DYNAMICAL THEORY OF X-RAY PROPAGATION IN DISTORTED CRYSTALS

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The Ewald-Laue theory is exact (it is a solution of Maxwell equations of propagation) ; but its use is limited because it deals with a very special type of experimental situation, seldom encountered in reality, namely :

- an incident plane wave, in vacuum,
- an ideally perfect crystal, extending to infinity in the x and z directions (see fig. 1).

I.- CHARACTERISTIC PARAMETERS

In order to evaluate the degree of ideality (or reality) of these assumptions, it is important to realise what are the orders of magnitude of the characteristic parameters involved in the problem. Bragg diffraction occurs because the crystalline field induces a split of degeneracy in the dispersion relation $E(\vec{K})$. If there were no crystalline field, the dispersion surface would consist of two spheres of radius $K = n/\lambda$, λ being the wave-length in vacuum of the radiation and n the index of refraction, centered on the 2 reciprocal lattice points O and H : the crystalline field introduces a gap in the region where these spheres intersect, of dimension $(K\chi_h)$ (χ_h = h-component of the crystal polarisability).

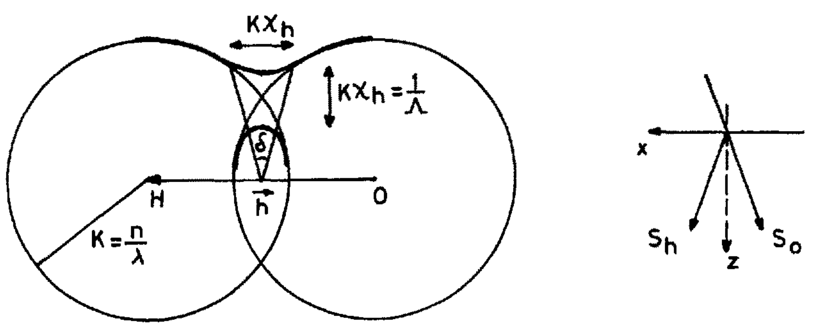

Fig. 1.

$(K\chi_h)$ is the characteristic dimension of the region of reciprocal space involved. A characteristic length Λ can be defined through the basic relation :

$$\Delta\vec{K}.\ \Delta\vec{r} \simeq 1 \qquad (1)$$

$$\Lambda = 1/(K\chi_h) \qquad (2)$$

Since $\chi_h \simeq 10^{-5}$, $\Lambda = 10^{+5}.\lambda \simeq 10^5 \mathring{A}$, for a typical value $\lambda = 1\ \mathring{A}$. It is sometimes convenient to use the angular characteristic parameter :

$$\delta = (K\chi_h)\ /\ K \simeq 10^{-5} \qquad (3)$$

which is the "width" of the Bragg reflection (or rocking curve).

The physical meaning of the characteristic length Λ is the following :

- along the direction of propagation, Λ is the distance over which appreciable changes in the amplitude occur ; it is a "modulation" length ;
- perpendicular to that direction (i.e. on a wave-front), Λ is the minimum size of aperture for the wave to propagate without alteration : it is clear from (1), that collimation by a slit smaller than Λ introduces some additional components in the wave-vector spectrum and therefore modifies the structure of the wave.

II.- LANG TECHNIQUE - PLANE WAVE OR SPHERICAL WAVE

Under these circumstances, a real wave can be considered as a plane wave if, and only if, its spread in wave-vector, specified by δK_x and δK_z, is much smaller than $(K\chi_h)$:

$$\text{Plane wave} \leftrightarrow \delta K_x \text{ and } \delta K_z \ll (K\chi_h) \qquad (4)$$

In the most popular imaging technique (the so-called "Lang topography"), the incident wave is produced by the point like focus of an X-Ray tube at a distance L from the crystal ; the wave thus emitted is monochromatic and spherical. A plane wave can be extracted out of it by introducing a collimating slit (of size Δx) on the beam trajectory, along the entrance surface of the crystal to be investigated ; the wave thus selected is characterized by :

$\delta K_z \simeq 0$ (the incident wave is almost perfectly monochromatic)

$$\delta K_x \simeq K\ (\Delta x/L) \qquad (5)$$

From (4), it is clear that this wave cannot be considered as "plane" unless :

$$K\,\Delta x/L \ll K\chi_h \tag{6}$$

a condition which imposes an upper limit to the width of the slit :

$$\Delta x \ll L\,\chi_h \tag{7}$$

i.e., for a typical value L = 10 cm : $\Delta x \ll 10^4$ Å, a value which is less than the characteristic length Λ (10^5 Å). Such a collimated wave, though "plane" at the entrance surface, does not propagate as a "plane" wave in the crystal ; its wave-vector distribution suffers an inescapable spread in K_x, which can be easily evaluated, from (1) and (7) :

$$\Delta K_x > 1/(L\chi_h) \simeq 10\ (K\chi_h) \tag{8}$$

We therefore come to the conclusion that the ideal plane wave of the Ewald-Laue theory has no reality whatsoever in Lang topography. As a matter of fact it would be more relevant to speak of a "spherical" wave, since the spread in K_x extends on the whole range (and even more) of K_x available in the problem.

III.- RAY THEORY FOR A PERFECT CRYSTAL (wave optics based on ray trajectories)

It is remarkable that the amplitude of this "spherical" wave can be calculated in the conceptual frame of the Ewald-Laue theory... once it has propagated far enough in the crystal (still assumed to be perfect).

Let us expand the wave entering in the crystal as a superposition of plane waves, each being specified by its departure from the exact Bragg angle $\Delta\theta$. Each component excites 2 Bloch waves in the crystal (represented by 2 points P_1 and P_2 on each branch of the dispersion surface), according to the wave matching condition at the entrance surface (fig. 2).

At a given point Q in the crystal, the phase and amplitude of these Bloch waves are known from the Laue theory and the amplitude at Q due to the considered incident wave is just (!) the sum of all of them, with their proper phase relationships.

Such a calculation would be very complicated if, precisely, the spread ΔK_x were not so large ; under these circumstances, the range of phases involved in the summation, which is just the product of ΔK_x by the lateral width Λ of the beam, is larger than 1. Which means that the contribution of any component in the summation can be cancelled out by that of another one, so that the total amplitude finally adds to zero. This is true for all components, except those for which the phase is stationary . Imagine that some value of the deviation parameter $\Delta\theta$, call it $\overline{\Delta\theta}$, makes the phase of the Ewald-Laue theory stationary ; then those components which correspond to a value of $\Delta\theta$ very close to $\overline{\Delta\theta}$ are nearly in phase and add constructively and the only contribution to the amplitude at Q comes from those components which correspond to a value of $\Delta\theta$ close to $\overline{\Delta\theta}$. The calculation which looked, at first sight, very complicated, becomes much simpler.

It has been performed exactly by Kato[1] in the case of a "perfect" spherical wave, i.e. assuming that the entrance slit is restricted to a single point $\Delta x=0$. The result (see fig. 2) is that only 2 wave-fields contribute to the amplitude at a given point Q, those which propagate along the OQ direction.

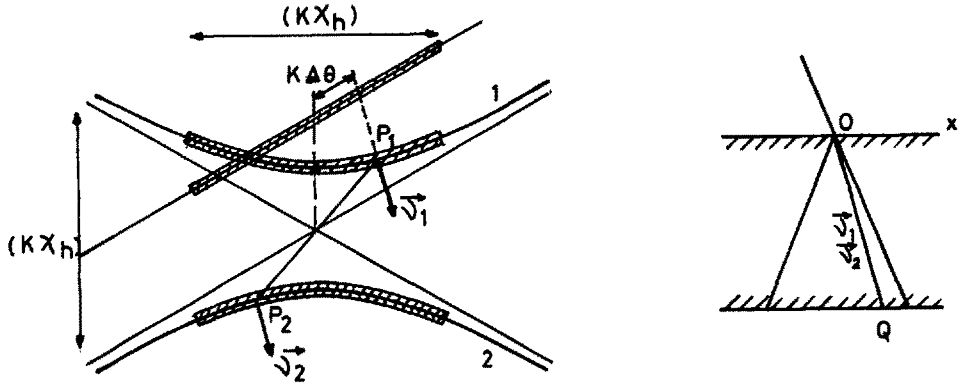

Fig. 2

Up to now, we have assumed a perfect crystal. As a matter of fact, Kato's "ray" theory can be easily extended in order to include the case of a crystal containing some kind of planar defect, a stacking fault, for instance (fig. 3).

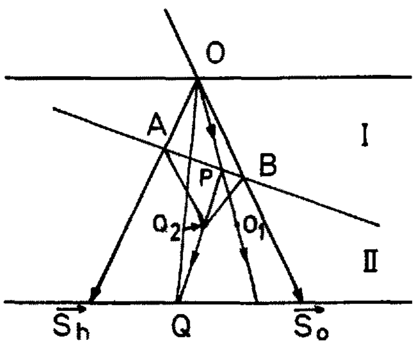

Fig. 3

Let P be a point on the fault line AB ; the energy arriving at P travels along OP in part I of the crystal. Viewed from part II this same point P can be reached either along O_1P parallel to OP or O_2P, O_2 being such that O_2B//OA and O_2A//OB. This means that at a given point Q on the exit surface, the amplitude is the sum of that corresponding to the direct trajectory OQ and that corresponding to the indirect trajectory OPQ. The exact calculation of the amplitude distribution on the exit surface of a crystal containing a stacking fault has been performed by several authors (2) and is in good agreement with the experimental observations.

IV.- DISTORTED CRYSTALS - THE EIKONAL THEORY

The classical theory is not restricted to planar defects and can be extended in order to account for the contrast of other types of defects. Nevertheless, if we try to apply the technique of the previous section without any change (i.e. expand the incident wave in plane components and then make an argument about the real trajectory being that for which constructive interference between neighbouring paths occurs), then we come across a difficulty which is that the Ewald-Laue theory cannot be applied to the calculation of the phase along a given path, since this theory assumes a perfect and infinite crystal, which is no more the case. Generally speaking, it is well known that plane wave (Fourier) analysis is well adapted only to those systems which exhibit invariant translational properties (in space, or in time, for instance). Such an invariance exists for a perfect and infinite crystal and this is the reason why the (plane) Bloch-waves of the Ewald-Laue theory are the "normal" modes of the problem in that case ; but when the presence of a defect breaks this translational invariance, the plane wave analysis looses its relevance.

The situation here is very similar to that encountered in ordinary optics (3) . When the medium in which the light propagates is characterized by a varying index of refraction, the plane waves

$$A \exp 2\pi i \vec{k}.\vec{r} \quad (\vec{k} \text{ fixed and } A = \text{Cste}) \qquad (10)$$

are no more the "normal modes" of the problem and have to be replaced by "modified plane waves" of the type :

$$A(\vec{r}) \exp 2\pi i\ S(\vec{r}) \qquad (11)$$

where $A(\vec{r})$ is a slowly varying function of $\vec{r}$ and $S(\vec{r})$ is called the Eikonal function. A local wave-vector can then be defined through the relation

$$\vec{k}(\vec{r}) = \overrightarrow{\text{grad}}\ S(\vec{r}) \qquad (12)$$

Applying the same type of argument to the propagation in a non-perfect crystalline medium, we replace the Bloch-waves by "modified Bloch-waves" (4) :

$$A_o(\vec{r}) \exp 2\pi i\ S_o(\vec{r}) + A_h(\vec{r}) \exp 2\pi i\ S_h(\vec{r}) \qquad (13)$$

and define two local wave-vectors :

$$\vec{K}_o(\vec{r}) = \overrightarrow{\text{grad}}\ S_o(\vec{r}) \text{ and } \vec{K}_h(\vec{r}) = \overrightarrow{\text{grad}}\ S_h(\vec{r}) \qquad (14)$$

such that

$$\vec{K}_h(\vec{r}) = \vec{K}_o(\vec{r}) + \vec{h}(\vec{r}) \qquad (15)$$

$\vec{h}(\vec{r})$ is the so-called local reciprocal lattice vector ; it depends on the atomic displacement $\vec{u}(\vec{r})$ due to the defects and is related to the reciprocal lattice vector of the perfect crystal $\vec{h}$:

$$\vec{h}(\vec{r}) = \vec{h} - \vec{\nabla}\left(\vec{h}.\vec{u}(\vec{r})\right) \qquad (16)$$

With these definitions, it is then possible to calculate the phase along a given path and determine the real trajectory by a condition of stationarity along the same lines as those developped in the previous section. As a matter of fact, this procedure is just an extension of Fermat's principle to a crystalline medium. The main result is that the two wave-fields which would propagate along the same direction in the perfect crystal (see fig.2) now separate ; since their separation depends on $\vec{u}(\vec{r})$, and thus on the local defects, it is clear that the changes in amplitude distribution on the exit surface give some information on the defects themselves ; this is commented by C. Malgrange in this issue (5).

V.- LIMITS OF VALIDITY OF THE EIKONAL THEORY - GENERAL TREATMENT

V.1.- In order for the Eikonal approximation (13), and its consequence (15), to be valid, it is necessary that :

$$\begin{pmatrix} \text{the variation } \Delta_\Lambda \vec{K}_h \text{ of } \vec{K}_h \text{ over} \\ \text{a segment } \Lambda \text{ of ray trajectory} \end{pmatrix} << \begin{pmatrix} \text{characteristic length} \\ \text{in } \vec{K} \text{ space: } (K\chi_h) \end{pmatrix} \qquad (17)$$

The left hand side of this inequality involves the derivative of $\vec{K}_h$; since $\vec{K}_h$ itself involves (see (16)) the first derivative of $\vec{h}.\vec{u}(\vec{r})$, it is clear that the left side of (17) contains the second derivative of $\vec{h}.\vec{u}(\vec{r})$. Exact calculation (6) leads to :

$$\frac{\partial^2[\vec{h}.\vec{u}(\vec{r})]}{\partial s_o \, \partial s_h} \cdot \Lambda << K\chi_h \qquad (18)$$

($\vec{s}_o$, $\vec{s}_h$: unit vectors along the incident and reflected direction).

This is usually written in the form :

$$f = \frac{\partial^2[\vec{h}.\vec{u}(\vec{r})]}{\partial s_o \, \delta s_h} \cdot \Lambda^2 << 1 \qquad (19)$$

Condition (17) can also be expressed in an alternative and equivalent manner as :

$$|\Delta_\Lambda \vec{K}_h/K| << \chi_h \qquad (20)$$

Since $|\Delta_\Lambda \vec{K}_h/K|$ is just the disorientation of the reflecting planes over a distance Λ , the conclusion is that the Eikonal approximation holds only as long as this disorientation is less than δ , the width of the rocking curve.

V.2.- When condition (17) is not fullfilled, neither the crystal wave nor the normal modes have something to do with plane waves. Rather than find out, for each type of deformation, the new normal modes along which the amplitude $\psi(\vec{r})$

should be expanded, it is more rational to solve Maxwell equations of propagation directly in the non-perfect crystal. Such a theory has been developped by Takagi [(7)] and is, at the present time, the basic and general theory of X-Ray propagation in crystals.

In this treatment, nothing particular is assumed concerning the structure of the crystal wave, except that it has an "o" and an "h"-component. $\psi(\vec{r})$ is written as :

$$\psi(\vec{r}) = \sum_h \psi_h(\vec{r}) \exp - i\vec{k}_h.\vec{r}, \left(h = o,\ h\right) \qquad (21)$$

$\vec{k}_h.\vec{r}$ represents a fast oscillation of constant periodicity λ , (wavelength in the perfect crystal) ; superimposed on that fast oscillation, $\psi(\vec{r})$ exhibits a modulation of its amplitude, extending over a range of order Λ and depending on the local state of deformation. Introducing (21) in Maxwell equations, Takagi obtains a set of partial differential equations of second order and hyperbolic type, which can be solved (in principle) by the Green function technique.

V.3.- The Green function (or propagator) G(O,Q) represents the effect of a unique point source at a further point Q ; since the total amplitude at Q, is the superposition of all the wavelets emitted by the different field sources (Huyghens's principle), it can be calculated by performing the convolution product of the source distribution by the Green function.

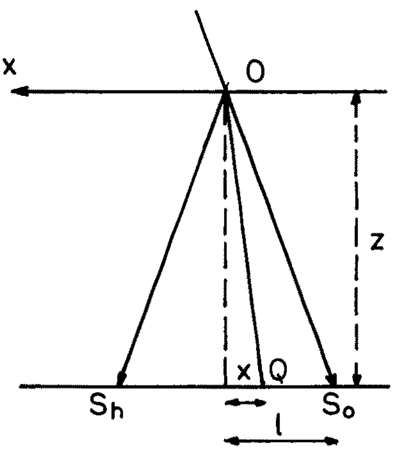

Fig. 4

It is enlightening to see how the Ewald-Laue theory comes out of this general treatment as a special case (perfect crystal and plane incident wave). Let $\lambda_x = 2\pi/K_x$ be the periodicity of a plane incident wave along the entrance surface Ox ; in the present case, the source distribution consists of all the points of this entrance surface which are "lightened" by the incident wave (each of them acts like a point source). On the other hand, Takagi has shown that the Green function corresponding to the propagation in a perfect crystal is :

$$G(O,Q) = J_o\left[\frac{z}{\Lambda}\sqrt{1-(x^2/l^2)}\right] \quad \text{(see fig.4)}.$$

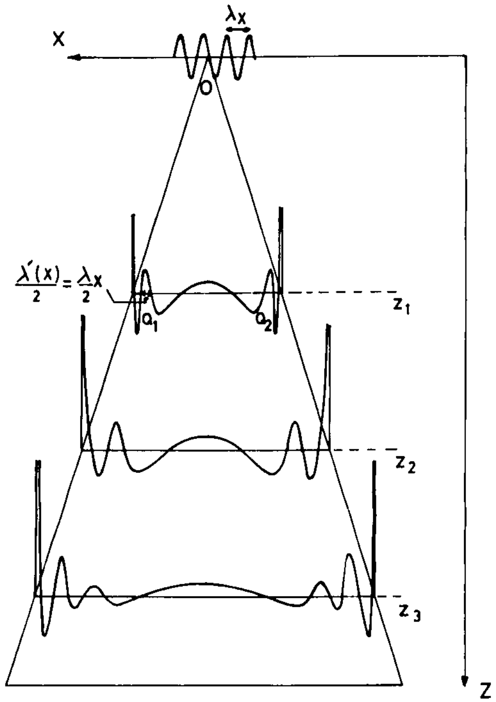

Fig. 5

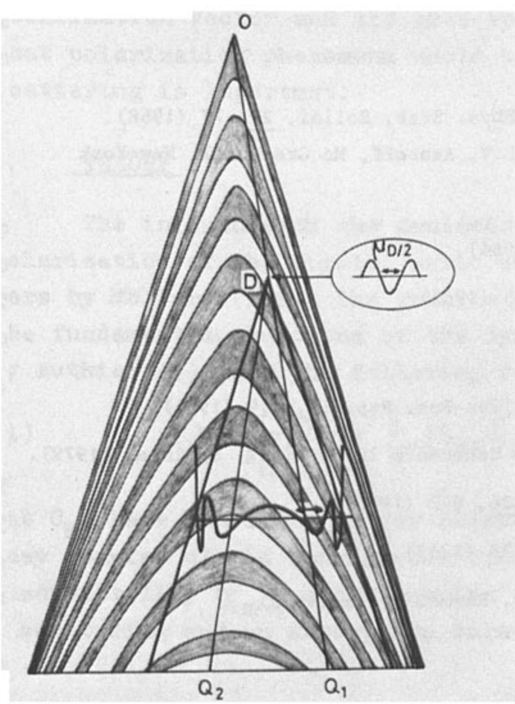

Fig. 6

At a given depth z in the crystal, this Green function exhibits oscillations of variable "wave-length" $\lambda'(x)$ (fig. 5). Convolution of this Green function with the periodic source distribution on the entrance surface, will amount to zero except at those points Q_1 and Q_2 where $\lambda_x = \lambda'(x)$. These points correspond to the two wave-fields of the Ewald-Laue theory.

Note that the Green function for a perfect crystal is just the amplitude distribution induced in the crystal by a "perfect" spherical wave, already calculated by Kato (see § III).

V.4.- For a non perfect crystal, the determination of the Green function is not so easy as in the case of a perfect crystal ; as a matter of fact, it is a problem still to be solved. Nevertheless, it can be shown(6) (on the basis of the mathematical theory of distributions(8) that it is not necessary to calculate the proper Green function for each case of imperfection and that the Green function J_o of the perfect case is sufficient, at least for a first order approximation. It can be shown, that the crystal wave can be calculated by assuming a perfect crystal and replacing the deformations by an extra distribution of fictive sources which are to account for the local deformations(6).

As an example of this technique, let us examine the effect on a section topograph (i.e. assuming a "perfect" spherical wave at the entrance surface) of the region which lies in the immediate vicinity (by this we mean at a distance <Λ) of the core D of a dislocation line. It has been shown that the strength of the equivalent "fictive" sources depends both on the local deformation and on the amplitude which would exist at the considered point if the crystal were perfect. It is clear on fig. 6 that the effect of the dislocation will be more or less sensible according to whether D lies inside a dark fringe or right in the middle of a bright one. In the present case, it is even possible to predict along which direction the disturbance in amplitude will be the most sensible. The argument is of the same type as in section V.3 : the distribution of fictive sources around D can be characterized by a certain periodicity, call it μ_D . Convoluting this distribution by the Green function J_o will give a null result except along those lines (originating at D) where μ_D matches the periodicity of J_o, i.e. along two lines DQ_1 and DQ_2 . In other words, we expect that two wave-fields will stem out of the highly distorted region around D.

Since it would seem that an "extra" wave-field has been created by the distorted region, this phenomenon is generally referred to as that of the "creation of new wave-fields" ; it is of great importance in the interpretation of the contrast of isolated defects such as dislocations.

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