Scattering Phenomena in Electron Diffraction

The Origin of the Dynamical Theory of X-Ray Diffraction

(SPECIAL LECTURE)

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Ladies and Gentlemen,

Let me go back to the spring of 1913, a year after Laue's discovery. By then, Laue had become Professor of Theoretical Physics at the University in Zürich; Friedrich went to the University Clinic in Freiburg as a co-worker to Professor Krönig for developing the methods of dosimetry in X-ray therapy; Knipping had accepted a job with the Siemens Laboratories in Berlin; and I was Sommerfeld's experimental assistant in Munich. As such, I continued X-ray diffraction work with Friedrich's instrumentation which is now in the Deutsche Museum in Munich.

My activity soon came to an end by the outbreak of war. I immediately went to the hospital in Munich in order to learn some of the medical applications of X-rays, and soon after followed our equipment when it was transferred to a military emergency hospital hastily established in one of the city's primary schools. After a year or so of applying X-rays to war casualties rather than to crystals, I grew nervous lest the glorious war might come to an end without my having seen the front. So, in early 1915 I applied for a position as 'Field X-ray Mechanic of the Army' and soon became an army employee (looked down upon by all true soldiers) in a gorgeous uniform which was often mistaken as that of an officer of the General Staff and treated with much respect.

The unit I helped to assemble in the Siemens works in Berlin consisted of a wagon housing a gasoline motor-dynamo, transformer, four X-ray tubes, dark room equipment, a table for the patients, etc., all very neatly and firmly packed for transportation over the roughest roads. The manual also provided for four horses to draw the wagon, one saddle horse for myself, and two men to help me —but these items were on paper only and I was told to organize any help I needed on the spot. I was ordered to report with my equipment at the headquarters of the fifth army (Hindenburg's), where my wagon would be sent by rail, but nobody in Berlin was willing to divulge the location of the headquarters. Finally I was given a voucher for a ticket to Königsberg in East Prussia.

When I arrived there, I found Königsberg a very attractive town. I installed myself comfortably in the best hotel, in an entirely unmilitary fashion, searched the railway freight yard every day for my wagon, made friends with the physicist, Prof. Kauffmann (who had been the first to show experimentally, in 1901, the dependence of the mass of the electron on its velocity), and settled down to writing a paper on optical refraction which I was keen to get published since it was an essential sequence to my thesis.

After about six weeks I ran out of money in this hotel and wrote a letter in as good a military style as I could muster to the Medical Department of the Fifth Army suggesting they send me the two months salary which were due. The repercussion which this letter had was immediate. I got told down over the phone in a way I never before, or after, experienced: how dare I not report, where had I been loitering all the time, was I sick, and woe to me if I didn't report next morning at seven at the depot in Tilsit—I would just be in time for the night train if I hurried.

This ended my experience in Königsberg. Needless to say I found the barking dogs in Tilsit quite friendly after a while and had three more weeks there before my wagon arrived. It was then hitched on to a hospital train and I had five comfortable days travelling slowly past the snow-flurried wide stretches of Russia to a small Lithuanian hamlet, Abeli. The forward section of an army hospital was established in the manor house of an estate and I soon set up my outfit in an asbestos hut under the high trees leading up to the house. Not long after, I made my one and only prisoner of war, by obtaining the release of a young Russian smith from the nearby barbed wire camp. We grew great friends and he soon learned to develop the X-ray plates, which gave me time to think over the problems of the dynamical theory of X-ray diffraction which gradually revealed themselves to me.

I saw clearly that the simple Laue theory, which we now call the kinematical theory, was in conflict with the energy principle. The original starting point for the dynamical theory was curiously enough the fact that neither Laue nor I were able to see the reason for Friedel's Law concerning the symmetry of the diffraction diagrams; I hoped to obtain the answer from a more elaborate theory. But I must confess that in the excitement of carrying through my theory, I forgot all about Friedel's Law. Later discussion dealt with absorbing crystals, and it was only much later that Professors Miyake, Uyeda and Kohra discovered that if there is more than one diffracted ray, Friedel's Law may be invalidated even in a non-absorbing crystal.

In this little hut in the farm yard I spent my first Russian winter, which was severe and long-lasting. When at last spring came, it was just wonderful. Within a few days the snow melted, the first buds appeared on the trees, the birds arrived and began nesting, and soon you found the plover's eggs hidden in the high tassels of grass in the marshes. It was a pleasure to explore the countryside, and as fighting had practically come to an end on this part of the front, I often indulged in taking long walks, observing and thinking about the dynamical theory. Without the uninterrupted solitude of these months I might never have found this initially rather formidable theory. I was later transferred to another small township, Novo-Alexandrovsk, also near Dvinsk (which lay on the Russian side of the river Dvina). Here my X-ray station was in the cellar of the court house turned into a hospital. It was a cosy place because half the room consisted of a big arched baking oven which was heated up with logs until the stones were hot; then the vent was closed and the hot air went to the room. I was aware of the danger of having it warm and managed several times to get away from my evening calculations just before the carbon monoxyde had fully put me to sleep.

It was not quite simple to see clearly the idea of the dynamical theory of X-ray diffraction. It was novel in many respects, but my previous work was a good preparation. This consisted of my thesis on the theory of dispersion in an anisotropic medium, and the paper on refraction which I finished writing in Königsberg. The thesis subject was given to me by my great teacher, Sommerfeld. I went to him in 1910 after I had heard his courses for two years and asked him whether he would accept me as a doctorand. Sommerfeld pulled out of the drawer of his desk a sheet of paper with a list of ten or fifteen topics for theses, which ranged from the propagation of radio waves, the incipience of turbulence, the self-induction of coils of various cross sections to many other problems requiring the solution of partial differential equations with boundary conditions. The last topic on the list was: to investigate whether an anisotropic arrangement of ordinary isotropic resonators would be sufficient to produce double refraction of a medium. When I saw this last topic, I knew at once that it was this one I wanted, even though Sommerfeld warned me that he would not be able to give me much help, such as he could give in the case of the more familiar boundary problems.—Only many years later a situation came back to my mind how, when I was fifteen or sixteen and still at school, I explained to my mother that I was convinced it should be possible to explore the structure of matter with light, extending the work of Helmholtz whose biography I had been reading. So, unknowingly to me, I picked the subject of Sommerfeld's list which was preformed in my mind—and I have never regretted it.

In the theory of dispersion one assumes that the optical properties of a body are caused by 'resonators' or 'dipoles' which form part of its molecules. Under the influence of the incident ray, each dipole oscillates and sends out spherical wavelets which are superimposed on the incident ray, so that together they form the refracted ray. There is, furthermore, a condition to be fulfilled by the optical field in the interior of the body, namely that it travel with a certain velocity q which is, in the case of light, in general smaller than the velocity of light in free space, c, and in the case of X-rays greater.

In the previous theories of dispersion of Drude, Lorentz and Planck there had been some confusion about the role of the incident ray. It was agreed that since we are dealing with solutions of Maxwell's Equations, which are linear equations, all individual fields can be superimposed. My study was carried out on a more precise model of the solid than had been used before, namely an orthorhombic arrangement of resonators, as suggested by Sommerfeld. This allowed me to calculate the field with all precision and without having to use statistical considerations. It showed that I had no use whatsoever for a superimposed incident ray in the interior of the body. This would have a velocity c like in vacuum, and it simply does not fit into the scheme of propagation of waves in the interior of the crystal. I had therefore to leave this incident wave out in constructing a self-supporting system of waves, i.e. one in which there are waves created by the dipole oscillations and dipole oscillations created by the waves.

In a crystal infinitely extended in all directions there is actually no such thing as an incident wave. In this case you construct by the just mentioned condition of self-consistency a proper state of vibration of the system consisting of resonators plus optical field. Such a state is characterized by a definite velocity of phase propagation for any frequency, and, in the case of an anisotropic medium, for any direction and mode of polarization. —An incident ray appears only when there exists a surface, so that there is an interior and exterior region, that is in the simplest case, when the crystal fills only one half-space. Since the superposition of the incident ray in the interior of the crystal would destroy the dynamical balance of the self-consistent field calculated for the interior, I came to the conclusion that by cutting away half of the unbounded crystal the incident ray must be annulled within the entire volume occupied by the crystal.

This was a rather remarkable conclusion which none of the people who had great credit in the theory of dispersion had pronounced. It was in order to find out Laue's attitude to this idea that I went to consult him early in 1912. In the course of this conversation he formed the idea of letting X-rays pass through a crystal, and this became the starting point for his famous experiment.

The paper on optical refraction which I finished in Königsberg carried the idea of the annullment of the incident field at the boundary of the half-crystal a step further by obtaining from this condition the relation between the internal and the external optical fields, that is, Fresnel's formulae for the ratios of the reflected and refracted amplitudes to that of the incident wave.

It was fortunate that I had carried through the whole theory for light waves, for when it came to the theory for X-rays, the same procedure could be followed. I assumed that the optical field in this case consisted of n strong waves, selected on the indication of the kinematical theory. Since none of these can exist in the crystal without generating the others, this bundle of n waves forms the unit replacing the single wave in the case of light. First the condition of its self-supported propagation in the unbounded crystal has to be found. This results in a definite phase velocity for each of the waves, and a corresponding ratio of their amplitudes. This first part of the investigation is thus the establishment of the proper field modes; the result can be visualized by constructing the 'Surface of Dispersion' in reciprocal space. In the case of n strong component waves, this surface consists of $2n$ sheets, and any point on any sheet represents one of the possible modes.

The next step is to connect these interior modes to an incident field, namely a single plane wave falling on the surface of a half-crystal. It turns out that by the cutting away of the upper half of the crystal, the mere summation of all the wavelets coming from the lower half-space produces a field in the interior which consists not only of the n strong waves previously considered ("mesowaves"), but further n strong waves of only slightly different directions and of phase velocity c as in free space ("epiwaves"). Proper modes can now be picked out by choosing representative points on the $2n$ sheets in such a fashion that the epiwaves of all modes coincide in direction, though differing in amplitude. By superimposing such modes in appropriate strengths, all the epiwaves and the incident wave can be made to cancel throughout the interior of the crystal. The vibration in the crystal is then the superposition of dynamically possible proper modes with no alien fields destroying the self-consistency. The result is that the amplitudes of the n diffracted rays at any point x are fully determined in terms of the amplitude of the incident wave, in analogy to Fresnel's formulae.

The presence of the epiwaves follows automatically from the breaking-off of the summation of wavelets after cutting away the upper half-crystal. Starting from a field-point x in the interior of the crystal, the field at x is found in the unbounded crystal by summing all contributions coming from atomic planes parallel to the surface which lie above x, and the summation goes from the nearest plane 1 to infinity; in the bounded crystal there are only L atomic planes above x, and the summation runs from 1 to L. Since the contributions from successive atomic planes to the field at x have constant phase differences, φ, the series to be summed is essentially a geometric progression with quotient $p=\exp(i\varphi)$; the infinite sum is $(1-p)^{-1}$, the finite sum, however, is $(1-p)^{-1} - p^{L+1}(1-p)^{-1}$ of which the first term gives the mesowaves, the second the epiwaves.

It was only considerably later that I saw the full analogy of my procedure with that generally applied in problems of mechanics. If we have a complicated mechanical system, say a girder bridge on which the load is suddenly changed, what we do first is to calculate the proper vibrations of the system. Once we know the spectrum and the proper modes, then a very simple mathematical procedure yields the amplitudes which the various proper modes of vibration acquire if the system is released from rest at given displacements.—In the case of X-ray diffraction the wave incident on the surface of the half-crystal takes the place of the initial displacement of the mechanical problem.

When I wrote the dynamical theory as my Habilitationsschrift in 1917, I sent it to Sommerfeld. He remarked to my mother that it seemed a nice piece of speculation, though hard to follow, and very unlikely ever to be of practical importance. It seemed so, for some time. The first experimental justifications for the theory were the deviations from Bragg's Law found in 1919/20 by Stenström and Hjalmar; next came Ernst Wagner's observations of dark and bright lines crossing spectra (Aufhellungslinien, 1920). Then came the measurements by Bergen Davis and von Nardroff on the intensity of reflection from atomic planes which are not symmetrically orientated with respect to the crystal surface. All these results could be accounted for very nicely by the theory, but the important verifications came later, in the 1930's, from Allison and Parratt's and Coster and Prins' measurements of the reflection curves of nearly perfect crystals, and somewhat later by Renninger's similar measurements.

I was quite aware, from the beginning of my work, that absorption could be incorporated in the theory by assuming a complex value for the polarizability of the dipoles, or a frictional term in their equations of motion. But since absorption is a quantum effect, it seemed a dubious procedure to describe it in this way; besides, the theory was complicated enough even for the non-absorbing crystal.

Only at the very end of my work in Novo-Alexandrovsk, when discussing the reflection in the Bragg case, did I remember the paper by C.G. Darwin of 1913 on which I had once given a colloquium report while still in Munich. Physicists in Germany looked upon the action of a single atomic net plane as one of diffraction, producing not only the one reflected ray but the whole range of cross grating spectra. The latter were not mentioned in Darwin's paper. Besides, the Laue diagrams, usually taken with incidence along a symmetry element, led directly to the notion of the coexistence of a bundle of diffracted rays, whereas Darwin's treatment took only a single one of these into consideration. These differences in outlook prevented the ready acceptance of Darwin's paper in Germany, and I was quite astonished that my results, in the case of a single diffracted ray, checked with those of Darwin.

The first dynamical theory of X-ray diffraction considered only a crystal formed by a lattice of point-atoms. An important generalization was that to a crystal with a multi-atom basis. Here the surface of dispersion depends on the structure factor S_h, and I pointed out in 1925 that the integrated reflection is proportional to $|S|$, not to $|S|^2$ — a result that was borne out by the measurements on crystals of sufficient perfection.

The dynamical theory of X-ray diffraction in its original form was a product of the classical approach to the interaction between radiation and matter so brilliantly expounded by H.A. Lorentz in his ' Theory of Electrons.' M.v. Laue, in 1931, transformed the theory to a more descriptive model of the solid by assuming the existence of a periodic dielectric constant for which Maxwell's equations have to be solved representing the optical field. The justification of his assumption in terms of the wave-mechanical perturbation theory was given by M. Kohler in 1935. The changed form of the theory has been preferred by most authors, but it seems not to lead any farther in the discussion of experiments than what could also be obtained with the classical form.

Of much greater importance was the extension of the same ideas to the case of electron diffraction by Bethe (1928), and later to neutron diffraction. The cross section for the interaction between a low energy electron and an atom is far larger than that between an X-ray and an atom. The coupling between the plane waves constituting the elementary optical field of n waves is therefore much stronger, and effects barely observable with X-rays become dominant for electron waves. It is thus that the Japanese school of electron diffractionists have in recent years much advanced the discussion of the phenomena depending on such interaction, and the presence here of such a large number of Japanese physicists who are fully acquainted with the intricacies of the dynamical theory bears testimony to the fact that this theory is more than a nice speculation.