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Introduction to the Dynamical Theory of X-Ray Diffraction

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The general features, terminology, and method of the dynamical theroy of X-ray diffraction are discussed, stressing the analogy with the general theory of small oscillations of a mechanical system.

My good friend the organizer of this session prepared me to speak to an audience consisting partly of experts in the applications of this theory and partly to people knowing the theory only by name. I shall dwell, therefore, on the method of the theory rather than on results or applications, of which the other papers of this session will contain examples.

Theories of X-ray diffraction

The geometrical theory, the kinematical theory, the dynamical theory and the developing quantum mechanical theory of X-ray diffraction can be regarded as successive stages of accounting for the same physical phenomena.

(i) In the most primitive stage, the *geometrical theory*, we ask only for the directions under which diffracted rays appear, *i.e.* those directions in which the wavelets scattered at each element of a crystal will reinforce one another without any cancellations. This theory alone leads to the concepts illustrated in Fig. 1, namely the *reciprocal lattice*, the *tiepoint* T determined by making $\overrightarrow{TO}=k_1$ the wave vector of the incident wave, and the *sphere of reflection* of radius k_0 with T as centre. Whenever this sphere passes through a lattice point, such as $\mathbf{h}$, the vector $\vec{T}\mathbf{h}=\mathbf{k}_h$ becomes the wave vector of a diffracted wave.

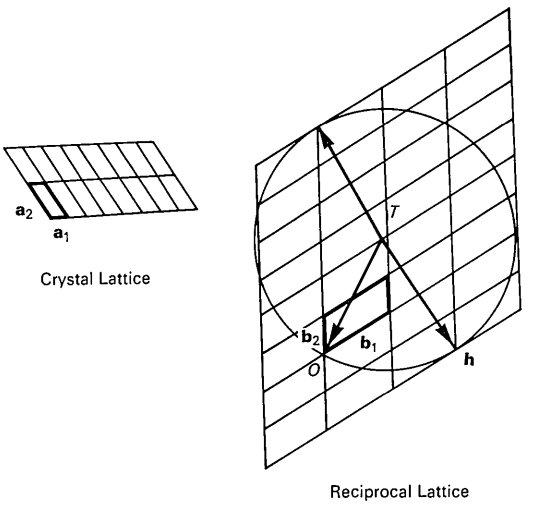

Fig. 1. Crystal lattice, reciprocal lattice, tiepoint T, sphere of reflection and wave-vectors of the primary and two of the secondary rays.

This simplest, geometrical theory leads to the *structure factor* by considering the differences of optical path caused by the dispersion of the atoms throughout the crystal cell. Thereby it opens the way towards crystal structure analysis of a primitive kind, namely by the use of absent reflections.

(ii) In the next stage, the *kinematical theory*, the combined effect of the scattered wavelets in directions other than those of maximum cooperation is taken account of. This requires a relaxation of the strict condition of periodicity of the crystal, a condition which would prevent the crystal from being bounded by a surface or of having a finite size, and which leads to the 'all or nothing' condition that a point of the reciprocal lattice lie exactly on the sphere of reflection. The scattering power – be it electron density $\varrho(x)$ for X-rays, or potential $V(x)$ for electrons – is rendered by a Fourier series in the case of the truly periodic crystal; but for any deviation from periodicity it is rendered by a Fourier integral, and its Fourier transform $F(\eta)$ is a continuous function in Fourier space. For slight departures from periodicity, such as finite size of the crystal, or temperature motion, the transform $F(\eta)$ will still have strong peaks at the positions $\boldsymbol{\eta}=\mathbf{h}$ but each lattice point $\mathbf{h}$ will be surrounded by a kind of halo of $F(\eta)$ instead of only carrying, in a δ-function like way, a Fourier coefficient $F_{\mathbf{h}}$. The purely mathematical and physically unreasonable 'all or nothing' condition of intersection of sphere of reflection and lattice point is now replaced by the more generous result of the superposition of the elementary wavelets, namely that the amplitudes of the diffracted waves at great distance are shown, for wave vectors of any direction, by the intersection of the sphere of reflection with the Fourier transform $F(\eta)$ of the scattering density $\varrho(x)$. This extension of the theory leads to the concept of *Lorentz factor* and thereby to a first primitive use of integral intensities for the determination of parameters in crystal structures. It also accounts for the broadening of the diffraction lines of fine powders, and for a host of other properties.

In the kinematical theory all waves are supposed to travel through the crystal with the same phase velocity.

This is generally taken to be c, the light velocity in empty space, and then all wave vectors have the same length $k_0 = \nu/c = 1/\lambda_0$ where λ_0 is the wave-length in vacuo. In that case the sphere of reflection, which always has radius k_0, passes through the origin O of Fourier space.

The kinematical theory forms the base for all crystal analysis and for the bulk of all work from 1912 on to now. But it has the serious shortcoming of not conserving energy since all energy contained in the diffracted rays is additional to that of the unmodified incident beam. The way to get rid of this defect is apparent from Fig. 1 and the obvious *reciprocity theorem*. This states that if any one of the n strong waves is considered to be the primary wave, the same set of n waves will be generated. Shifting the origin from O to any of the points $\mathbf{h}$ lying on the sphere makes no difference to the position of T nor of the sphere; only the order vectors will be re-named.

The n rays thus form an inseparable unit which we call the *optical field*; it takes the place of the single plane wave forming the simplest optical field in visible light optics.

(iii) In the *dynamical theory* there are two problems to be solved. The first is to find the conditions under which an n-ray field can exist in, and travel through, the crystal. This is an extension of the optical '*theory of dispersion*' to the short wavelengths of order 10^{-8} cm instead of $5 \cdot 10^{-5}$ cm in the visible range. The second problem is that of connecting the fields inside the crystal to those outside, including the incident wave; the answer is the '*theory of refraction and reflection*'. For the propagation of optical fields inside the crystal, we may assume this to be unbounded, *i.e.* filling all space. There is no outside and no '*incident wave*'. For the latter to appear we have to assume at least one surface to exist – we speak of the '*half-crystal*' filling the space below the plane $z=0$, or of a crystal slab limited by the planes $z=0$ and $z=D$.

In the dynamical theory interaction between the scattered or diffracted rays is taken into account; apart from the scattering there is no interaction between the radiation field and the crystal. If temperature motion and absorption are introduced, this is done in an *ad hoc* descriptive manner, which is not very satisfactory even if it is capable of rendering the experimental facts.

(iv) The theory of the future may well be the *quantum theory of diffraction* in which the radiation field and the state of the crystal are considered as parts of a single system. Such a theory was first formulated by M. Born in 1942 and it is being actively developed, in modern terms of Feynman diagrams by Ohtsuki (1964), Kuriyama (1966) and others. In quantum language the kinematical theory represents the simple collision of photon and crystal; the dynamical theory gives the multiple collision of photon and crystal. In both cases the crystal remains in its ground state. In the quantum theory multiple scattering occurs together with a possible change of state of the crystal, by collision of the photon with a phonon or with an electron, leading to photoeffects or to the formation of excited states as a preparation for the emission of characteristic radiation or for Auger effects. If this theory can be carried through in a realistic way, it would make the *ad hoc* assumptions for the incorporation of temperature effects, Compton effect and absorption obsolete.

Analogy between the dynamical theory and the theory of small oscillations in mechanics

Before returning to the 'dynamical' theory let us consider a well-known theory in particle dynamics, the theory of small oscillations of a mechanical system about a position of stable equilibrium. To fix the ideas, consider a system of n equal pendulums coupled by a taut string from which they are hung at equal distances. These pendulums are the counterparts of the n rays forming the optical field. In comparing the mechanical and the optical problems, time dependence in the first corresponds to space dependence in the latter. Thus the problem of exciting the n-wave optical field in a half-crystal by a wave incident on the surface $z=0$ corresponds to finding the motion of the row of pendulums after an impact on one of them at time $t=0$. How do we find this motion for times $t>0$?

The standard method is first to study the *proper modes* of the system, namely those motions which are self-consistent in the sense that each part of the system moves without the application of external forces under its intrinsic forces (gravity in the case of pendulums) and the forces transmitted to it by being coupled to its neighbours. Such proper modes can only be found for certain frequencies of the motion, called the *proper frequencies* $\nu^{(j)}$; they are the (always real) roots of an equation of order n, the *secular equation* or *equation of dispersion*. The set of frequencies $\nu^{(1)} \cdots \nu^{(n)}$ forms the '*spectrum*' of the mechanical system. For each proper frequency $\nu^{(j)}$ the amplitudes of the n pendulums, $A_1^{(j)}, A_2^{(j)} \cdots A_n^{(j)}$ stand in a fully determined ratio, and the motion can further be 'normalized' by making $\sum_{h=1}^{n} |A_h^{(j)}|^2 = 1$; the motion is then called a *proper mode*.

The optical analogue to this part of the solution of the mechanical problem is the *theory of dispersion*. In it, we find a self-consistent field of n waves, *i.e.* one in which each wave gives off as much energy to the others, as it receives from them *via* the coupling by means of scattering by the atoms. The amplitudes of the waves therefore remain constant during the progression of the field through the crystal. This corresponds to the constant amplitude simple harmonic motion of each pendulum in a proper mode.

Such a state of self-consistency can not be established for an arbitrary tiepoint, but only for tiepoints lying on a surface of degree $2n$, the *surface of dispersion* (SurfDisp). (The factor 2 arises because a transverse wave has two independent amplitude components; it

also holds for the pendulums if they are allowed vibrations in the x and y directions.) The SurfDisp is thus the analogue to the 'spectrum' of the row of pendulums; to indicate that a tiepoint lies on this surface, we speak of a '*proper tiepoint*'. It represents n waves of well-determined amplitude ratios which form the self-consistent optical field or '*proper X-field*'. Having determined the SurfDisp and proper X-field we know all there is to be known about fields inside the crystal.

We have now to tackle the problem of starting the motion of the pendulums from rest by a blow given to the 'primary' pendulum at $t=0$. This blow imparts a certain initial velocity to the number one pendulum, without directly affecting its position or the positions and zero velocities of the others. After the initial impact is over, the subsequent motion of the system can only consist of the superposition of its proper modes, since external forces are no longer acting. Optically, the corresponding situation is that the external '*incident*' *wave* creates at the surface of the half-crystal a '*primary*' *wave* of the same energy or amplitude. As this wave travels towards the interior ($z>0$, corresponding to $t>0$ for the pendulums) it is subjected to the coupling by radiation to the other directions and it gives off energy to the other $n-1$ waves which start with zero amplitude at $z=0$. Since the incident wave has been converted into the primary wave at the surface, it is not influencing the optical field inside the crystal any more, so that the field at $z>0$ can only be the superposition of proper X-fields.

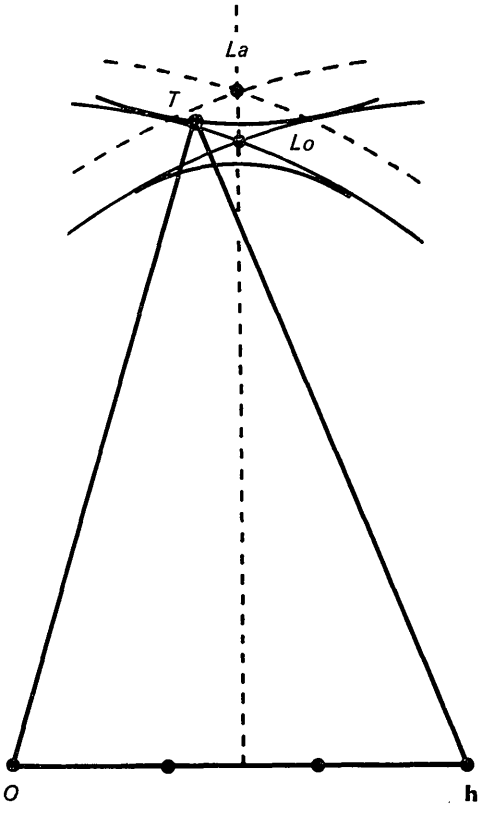

Fig. 2. The Lorentz and Laue spheres about the reciprocal lattice points **O** and **h**; branching of the Lorentz spheres near the Lorentz point *Lo*.

We have thus to superimpose (normalized) proper fields with such amplitudes that at $z=0$ the resultant amplitude is E_0, namely that of the incident wave, in the 'primary' direction, and zero in all secondary directions. Such a condition must hold at all points of the crystal surface and thereby allows, in each of the n directions of diffraction, the combination of only such plane waves as keep in step with one another along the surface, that is, for which the scalar product $\mathbf{u} \cdot \mathbf{k}_h^{(j)}$ where $\mathbf{u}$, a position vector in the surface, has the same value for all j. As Fig. 3 shows, this is achieved by combining proper fields represented by tiepoints $T^{(1)}$, $T^{(2)} \cdots T^{(2n)}$ which lie on the same normal to the crystal surface as the point A representing the incident wave. Each tiepoint $T^{(j)}$ is assigned a weight $W^{(j)}$ by the above conditions, and these weighted proper tiepoints represent the solution of the problem of adapting the internal wave-field to the conditions of incidence.

Two-ray fields within the crystal

Let us illustrate the general procedure by a discussion of the most important situation, namely when there exists only *one* secondary ray. If either the primary, or the secondary ray were travelling alone through the crystal, their wave vector would have to have length K_0, where $K_0=nk_0=n/\lambda_0$; λ_0 is the wavelength in free space and n the refractive index which for a single ray is given by the Lorentz formula of dispersion according to which the optical density is

$$n^2-1 = \frac{Ne^2/m}{\omega_0^2-\omega^2}, \qquad (1)$$

with N=number of electrons per unit volume, e^2/m the ratio for electrons, ω_0 the (circular) proper frequency of the resonator or dipole, and $\omega=2\pi\nu$ the circular frequency of the optical field. For X-ray frequencies ω_0/ω is usually very small, and the optical density is negative and very small, so that (1) can be replaced by

$$n-1=-(\tfrac{1}{8}\pi^2)\,(Ne^2/m)/\nu^2=\sim 10^{-5} \text{ to } 10^{-6}. \qquad (2)$$

The introduction of K_0 instead of k_0 accounts for the general refraction in the crystal and thereby for the '*deviations from Bragg's Law*' as first observed by Stenström in 1919. The geometrical locus for the foot of the wave vector of the (isolated) primary ray is thus a spherical surface of radius K_0 with the origin as centre, the '*Lorentz Sphere*' of ray 1 and similarly for the (isolated) secondary ray the Lorentz Sphere of radius K_0 about the point **h** of the reciprocal lattice. The two spheres intersect in a circle with centre at the midpoint of the vector **h**, and near this circle lie the tiepoints of the 2-wave fields in which the two rays are coupled by mutual scattering. This coupling leads to a splitting of the two spheres as indicated in Fig. 2:

the tiepoints for the simultaneous 2-ray field are bound to a ring-shaped surface consisting of 4 sheets of hyperbolic cross section. Two of the sheets – the outer ones – belong to the mode where the electric force E is normal to the plane of the two rays, and the other two to those modes for which E lies in that plane. Only the two outer sheets are shown in the figures.

For any plane of the rays 1 and 2 there exist two points of reference: the *Lorentz point Lo* which is at distance K_0 from points O and **h** of the reciprocal lattice and is, therefore, the point where the Lorentz spheres intersect in that plane: and the *Laue point La* where the corresponding spheres of radius k_0 intersect.

The distance $|\overrightarrow{LaLo}| = (k_0 - K_0)\cos\theta = k_0(1-n)\cos\theta$, where θ is the Bragg angle; it is therefore about 10^{-5} to 10^{-6} times smaller than the length of the wave vectors. Drawn to the scale adopted for showing the splitting of the Lorentz spheres the wave vectors should be about a mile long. The spheres or circles about O and **h** can therefore be replaced by their tangents at the Lorentz point, and these form the asymptotic planes or lines of the SurfDisp. Any point T on the surface is a *proper tiepoint*; the field represented by it is a *proper mode* of the optical field; the wave vectors of its two plane waves are fully determined by T, and so is the amplitude ratio of the two waves since each amplitude is inversely proportional to the excess of the length of the wave vector over the free-space value k_0. Thus all we can know about the proper modes of the 2-wave optical field in the crystal is contained in the SurfDisp.

Half-crystal; two-ray internal field adapted to incident field

In making the transition to the half-crystal, we have to distinguish two cases. In the first, the Laue case, the crystal surface lies on one side of both rays, *i.e.* both rays are directed towards the interior of the crystal. In the second case, the Bragg case, the surface lies between the directions of the primary ray and the secondary ray, so that the latter travels in the crystal towards its surface.

In either case the optical field inside the crystal is a superposition of proper modes, such that the amplitude E_0 incident on the crystal surface is taken over, at $z=0$, by the 'primary wave' and that, in the Laue case, the secondary wave begins with zero amplitude at the surface. In the Bragg case we cannot obtain a second boundary condition without introducing a lower surface of the crystal. In the simplest case this is parallel to the entrance surface, so that the crystal is a slab of thickness D. The condition is then that the secondary ray start with zero amplitude at the lower surface [*cf.* Fig. (4*b*)].

Since these conditions have to hold at *any* point of the surface, the waves in direction 1 have to remain in step with the incident wave along the whole surface, and the waves in direction 2 also have to have a common trace along the surface (the upper one in the Laue case, the lower one in the Bragg case). The wave vector of the trace is the resolved part of the spatial wave vector along the surface. The existence of such surface conditions therefore leads to a unique selection of tiepoints $T^{(1)}T^{(4)}$ which may be combined, namely those lying on the same normal to the surface as the point A representing the incident wave (A='Anregungspunkt'). For with this choice, the wave vectors $\overrightarrow{AO}$ $\overrightarrow{T^{(1)}O}$ $\overrightarrow{T^{(2)}O}$ all have the same resolved parts along the surface, and $\overrightarrow{T^{(1)}\mathbf{h}}$, $\overrightarrow{T^{(2)}\mathbf{h}}$ stand in a similar relation; besides the same holds for the other case of polarization for which the hyperbola is not shown in Fig. 3.

The point A in Fig. 3 corresponds to incidence on the reflecting planes under an angle θ smaller than the Bragg angle θ_B since θ_B would be the angle of incidence if A coincided with La. We have

$$|\Delta\theta| = |\theta - \theta_B| = |\overrightarrow{ALa}|/k_0 .$$

As A moves on the Laue circle, the distance $T^{(1)}T^{(2)}$ varies. It has its smallest value, namely the vertex dis-

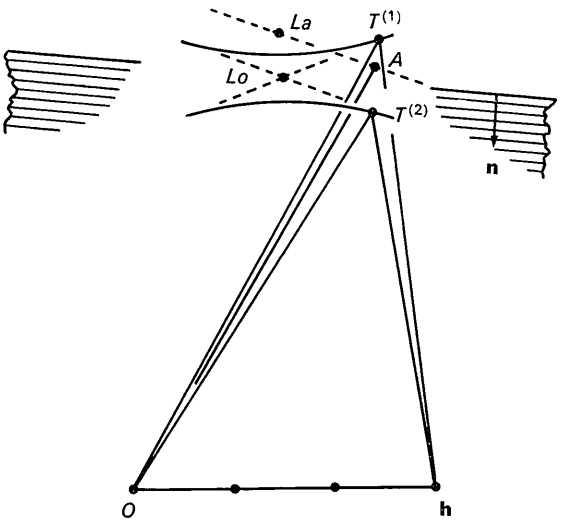

Fig. 3. The SurfDisp for two rays; selection of proper tiepoints for a half-crystal cut off with surface normal **n**.

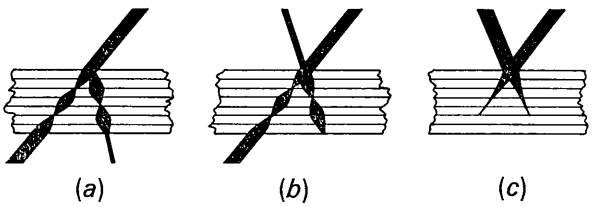

Fig. 4. Wave-fields in the crystal slab, (*a*) Pendellösung in the Laue case, (*b*) Pendellösung in the Bragg case, (*c*) primary extinction and total reflection in the Bragg case.

tance of the hyperbola, when rays 1 and 2 are equally inclined to the reflecting surfaces and A coincides with La. This minimum distance is $K_0|n^2-1|\,|\alpha_h|/\cos\theta$ where α_h, the '*coupling coefficient*' of the two rays, is the Fourier coefficient of order h in the development of the distribution of the polarizability, or scattering power, or electron density, in the crystal cell divided by the total polarizability. This was loosely called the structure factor in the kinematical theory, and we see that the vertex distance is proportional to its first power. The value given above holds when the electric field vector of the waves is normal to the plane of rays 1 and 2 ('*σ polarization*'); in that case the coupling is stronger than for '*π polarization*' when each ray is affected only by the component of the E field of the other ray normal to it. $|\alpha_h|$ is then to be replaced by $|\alpha_h|\cos 2\theta$ where 2θ is the angle between rays 1 and 2.

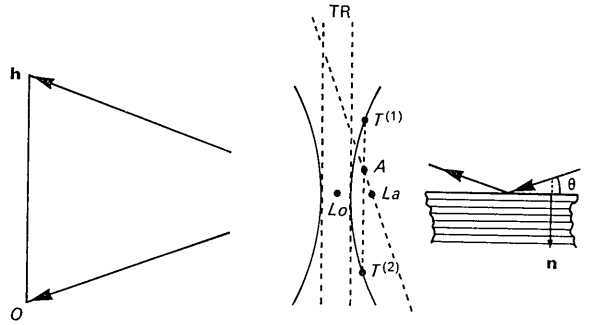

Fig. 5. Bragg case; regions of Pendellösung and of total reflection (TR) for symmetrical Bragg case.

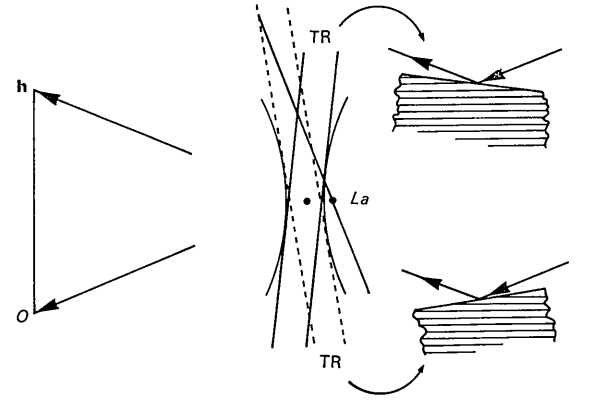

Fig. 6. (*a*) Bragg case; dependence of width of region of total reflection and of deviation from Bragg's law on the inclination of the crystal surface. (*b*) Experimental reflection curves showing this dependence (Bubakova, R., *Czech. J. Phys.* 1962 B12, 776).

The superposition of the proper modes produces two plane waves in direction 1 with vectors $\overrightarrow{T^{(1)}O}$ and $\overrightarrow{T^{(2)}O}$ and two plane waves in direction 2 with wave vectors $\overrightarrow{T^{(1)}\mathbf{h}}$ and $\overrightarrow{T^{(2)}\mathbf{h}}$. The difference between the vectors of each pair is given by the vector $\overrightarrow{T^{(1)}T^{(2)}}$ and is very small compared to the lengths of the wave vectors themselves. Thus in each direction we obtain beats, and, since $\overrightarrow{T^{(1)}T^{(2)}}$ is normal to the crystal surface, these beats are of constant amplitude in planes parallel to the surface. It is by means of these spatial beats that the flux of energy is transferred from direction 1 to direction 2 and *vice versa*, in the same manner that energy oscillates from one to the other of coupled equal or nearly equal pendulums. This type of solution was therefore called originally '*Pendellösung*', and this name has been accepted internationally. The full analogy however, of the procedure of the dynamical theory to that of the general theory of small oscillations in mechanics was not recognized until recently and is published here for the first time. In the hands of N. Kato and others, the fringes obtained by the Pendellösung type of optical field near the exit surface of a slightly wedge-shaped crystal slab have been used for precise determination of structure factors $|F_h|$ – a method understandable in the light of the above remarks on the smallest $T^{(1)}T^{(2)}$ distance which leads to the longest beats.

The Pendellösung type of solution is illustrated in Figs. 4(*a*) and (*b*) for the symmetrical Laue and Bragg cases. The amplitudes of the fields in directions 1 and 2 are indicated by the thickness of the rays. Energy, of course, is conserved; though the direction of the energy flux varies with depth, the integrated flux component parallel to z is the same everywhere – provided of course that absorption is absent.

Whereas in the Laue case the proper tiepoints $T^{(1)}$ and $T^{(2)}$ lie on different branches of the hyperbola, they are on the same branch in the Bragg case. As the point of excitation moves from A in Fig. 5 towards greater glancing angles θ, $T^{(1)}$ and $T^{(2)}$ approach one another and the length of the Pendellösung beats increases, without, however, there occurring any fundamental change in the type of solution until $T^{(1)}$ and $T^{(2)}$ coincide. The first beat then changes, for direction 1, into a linear decrease and for direction 2, into a

linear increase, with increasing depth – *cf.* the behaviour of a pendulum starting its oscillations at time $t=0$ under the influence of a periodic force having as frequency the proper frequency of the pendulum.

As A moves upwards into the region defined by the tangents to the hyperbola parallel to the surface normal n, the intersections with the SurfDisp become complex, indicating that the wave vectors have a real and an imaginary part. The latter produces an exponential increase or decay of the field, as it proceeds in the crystal, of the form $\exp(\pm\kappa z)$ which leads to Darwin's '*primary extinction*', whereas the real part corresponds to a straight displacement of the tiepoint from one tangent point to the other. For points A between the two tangents all of the energy flux of the incident wave must emerge again as the 'reflected' wave 2 from a sufficiently thick non-absorbing crystal, since the primary extinction weakens the field to zero amplitude at the lower boundary of a crystal slab [Fig. 4(*c*)]. The region between the tangents is therefore the '*region of total reflection*' TR. As Fig. 6(*a*) shows, its centre is displaced from the Bragg angle which is the angle of incidence when A coincides with La.

The displacement as well as the width of TR depend on the direction of the crystal surface. Measured in θ, they are largest when incidence on the surface of the crystal is nearly glancing, and the reflected ray leaves the surface at a steep angle [Fig. 6(*a*)]. This effect is well shown in the reflection curves published by Bubakova (1962) [Fig. 6(*b*)]. It was first observed by Bergen Davis & Terrill (1922) and von Nardroff (1924) in their 'rocking curves' and has been used in reverse for converting a wide incident beam into a narrow monochromatized reflected beam [*e.g.* Renninger (1961)].

The reflection curves in Fig. 6(*b*) also show the rounding-off of the curve produced by the variation of the width of the region of total reflection for the σ and the π polarizations and the asymmetry of the curve owing to absorption.

The dynamical theory was formulated in its essential parts in 1917, but it lay dormant for some thirty years with only a few applications. It came into prominence when the art of growing perfect or near-perfect crystals was developed, and when the discussion of electron diffraction and electron microscope pictures demanded some such theory because of the much stronger interaction of matter with electrons than with X-rays. The discovery of the Borrmann effect in 1941 and of its use as a means for the study of dislocations and other imperfections in crystals heightened the interest in the theory. In 1930 the theory was re-cast in a slightly different form by M. v. Laue and it has been used in that form by most workers. The presentation given above reverts more closely to the form and the ideas of the original publication.

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