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Ewald Dynamical Diffraction Symposium

Over 250 scientists from four continents gathered at the University of Oklahoma on 23 March 1978 to honor Professor Paul P. Ewald in his 90th year with a special, day-long, Dynamical Diffraction Symposium. Recent developments in the applications of dynamical diffraction theory, created by Professor Ewald in 1917, were presented in a series of eight invited papers. It is noteworthy that his theory has only begun to have wide impact within the last twenty years, as experimental techniques and other theorists have caught up with it. The first paper was presented by Professor Ewald, who received standing ovations before he commenced and after he completed his talk.

It is now well known that Ewald's discussion of his doctoral thesis with Max von Laue led to ideas which played a significant part in the discovery of X-ray diffraction by von Laue, Friedrich and Knipping. Ewald then worked out much of the early theory of such diffraction, including the concept of the reciprocal lattice. Professor Ewald, with his wife Ella (who also attended the meeting) and their four children fled Nazi Germany in the 1930's and went to Cambridge in 1937, then to Queen's College, Belfast in 1939, and finally to the Polytechnic Institute of Brooklyn in 1949, where he remained until 1959. The Ewalds now live in Ithaca, New York.

The Dynamical Diffraction Symposium was a special part of the American Crystallographic Association meeting for 1978. The invited speakers were asked to submit manuscripts based on their talks for possible publication in *Acta Crystallographica* as a group, in homage to Professor Ewald. Each manuscript received was accorded normal editorial and refereeing treatment.

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Dynamical Diffraction Symposium

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A Review of my Papers on Crystal Optics 1912 to 1968*

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Abstract

The theory of the diffraction of X-rays by crystals was developed by Ewald as part of a unified study of the interaction of light of all wavelengths with crystals, beginning with the work for his 1912 thesis and extending to his papers in 1968. The formulation of the problem in terms of the interaction of electromagnetic radiation with a periodic array of dipoles is placed in its historical perspective and is compared with Laue's version based on the assumption of a continuous electron density distribution. The Borrmann effect, hinted at in 1917, is derived readily from consideration of the dispersion surface.

Introduction

On this happy occasion of a symposium celebrating my ninetieth birthday may I be excused for giving a review of my own papers? The main reason for this is that very few have read my original papers. Yet I see some merits in them as compared to later expositions of the same subjects. Besides, my work has been attempting to establish the unity of classical optics throughout the entire range of wavelengths from infrared to X-rays. This general aspect has received little resonance.

The papers I am going to comment on fall into two groups. There are four main papers under the title *Zur Bergründung der Kristalloptik* (*Foundations of Crystal Optics*; Ewald, 1916*a*,*b*, 1917, 1937; quoted hereafter as *Optics* I–IV). Of these the first is a slight remodelling of my Munich PhD thesis of 1912 *Dispersion und Doppelbrechung in Elektronengittern* (*Kristallen*) [*Dispersion and Double Refraction in Lattices of Electrons* (*Crystals*)] (Ewald, 1912), while the later papers

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* *Editors' note:* This summary of his contributions to crystal optics was prepared by Professor Ewald as a basis for his talk at the Oklahoma meeting and for private circulation. In response to insistent requests from the Editors, Professor Ewald has granted permission for the publication of the summary in full.

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develop the theory further, reaching a satisfactory generality in the fourth paper of the series. Surrounding this backbone of my work are shorter papers on topics of detail, some of which I shall mention when I have to comment upon them.

The optical field from assumed dipole oscillations

In 1910, when I began work on my thesis, there was no quantitative proof for the internal periodicity of crystals. My teacher Sommerfeld had the idea that a proof might be obtainable by investigating whether oscillating electrons or dipoles, when placed in an anisotropic periodic array, a lattice, would produce observable double refraction by the mere fact of that arrangement. It was hoped that if this were so, then the observed double refraction could be used to gain information on the internal structure of the crystals. Sommerfeld handed me the reprint of a paper by Planck (1902) on the theory of dispersion, and with that I went hiking up the Rhine Valley for the summer vacations.

The paper by Planck was quite incomprehensible to me. I could not disentangle the 'incident wave', the 'total field', and the 'field of excitation'. But I came to the conclusion that my first aim would be to find out what kind of field would be generated by the dipoles if they were set in motion by a plane optical wave travelling through the lattice with some unknown velocity q; that is, according to a 'wave of dipole excitation' described by a wave vector $\mathbf{K}$ whose length is ν/q, where ν is the frequency. Each dipole emits a spherical wavelet which travels away from it with the speed of light *in vacuo*, c, or with a wave constant $k_0 = \nu/c$. Because fields can be simply superimposed, to find the sum of all wavelets is a purely wave-kinematical exercise.

This summation kept me busy for about a year. Assuming an orthorhombic lattice with translations $\mathbf{a}_1$, $\mathbf{a}_2$, $\mathbf{a}_3$, the positions of the dipoles are given by $\mathbf{X}_l = l_1\mathbf{a}_1 + l_2\mathbf{a}_2 + l_3\mathbf{a}_3$ and each spherical wavelet proceeds according to the distance between the source and the field point $\mathbf{x} = x_1\mathbf{a}_1 + x_2\mathbf{a}_2 + x_3\mathbf{a}_3$, namely $r(l,x) = \{\sum_i [(x_i - l_i)\mathbf{a}_i]^2\}^{1/2}$. The irrationality of this expression prevents a straightforward summation.

A first helpful suggestion came from Sommerfeld. He showed me the development of the function expressing a spherical wave, namely $(\frac{1}{4}\pi r)\exp(2\pi ikr)$, into an integral over the product of a Bessel function of the planar distance ρ and an exponential function whose argument is linear in the third coordinate x_3 or z. He suggested that if it were possible to linearize ρ by using a similar integral technique, then it would be easy to sum the wavelets.

After weeks I found the proper integral technique and when I met Sommerfeld to show him this result, he said 'Wait, I also have to show you something' – and with that he showed me the same integral I had found. It is remarkable that neither of us spoke of the Fourier integral. At that time one spoke of Fourier developments, mainly in cosine and sine series, and the Fourier integral, in spite of its having been known for a long time, was still a bit strange; the term Fourier transform had yet to be invented, or to be commonly used in Germany.

The field generated by the dipoles was now easily calculated; it kept step with the 'wave of dipole excitation'.

The field of excitation and double refraction

My triumph at arriving at this field did not last long. I was now confronted with the task of extracting from this field that part that came from any individual dipole for which one sought to establish the motion. For it seemed obvious that a dipole would not be activated by its own field, but only by the fields radiated to its position by all the other dipoles. This, certainly, was the assumption upon which Planck and H. A. Lorentz based their theories of dispersion. The extraction of one dipole field seemed, however, hopeless because it was thoroughly mixed up with all others. Here a remark of Debye (at the time Sommerfeld's assistant) at a ski meeting in Mittenwald helped. He recalled a method Riemann had used in a similar case. Debye said 'Take the expression for the total field, get the denominator of the sum into the argument of an exponential function by introducing a new integration, and then, by interchanging summation and integration you obtain as the integrand a Θ function. To this you can apply the Θ transformation formula, and that will once more allow you to recognize the contribution of the individual dipole'. The method worked! I could now apply the field of excitation to the individual dipole and relate to it the amplitude of oscillation by the dipole's only physical constant, its polarizability.

There resulted the following situation: the optical field generated by the dipole oscillations had to be such that it just sustained these very oscillations. This condition could be fulfilled by the proper choice of the assumed wave vector $\mathbf{K}$ of the optical field, and with it of the refractive index $n = |\mathbf{K}|/k_0$. I called the situation obtained in this way one of 'dynamical balance'; since the work of Hartree it is now called a 'self-consistent' regime.

I was now able to conclude my thesis by a numerical calculation of the double refraction of a dipole lattice. On the recommendation of P. von Groth I used the axial ratios of anhydrite. The result was: no correlation between calculated and observed principal refractive indexes; however, indexes and double refraction of the same order of magnitude as observed; furthermore, the correct angular dependence of the optical properties as given by the various crystal-optical surfaces. Thus the characteristic optical properties of crystals were caused, at least partly, by the anisotropy of their internal arrangement.

I often feel embarrassed when the now generally accepted method of summation of potentials in crystals is given my name. True, I extended it later to non-orthogonal axes and I gave an explanation of the way it produces very rapid convergence – but essentially the method seems to go back to Riemann.

General insights gained in the thesis

My thesis brought with it several innovations for the then accepted theory of dispersion. This appelation was, in a way, a misnomer. Dispersion in the strict sense is the spreading of a pencil of light into a spectrum; it results from the dependence of the refractive index on the frequency, itself a consequence of the variation of the polarizability of the resonators with frequency. This dependence is nowadays the main objective of the quantum theory of dispersion which is based on our knowledge of the atom. How the polarizability affects the passage of light through a body is a second, now often neglected, aspect of the old designation 'theory of dispersion'. This part would better be called 'theory of light propagation'. Both the speed of light in a medium and its variation with frequency are *internal* properties of the medium. There was therefore in my thesis no room for the introduction of an 'incident wave' which played a role in the previous theories. In fact, my work established that selecting the proper phase velocity was the condition required for obtaining a self-consistent state of an optical wave in a medium containing resonators. This state was a free vibration of the medium in the same sense that in mechanics one speaks of the free vibrations of an unbounded string, plate or any other system where the internal forces balance and no external force is required.

Both the older theories and mine are essentially linear, which means that fields can be superimposed without their interaction. How then was it explained by the older theory that an incident wave falling from outside on a medium bounded by a plane surface could not be detected inside the medium? It would have phase velocity c, would not be refracted, and would thoroughly destroy the self-consistent state inside the medium. My answer was: this wave does not enter the medium; it is destroyed at the surface by the mere fact of the limitation of the array of dipoles.

As this statement was rather revolutionary, I was keen to get Laue's opinion on it. But his thoughts took another turn and he asked repeatedly 'What would happen in the case of very short waves?' I pointed out that the transformation of the optical potential was valid for all wavelengths and that his question could be easily discussed, but that I was preoccupied with finishing the write-up of my thesis and would have to leave it to him.

This concludes the report on my thesis which was handed in early in 1912 and published later in that year. The same material was slightly shortened and brought up to date when it was republished as *Optics* I in *Annalen der Physik* of 1916.

The optical potential in two forms

Let me make the first achievement of my thesis more precise, although in a more modern form, because it contains the key to all later papers.

Field quantities like the electric-field strength are best obtained by simple processes of differentiation from a vector potential which was introduced by H. Hertz in deriving the field of a single dipole of moment

$$\tilde{\mathbf{p}} = \mathbf{p}_0 \exp(-2\pi i \nu t), \tag{1}$$

namely

$$\mathbf{z}(\mathbf{x}) = (\tilde{\mathbf{p}}/4\pi r) \exp(2\pi i k_0 r); \tag{2}$$

here r is the distance of the fieldpoint $\mathbf{x}$ from the dipole and $k_0 = \nu/c$ is the constant of wave propagation in free space.

We have been considering dipole moments phased according to a progressive wave of an unspecified wave vector $\mathbf{K}$; therefore, the amplitude $\mathbf{p}_0$ now becomes $\mathbf{p}_0 \exp[2\pi i(\mathbf{KX})]$, where $\mathbf{X}$ is the position of the dipole and $(\mathbf{KX})$ is the scalar product of the two vectors. The distance from dipole to field point $\mathbf{x}$ is then $r = |\mathbf{x} - \mathbf{X}|$ and the total Hertz potential of the optical field at point $\mathbf{x}$ appears as a sum over all sources of wavelets, that is over the three integers (l_1, l_2, l_3) contained in $\mathbf{X}_l$:

$$\mathbf{z}(\mathbf{x}) = \sum_l \frac{P_0 \exp(j\mathbf{K}\mathbf{X}_l)}{4\pi|\mathbf{x} - \mathbf{X}|} \exp(jk_0|\mathbf{x} - \mathbf{X}_l|) \quad (j \equiv 2\pi i). \tag{3}$$

In its transformed shape the same $\mathbf{z}(\mathbf{x})$ is found to be a sum of plane waves:

$$\mathbf{z}(\mathbf{x}) = v_a^{-1} \sum_h p_0/[4\pi^2(K_h^2 - k_0^2)] \exp[j(K_h x)]; \tag{3'}$$

here the summation is extended over all points $\mathbf{h} = (h_1\mathbf{b}_1 + h_2\mathbf{b}_2 + h_3\mathbf{b}_3)$ of the 'reciprocal lattice'; the wave vectors $\mathbf{K}_h$ connect the 'tie point' T (*Ausbreitungspunkt*) to the points $\mathbf{h}$ of the reciprocal lattice, and the tie point is defined as the point $-\mathbf{K}_1$, where $\mathbf{K}_1$ (which above was called simply $\mathbf{K}$) is the assumed wave vector of the dipole phasing. The $\mathbf{b}_i$ are the reciprocal vectors to the $\mathbf{a}_i$.

The transition from the source-conscious form (3) to the plane-wave form (3′) can nowadays easily be made by applying the technique of Fourier transforms; in the original papers complex integrals were evaluated by the method of residues.

The reciprocal lattice was not yet explicitly envisaged in the thesis; its importance became manifest only in connection with X-ray diffraction. Also it was sufficient to take the simplest model of a crystal for clarifying the optics, namely a simple orthorhombic lattice. Generalization to non-orthogonal axes occurs only in *Optics* III, while the restriction to a basis of a single dipole could be dropped only in *Optics* IV.

The impact of Laue's discovery

Laue, Friedrich and Knipping found X-ray diffraction in crystals round about Easter 1912. I had become an assistant in Göttingen, and I first heard of the discovery some seven weeks later when Sommerfeld came to deliver a lecture on this discovery to the Göttingen Physical Society. Scientific news travelled slowly in those days.

The evening after Sommerfeld's lecture, I finally sat down to discuss the case of short wavelengths. It was hereby that the reciprocal lattice became an essential tool and that the 'sphere of reflexion' (Bragg) or '*Ausbreitungskugel*' was an obvious construction (Ewald, 1913).

Naturally, interest soon became centered on X-ray diffraction. Methods of indexing had to be worked out. One of the early problems was why many more spots did not appear on the Laue diagrams. In a discussion of the original Laue diagrams of zinc blende which Sommerfeld prepared roughly for the second Solvay Conference in 1913 and which I worked out, this question was answered; here the concept and name of the structure factor appears for the first time. In 1914 W. L. Bragg had succeeded in determining what he then called 'the terribly complicated' structure of pyrites, FeS_2, the first example of a cubic structure with non-intersecting trigonal axes and the second type involving a parameter. For the latter, Bragg found a value from the spectrometer readings. By considering the structure factor as a function of the parameter I derived from Friedrich's Laue photograph of pyrites a different and much more accurate value of the parameter – in fact, this may be considered the first precision determination of a parameter (Ewald & Friedrich, 1914).

War service; the extinction theorem

In August 1914 war broke out. The X-ray equipment was transferred from Sommerfeld's institute to a hospital for medical work, and I accompanied it. In 1915 I was attached to the army and went, together with a mobile X-ray station, to the Lithuanian front. It took several weeks before the unit made its way from the Siemens factory in Berlin to Königsberg in East Prussia, where I was waiting for its arrival. During this period of forced leisure I returned to the nagging problem left over from my thesis, namely that of the cancellation of the incident field.

In order to prove the extinction statement, I had to know the optical field generated by a 'half-crystal', that is by a lattice of phased dipoles filling only the space under the plane $z = 0$. This was again a problem of wavelet summation. I tackled it with the familiar methods of evaluating complex integrals by the method of residues. I stuck to the assumption of an orthorhombic lattice because then the integrals could be evaluated separately for the three orthogonal variables.

The result of the summation was that the field inside the crystal consists of the same waves as in the unbounded crystal ('mesowaves'), but that each such wave is accompanied by a 'boundary wave' ('epiwave') of the same amplitude, opposite phase, wave constant k_0 (instead of $|K_h|$) and wave vector $\bar{\mathbf{K}}_h$ such that $\bar{\mathbf{K}}_h$ and $\mathbf{K}_h$ have the same tangential component along the surface. The last condition is an expression of Snell's law of refraction: it assures that the traces of meso- and epiwave move along the surface of the half-crystal with the same speed, so that if extinction occurs at one point of the surface, it is true for all points.

Besides the epiwave inside the half-crystal, there exists an outer epiwave whose wave vector $\bar{\bar{\mathbf{K}}}_h$ is the mirror image of that of the inner epiwave with respect to the surface.

For light, only the term 000 of the sums yields three ordinary plane waves, namely the mesowave and the two epiwaves, which form, respectively, the refracted, the reflected and the extinguishing waves. All other terms in the sums represent 'inhomogeneous waves' whose complex wave vectors have a real part directed along the surface and an imaginary part normal to the surface. The latter produces a rapidly decreasing amplitude along the wave front with increasing distance from the surface. For visible light this inhomogeneity is so strong that the amplitude diminishes from the top layer of dipoles to the next one at depth d by at least a factor of 1000. The inhomogeneous waves mediate the transition from the field outside the crystal to that inside; in fact, there exists a region just beyond the surface, namely $0 < z < d$, where the description of the field can be given by the formulas for either the inside or the outside space. After cancellation of the incident wave, there remained the well known self-consistent optical field inside the crystal and the reflected wave outside. The Fresnel formulas for the amplitudes of these waves resulted.

The investigation supported in all detail the extinction theorem for visible light. It proved invaluable in its more complicated application in the theory of X-ray diffraction. I was happy to mail the manuscript to Sommerfeld for publication as a sequal to the 1912 thesis before my X-ray unit arrived in Königsberg (Ewald, 1916*a*,*b*).

Theory of X-ray diffraction

Hardly any fighting went on across the Dwina River in November 1915 when my car and I were installed in a makeshift hospital some 10 miles behind the front. Exhausted troops were sent there from the French front to recover – mostly old men no longer fit for active fighting and prone to break their brittle bones on the icy morass into which the few roads had been turned by the military traffic. I now had time to consider the 'case of short wavelengths' not in the purely geometrical sense, but as the dynamical problem of how an X-ray optical field could travel in the crystal.

The first step was to recognize the importance of the 'reciprocity theorem' which I had already stated in the paper written after Sommerfeld's lecture in Göttingen. If n points of the reciprocal lattice lie on the sphere of reflection, then the corresponding n rays form an inseparable unit, in the sense that none of them can exist without engendering all others. Thus a bundle of plane waves takes the place of the single plane wave in the light case. In the unbounded medium this composite field has to be subjected to the condition of self-consistency. The adapter for achieving this is the choice of the length of the wave vector of one of the rays; by this, the length of all other wave vectors is determined, or as we now say, the common origin of these vectors, the 'tie point' T, is fixed. The refractive index for X-rays differs from 1 by only about one part in a million; this means that the difference in length between any of the wave vectors $\mathbf{K}_h$ and the vacuum value k_0 is of that order of magnitude. There is no general refractive index; each of the n rays has its own. In spite of the nearly equal lengths of the wave vectors, their differences regulate the amplitudes of the waves according to the 'resonance factors' $(K_h^2 - k_0^2)^{-1}$ in the potential (3′). This all-important influence can be understood by a Fresnel zone argument: the smaller the difference $(K_h^2 - k_0^2)$, the less difference there is between the assumed phase velocity q of the dipole excitation and the velocity c with which the spherical wavelets travel; therefore, the greater is the number of dipole-containing planes which send their contributions approximately in the same phase to a field point $\mathbf{x}$, and the greater is the build-up of their amplitudes. Thus in order to balance the amplitudes of the n co-existing beams, so that none gains or loses amplitude (and energy) from the other beams, the lengths of all wave vectors have to be finely adjusted, and with this adjustment the amplitude ratios of the n beams in a self-consistent mode are fixed.

The simple geometrical expression for this intricate balancing is the 'surface of dispersion' on which the tie point must lie. The general equation of the surface is given in *Optics* III for the case of n co-existing rays. The restriction to an orthorhombic lattice has been dropped, but not that of having only one dipole per cell. The general properties of the surface are discussed. It is shown to consist of $2n$ sheets, corresponding to the number of amplitude components of n transverse waves. There are then $2n$ different modes of the optical field. The unrealistic condition of the kinematical theory, that for diffracted rays to exist the sphere must pass *exactly* through the points of the reciprocal lattice, is now softened to the condition of a close approach. Finally the important case of two rays is discussed in detail.

In order to obtain results that can be checked by experiment the case of the half-crystal has to be treated upon whose surface a plane X-ray wave is incident. There are now n homogeneous waves forming the mesofield, each of them accompanied by an internal epiwave of phase velocity c whose wave vector $\overline{\mathbf{K}}_h$ has the same component along the surface as the mesowave vector $\mathbf{K}_h$. In the outer space there are 'reflected' epiwaves whose vectors $\overline{\overline{\mathbf{K}}}_h$ are the mirror images of $\overline{\mathbf{K}}_h$ across the surface, but their amplitudes are not the same. Besides, there are inhomogeneous waves travelling along the surface whose amplitude is restricted to the first few dipole layers. The condition of self-consistency inside the crystal can be fulfilled by superimposing dynamically balanced proper modes with such amplitudes that in $(n - 1)$ directions the internal epiwaves cancel and that in the remaining direction, chosen as (000), the sum of the internal epiwaves cancels the incident wave. This is achieved by superimposing fields represented by $2n$ tie points T, T^*, ..., T^{2n*} situated on the $2n$ sheets of the surface of dispersion along the direction of the surface normal, each field taken with a suitable amplitude. The result is that in each direction of diffracted rays a bundle of plane waves of very slightly differing wave vectors $\mathbf{K}_h^*$, $\mathbf{K}_h^{**}$, ..., $\mathbf{K}_h^{2n*}$ is propagated; these waves form beats depending on the depth below the surface. Energy is exchanged between the rays (*Pendellösung*) and a group velocity for the propagation of energy through the crystal can be defined.

The full solution was easy to establish in the case when all diffracted rays travel from the surface of incidence towards the interior ('Laue case'). It was shown that in this case all $2n$ intersections of the surface normal with the sheets of the surface of dispersion are real, that the solution is fully determined, that the primary ray (000) at the upper surface takes over the energy of the incident ray and that the diffracted rays start with zero amplitude before taking out energy from the primary ray. There is a perfect analogy to a system of n coupled pendulums one of which receives at time $t = 0$ an impulse while the others are at rest. The initial energy is then transferred through the coupling to the other parts of the system, and if there is no damping beats will continue forever. The analogy of the spatial behavior of the diffracted rays and the time development of the motion of coupled pendulums has been stressed by me on many occasions. The method followed in the dynamical theory is the strict analogue to the theory of small oscillations in classical mechanics.

In the non-Laue case at least one upwards-directed diffracted ray leaves the half-crystal by the surface upon which incidence occurs. This may be the directly reflected or any other diffracted ray. In either case this internal ray produces an equally strong epiwave only in the outer half-space. It does not produce a strong internal epiwave, and it therefore leads to no condition of annihilation in the interior. The amplitudes remain indeterminate. This dilemma is physically justified because of the unrealistic assumption that the half-crystal fills the entire lower half-space. Sufficient conditions are obtained for making the problem, and the answer, precise by assuming a second boundary plane parallel to the upper one, that is at depth D. The drawback of this assumption is that the answer now depends on the thickness D of the crystal plate and that it is complicated by the interference effects between the waves reflected at the upper and the lower boundaries, like in the theory of the Lummer–Gehrcke plate. In order to get rid of this unwanted complication and because in actual crystals the thickness D is rarely constant, I averaged over D. There resulted in the case of two rays the well known 'top hat' reflexion curve. This is, in fact, the curve already found by C. G. Darwin in 1913. It shows the angular region of complex wave vectors and therefore total reflexion, bordered by a falling-off intensity in the Lummer–Gehrcke regions. Prins (1930) refined the curve by including absorption and superimposing for the two cases of polarization and in due course this curve was confirmed with ever-increasing resolution on selected crystals.

Comparison of my theory with Laue's version

Laue's version of the theory dates from 1931 (Laue, 1931), that is five years after wave mechanics had changed our ideas of the constitution of atoms and solids and two years after Bragg and West had shown the usefulness of Fourier methods in the determination of crystal structures. The starting point for Laue is the assumption that each cell of the crystal structure is filled with a continuous electron density and dielectric constant, and that Maxwell's equations can be applied to this medium. For the half-crystal a continuous boundary is assumed, across which the usual boundary conditions for the field can be used.

It seems strange to transfer the concept of dielectric constant, derived as it is from large-scale experiments, to the spaces between atoms. True, H. A. Lorentz had shown in his theory of electrons how in a model medium containing only positive and negative point charges the material constants of the Maxwellian theory can be obtained by considering average values of fields over regions containing many point charges. But this kind of averaging is not applicable to the rapidly varying X-ray fields. A similar objection can be raised against the application of boundary conditions to electron-density and dielectric-constant distributions which protrude into the upper half-space.

In contrast to this, the model I used, even if it may be less realistic, is that of an open structure: there is no closed boundary, the external field is superimposed up to any depth in the medium and the condition of self-consistency holds everywhere. It is logically the simpler model.

In spite of the conceptual differences, Laue obtained the same results as I did. His assumption of the dielectric constant was justified by Kohler (1935) on the basis of wave mechanics. Schrödinger's wave-mechanical perturbation theory also showed that each element of volume of electron density reacted to an optical field like a dipole; and with that the models became fundamentally the same, provided I could carry my approach through to a basis containing an arbitrary number of dipoles. I had achieved this already in 1925 for the case of two rays, but not for the general case of an arbitrary number; Laue's theory and discussion did not go farther than this.

My 'habilitation'; Borrmann effect

When World War I neared its end I used my theory of X-ray diffraction as a thesis for being admitted as a lecturer at the University of Munich. Even though Sommerfeld thought that my speculations would never find an application, he accepted the thesis. I had to give a trial lecture and put up a number of statements which I was willing to defend against interventions by the faculty. I had lost contact with physics during the solitary work on my problem and found it difficult to formulate significant statements. The second of my statements was based on the insight I had gained from my work. It seemed rather strange at a time when absorption of X-rays was considered to be entirely conditioned by the amount of matter transversed. It read (in translation): *In case the absorption of X-rays like that of light can be traced to a consumption of energy in the oscillations of dipoles then under some circumstances diffracted X-rays will not suffer any weakening in an absorbing crystal.*

This statement was based on my knowledge that the field for compensating the incident field would be produced by a dipole amplitude which would be smaller the closer the tie point lies to the 'Laue point', *i.e.* to the only point giving rise to diffracted rays according to the kinematical theory. This statement in December 1917 is a prediction of the effect G. Borrmann discovered experimentally in 1941 (Borrmann, 1941); Laue deduced its laws while he was held 'at His Majesty's pleasure' in Farmhall at the end of the European war (Laue, 1949). In 1917 X-ray absorption, like emission, was known to be a typical quantum effect; therefore the carefully worded beginning of the statement. I forgot the statement even after Laue's explanation of the effect until some fifty years later I happened to come across a copy of the invitation to the inaugural meeting.

Preliminary summary

By 1918 the dynamical theory of X-ray diffraction had been successfully established, first for an orthorhombic lattice, later for a general lattice. The theory was, however, still restricted to a basis of a single point-dipole. No structure factor occurred. The general case of n rays had been treated, but only the case $n = 2$ had been discussed in sufficient detail to check with experiment. This comparison had to wait for many improvements in the resolution of the experimental methods and the availability of near-perfect crystals.

Book and *Handbook*; amplification of the reciprocal lattice

In 1921 I left Munich for a chair in theoretical physics at the Polytechnic School in Stuttgart. Besides assembling a full course of lectures in the first years, I wrote a book: *Kristalle und Röntgenstrahlen* (Ewald, 1923). The way I treated the subject was broad and introductory. Later on, I hated to read in it, but judging by the unexpected compliments from crystallographers of many nations and after many years had passed, it must have served its purpose well. Characteristically neither the dynamical theory, nor even intensity, is mentioned in the book; both were in my view subjects about which too little was known at the time. The book contained a list of all known structures which I meant to continue in a second edition. This became the origin of *Strukturbericht*. In spite of its rapid sale the book never achieved a second edition because I was soon writing a full and concise review of the subject for the *Handbuch der Physik*, which was more to my liking. The 1927 edition of this report (Ewald, 1927) was followed in 1933 by a second edition which was brought up to date (Ewald, 1933).

The last manuscript I finished in Munich (Ewald, 1921) was prompted by a request of the Editor of *Zeitschrift für Kristallographie*, Paul Niggli, as he took over from P. von Groth. The 'polar lattice' had been used by the older morphologists only for determining the orientation of the normals to crystal faces with respect to a set of axes. Because of X-ray diffraction one now became interested both in the orientation and the spacing of atomic net planes. The spacing was the inverse of the spacing of a row of points of the reciprocal lattice passing through the origin; this relation required the 'polar axes' to be renormalized to 'reciprocal axes' in the way Willard Gibbs had defined these, and the reciprocal lattice formed by using these axes as translations.

In crystal structures we were now confronted with motifs consisting of many atoms, repeated by the translations. In the obvious point-by-point description of the structure the coordinates of each atom are listed. Could not the structure also be described by referring only to atomic planes? This is a purely geometrical question, and the answer is: yes – but it was very surprising at the time. No new lattice points can be added to those of the reciprocal lattice for they would indicate irrational positions of net planes. But one can add 'weights' to the existing reciprocal-lattice points. These must be invariant against changes in the description of the structure, such as doubling an axis and the basis. To my astonishment the weights necessary to achieve invariance turned out to be identical with the structure factors! The entire row of reciprocal-lattice points passing through the origin suddenly acquired significance: their weights were the Fourier coefficients representing the sequence and population density of the atomic planes normal to the row.

With this result my geometrically conceived extension of the reciprocal lattice tied up with the suggestion by W. H. Bragg (1915) to use Fourier series and their coefficients in determining crystal structures. This method, first tried out by W. L. Bragg and West in 1929 on diopside became more generally applied when the technique of summing Fourier series in two dimensions had been facilitated by the Beevers and Lipson strips in 1934, the same year in which A. L. Patterson showed what use can be made of intensities instead of the unknown Fourier amplitudes. This last problem had been dealt with already in my 1921 paper although in algebraic terms, anticipating convolution and stressing that only the differences of atomic positions are obtainable from the intensities.

The Paris lectures

In 1932 I returned once more to the dynamical theory on the occasion of a lecture series I gave at the Institut

Henri Poincaré in Paris (Ewald, 1938). Here again I stressed the continuity of my approach for all parts of the spectrum, treating with greater care the determination of the field of excitation. The equation of motion of a single isolated dipole requires a damping term because of the loss of energy by radiation. This can only come from the action of its own field on the dipole. By a more thorough discussion than in my previous papers the field radiated to a dipole forming part of a lattice from the other dipoles is shown exactly to cancel the damping term in the dipole's equation of motion. There is no loss of energy by radiation for the individual dipole in a lattice if it contributes to the self-propagating plane-wave optical field. This result was already found by H. A. Lorentz for a dipole in a periodic medium. The paper ends with the derivation of the crystal-optical surfaces, the 'surface of normals' for visible light and the 'surface of dispersion' for X-rays.

Optics IV

Up to this point the dynamical theory had been able to deal in a general way only with crystals having a basis of a single dipole. Already in 1925 I had solved the case of a general basis, but only for one diffracted beam ($n = 2$) (Ewald, 1925). For this most important case the result was that for a perfect crystal the diffracted intensity is proportional to $|F_h|$, where F_h is the structure amplitude, and not as for the mosaic crystal to $|F_h|^2$. The reason for this result was published, but not the detailed derivation.

In 1937 I could finally return to the general problem of an arbitrary basis and an arbitrary number n of diffracted rays. The difficulty of this generalization lies in the following fact: As long as there is only a single point-dipole at the origin of the crystal cell all vectors $\mathbf{K}_h = \mathbf{K}_{000} + \mathbf{h}$, where $\mathbf{h}$ is a lattice vector, describe the same oscillations of the dipoles. The difference of description lies only in interpolating a smaller or greater number of sinusoidal waves in the empty space between the dipoles, and this is physically irrelevant. If, however, a second dipole is placed somewhere inside the cell, then it will pick up a different phase according to each $\mathbf{K}_h$. For the scattering of one beam into another there will thus result a different phase relation between the atoms of the basis according to the value of $\mathbf{h}$. Thus for each scattering process there exists a different moment of the entire basis. This is the vectorial structure amplitude

$$\mathbf{S}_h = \sum_s \mathbf{p}^s \exp\left[-j(\mathbf{h}\mathbf{x}^s)\right] \quad (j \equiv 2\pi i); \tag{4}$$

the summation goes over all dipoles in the cell, each sort marked by an index s and shifted by $\mathbf{x}^s$ from the origin of the cell.

Self-consistency of the optical field now requires that

$$\mathbf{S}_h = \sum_{i=1}^{n} A_{h-i}\, \mathbf{S}_{i\perp \mathbf{K}_i} \frac{k_0^2}{K_i^2 - k_0^2}, \tag{5}$$

where

$$A_m = \sum_s (\alpha_s/v_a) \exp\left[-j(\mathbf{h}_m \mathbf{x}^s)\right] \quad (v_a = \text{volume of cell}). \tag{5'}$$

It will be seen that A_m is nothing other than the Fourier coefficient in the development of the distributed polarizability in the cell. $\mathbf{S}_{i\perp \mathbf{K}_i}$ is the part of the moment of the cell that is effective in producing a transverse optical field progressing in the direction of the wave vector $\mathbf{K}_i$.

Equations (5) and (5′) are linear homogeneous equations for the n structure amplitudes $\mathbf{S}_h$; they can be solved only on condition that one of the vectors $\mathbf{K}$ be chosen properly. After splitting the $\mathbf{S}_h$ into a component transverse to $\mathbf{K}_h$ and another one parallel to it the condition of solubility takes the form of the vanishing of the product of two determinants. This is done in a general way for n rays, whereby the restriction that these be 'strong' rays becomes immaterial. Thus one should be able to take into account points of the reciprocal lattice lying inside or outside the sphere of propagation at some distance from its surface. This may be of value in discussing the optics of the ultrasoft X-ray region which forms the transition to light optics.

Needless to say, the general determinantal condition leads to the old results if $n = 2$, and if n unlimited in the case of a single dipole basis.

The enhanced Borrmann effect

Borrmann & Hartwig (1965) discovered that when three strong rays coexist in a near-perfect Si or Ge crystal the absorption drops even below the value reached in the case of only two rays. The condition of coexistence limits this 'enhanced Borrmann effect' to a small angular region; thus when the crystal is rotated under preservation of the reflecting condition for one set of net planes then within the dark line on the film which is produced by the simple effect there will be found a small darker spot due to the enhancement. The simple interpretation of this spot is that for three beams the surface of dispersion approaches the Laue point even closer than that for two beams. This prompted the papers by Y. Héno and myself (Ewald & Héno, 1968; Héno & Ewald, 1968) [(I) and (II)]. In them the surface of dispersion was analytically and geometrically discussed for the special three-beam case observed. It is shown in (I) that for $n = 3$ one sheet of the surface of dispersion passes at a smaller distance from the Laue point than does any of the three surfaces for only two out of the three beams. Thus, without even introducing absorption, the origin of the enhancement is manifest from the general resonance denominator $(K_h^2 - k_0^2)^{-1}$. In (II) absorption and atomic factors are introduced and a graphical and numerical rendering of the surface of dispersion is given for variable wavelength and, consequently, angular settings. Finally the effective coefficient of absorption, μ_{eff}, is given for two different three-beam cases in germanium for each of the six sheets of the surface of dispersion. The curves show μ_{eff} as a function of λ/a for the cases $n = 1$, 2 and 3. They show strikingly to what extent a perfect crystal can become transmittant in one of its proper modes, while in others it becomes more opaque. The value of this analytical treatment lies in the general survey it provides, and as such it is satisfactory. On the other hand, it is labor saving to use computer methods in future cases.

Retrospect

I am lucky to have spent so many of my best years in doing research I have loved; in having been well prepared for Laue's discovery by my thesis work; by taking an active part in developing the theory of the subject; by gradually simplifying the mathematics and by arriving at an increasingly deeper understanding of the varied aspects of crystal optics.

On the other hand, my interest always centered on the perfect crystal in which I saw the preferred material for exacting optical investigation. Herein lies a strong limitation, and an abstraction which in important aspects is contrary to nature. I am happy to see how others have been, still are, and will be carrying on beyond the limitations I set for myself.

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On Extinction.
III. An Improvement of the Secondary-Extinction Theory*

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Abstract

The previous theory [Kato (1976). *Acta Cryst.* A**32**, 458–466] is improved by taking into account the higher-order correlations of lattice phase factors. The previous coupling constants in the energy-transfer equations have, in general, to be reduced. In the simplest case of non-absorbing crystals the reduction factor can be given as

$$R = 1 + (\tau_2)^{-1} \sum_{j=1}^{\infty} V_2^{(j+1)}(-\kappa_g \kappa_{-g})^j,$$

where $\kappa_{\pm g}$ is the kinematical diffraction amplitude per unit length for $\pm\mathbf{g}$ reflection, and $V_2^{(j)}$ is the correlation

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