solid state, namely its solidity, was yet unexplained, and indeed, hardly explored by physicists. I found that in the light of the recently discovered structure of rocksalt, such a crystal should be thousands of times stronger in resisting rigid rupture or plastic deformation, than it actually was. In facing this paradox I appealed to two features of modern physics (1) that a crystal of rock salt was one giant molecule and (2) that inside a molecule energy could be transferred by quantum jumps not controlled by the laws of classical mechanics. From a calculation based on the actual strength of rocksalt I concluded that the energy required for producing the new surface formed by breaking the crystal would have to be supplied from the stress stored up on either side of the future break, in an area extending two or three millimeters in both directions of it. So I set out to show experimentally that crystals shorter than a few millimeters were stronger than those of greater length. The result was inconclusive and the whole idea may have been wrong, but in pursuing it I stumbled on an important aspect of the strength of materials which seemed to reflect my original paradox and to encourage the way I was trying to solve it; I came to know about the hardening of materials by cold working.

I was deeply struck by the fact that every process that destroyed the ideal structure of crystals (and thus reduced the areas which could be regarded as single molecules) increased the strength of crystalline materials. This seemed to confirm the principle by which I explained the low resistance of crystals to stress and to refute the rival theory—inspired by the work of Griffith on quartz threads—that the weakness of crystals was due to cracks or other imperfections of structure. The cold working of rocksalt crystals by vigorously filing their sides and of tungsten crystals (obtained from filaments for incandescent lamps) by drawing them through a die, confirmed this. The results, presented in September 1921 under the title 'The Hardening of Crystals by Cold Working' to the meeting of the Bunsen Gesellschaft, were received with uneasy surprise. Gustav Tammann, speaking as an elder statesman, expressed this in the discussion. Yet, as later work was to show, my observations were fundamentally sound.

Meanwhile, I took up antecedent questions. Some metallurgists, interested in my work on the hardening of single crystals, told me of a method invented by Czochralski for producing metal crystals in the form of wires. It consisted in pulling out a thread from a pool of molten metal, so that the thread continued to solidify at the rate at which you were pulling it out. Erwin von Gomperz, who was doing his thesis with me, was put to growing single crystals of tin and zinc in this way. Unfortunately, the metal tended to come out in lumps, and the project was saved only by the intervention of Hermann Mark who covered the liquid metal by a sheet of mica with a hole in the middle, through which the thread came out as a smooth cylindrical wire. But for this ingenious intervention, our subsequent investigations of the plastic flow of metals might not have come about.

The next stage of our work elucidated the crystallographic laws of plastic flow in zinc and tin. These are now well known. Of this work I should like to say only that it is a rare instance of something supposed to be a common occurence, namely of the participation of as many as three scientists as equals in a fairly important piece of work. We were lucky in hitting on a problem ripe for solution, big enough to engage our combined faculties, and the solution of which was worth this effort. The ripeness of the problem was confirmed, when a few months after our paper on zinc had came out, a similar investigation was published by G. I. Taylor and Miss Elam in England, that solved the same problem for an entirely different system, namely aluminium. Though these two parallel papers applied very different methods, they both evaluated identical possibilities concealed in a common intellectual situation.*

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* The wooden model of slip in crystals which I often see reproduced in current litterature,

The following episode might illustrate this principle on a smaller scale and show incidentally also, how crude was the knowledge of crystal structures on which we were relying at the time. Shortly after Mark and I had published the structure of white tin, we received the visit of the Dutch scientist Van Arkel, who told us that our result was wrong, for he had established an entirely different structure of the metal. Only after hours of discussion did it become apparent that his structure was actually the same as ours, but looked different because he represented it with axes turned by 45 degrees relative to ours.

Most of this work was completed in a little over two years from the day when I first 'cooked a piece of meat' in Dahlem—all of it being supported as a rather odd kind of fibre chemistry by our noble-hearted director, R. O. Herzog. Having established the geometrical mechanism of deformation in crystalline solids, we could now take up effectively the physical problems of deformation that had first drawn me into this field. But Mark had become engrossed in structure analysis and Weissenberg had also taken up problems of his own; it fell, therefore, mainly to Schmid and myself to embark on the physics of solid strength, equipped with the crystallographic results to which our whole group had contributed before. Schmid established his law of shearing stress at the yield point. Together we observed that minimal deformation of crystals can lead to noticeable hardening and discovered the fact of 'recovery' which cancelled hardening without recrystallization. Schmid established hardening for greater deformations, by applying his law of shearing stress. Jointly we proved by strains applied under hydrostatic pressure, that stress vertical to a slip plane leaves its resistance to shearing unaffected. Experiments with W. Meissner and E. Schmid at the temperature of 1°K demonstrated the athermal quality of slip in crystals, and this brought out the fundamental contrast of crystal plasticity to the deformation of amorphous solids, which become perfectly rigid at absolute zero. Observations with G. Masing on fine grained zinc ruptured at liquid air temperature, confirmed the fact that the internal fragmentation of a crystalline material increased its resistance to brittle rupture. In every instance so far the strength of crystals was found rising from its paradoxically low value, towards the much higher theoretical strength which Griffith has actually observed in amorphous quartz threads—to

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was made on my instructions in the small workshop of a joiner in Dahlem. At one time the mathematician R. v. Mises remarked acidly on its widespread use by members of our group: 'All the problems of plasticity are apparently to be solved by pushing this model to and fro.'

the extent that the disturbance of the crystalline order shifted the condition of the crystal towards the amorphous state. This was also the explanation I found for the curious Joffé-effect: I showed by experiments with W. Ewald, that the water dissolving the surface of a rocksalt prism reduces its resistance against plastic flow; and that it is the onset of this flow, acting as cold working, that increases the crystal's resistance to brittle rupture, as observed by Joffé.*

I shall pass over our enquiries starting from the discovery of fibre structure in cold worked polycrystalline metals, by merely mentioning that it was nice to be able to account for this phenomenon by the crystallography of plastic deformation as observed in single crystals. More interesting perhaps was the fact (found with P. Beck) that while the annealing of a bent aluminium crystal caused it to recrystallize, no recrystallization took place if it was straightened out before annealing. These were sidelines, for they threw no light on the nature of solid strength, which remained shrouded in the paradox that all effects which would tend to restore the ideal crystal structure appeared to weaken the material far below its ideal strength, whereas every disturbance of this structure tended to raise its strength towards its ideal value.

My fascination with this fact had borne fruit—but it had proved excessive. From a paper written by Erich Schmid on the occasion of my 70th birthday, I gather that the picture I had formed of the hardening and weakening of crystals made me overlook an important clue for modifying it. My experiments with W. Ewald (1924) show that bending a rocksalt crystal in one direction hardens it only for further bending in the same direction and actually weakens it for bending it back. Schmid says that such mechanical recovery has subsequently been effected in various crystals, including those of metals. Had I noticed the fact that deformation may actually weaken a crystal, my mind would have been more receptive to the idea that the extremely low resistance of crystals against plastic deformation might be due to the kind of irregularity in the structure of the crystals that is now known as *dislocation*. However the idea of dislocations causing a high degree of plasticity did gradually take shape in my thoughts. I gave a full account of this theory in April 1932 in a lecture addressed to the members of Joffé's institute in Leningrad, who received it well. On returning to Berlin I talked about my theory to Orowan who told me

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* Joffé himself contested my explanation, especially since the lowering of the yield-point of rocksalt by dissolving its surface with water had not been observed before. But I gather that this effect is now well established.

that he had developed a similar idea in his thesis about to be submitted for a degree. He urged me to publish my paper without considering his rival claim, but I preferred to delay this until he too was free to publish. (This explains why my communication appeared in print in Germany and in German a year after I had left the country and had already published many papers in England.)

Meanwhile the principle that scientists only reveal hidden knowledge which has become accessible by the intellectual situation of the moment, reasserted itself. Once more the same intimations had matured to the same solutions in another, totally different, mind, that of G. I. Taylor in England. What I had published as 'Versetzung' in German, he published simultaneously as 'dislocation' in English.

By the autumn of 1923 I left the Institute for Fibre Chemistry through promotion to independent Membership in the Institute of Physical Chemistry. Haber received me now with full confidence in my ability to work as a scientist and I immediately plunged back into reaction kinetics. My time with X-rays and crystals had lasted three years.

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