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DYNAMICAL THEORY OF MOIRE FRINGE PATTERNS

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SUMMARY

The dynamical theory of moiré patterns is developed on the basis of the two-beam approximation. The method used is the column approximation introduced by Howie and Whelan.

First, the geometry of the moiré patterns is discussed. Then the intensity profile of the fringes is calculated. Assuming that the anomalous absorption is negligible, the effect of a step on the surface and of tilting the crystal is examined. During tilting the fringes will be displaced and the contrast between dark and bright fringes will die away, possibly in an observable fluctuating way. The asymptotic expressions of the results yield the formulae valid for the kinematical approximation, which is also discussed. It is suggested that under favourab conditions the crystal thicknesses may in principle be determined from the fluctuations of the contrast during the tilting of the crystal.

The anomalous absorption effects have then be considered; they do not influenc the geometry of the pattern, but introduce corrections for the fringe profile if the considered crystal is of intermediate thickness. For thick crystals the anomalous absorption effects become predominant.

It is shown that the results of the column approximation can be obtained and generalized in a simple way. The transmitted, respectively diffracted, wave is described as arising by the interference of two transmitted, respectively diffracted, waves with slightly different wave vectors. The column approximation reveals itself as a very good approximation for sandwiches of crystals of the same material.

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§ 1. INTRODUCTION

It is well known that if two parts of a crystal foil, whose interface is parallel to the surface, are rotated over a small angle or have parallel lattices with slightly different parameters, dark and bright fringes, known as moiré fringes, show up in the electron microscope.

The kinematical treatment of this effect is due to O. Rang (1960), whilst Hashimoto et al (1961) have recently developed a dynamical theory using the Schrödinger equation. A review of the literature on the subject is given in the quoted papers.

In the present paper we propose, first, to derive the intensities of the moiré pattern fringes from the system of equations introduced by Howie and Whelan (1960a, 1961) expressing the column approximation. The latter consists in identifying the intensity at a given point of the back surface, with the intensity of the wave transmitted and diffracted by a narrow column normal to the foil surface. The final result will be obtained by means of calculations involving only elementary mathematical methods.

It will be shown, next, that this result can also be deduced by a simple reasoning, without any calculations. Moreover, the formulae obtained from the column approximations are generalized and can be applied even when the latter breaks down. It will be seen, however, that in most cases the column approximation is a very good one.

The method described here represents a simplification of the dynamical theory of moiré pattern. Furthermore, we shall examine some aspects of the moiré pattern which have not been discussed by Hashimoto et al (loc.cit.), in particular the anomalous absorption effects. The generalisation for more than two superposed crystals or more than one strongly diffracted beam does not introduce major difficulties. This will be examined in a paper to be published elsewhere.

The amplitudes $\mathcal{T}$ and $\mathcal{S}$ of transmitted and reflected waves are solutions of the system

$$\begin{cases} \dfrac{d\mathcal{T}}{dz} = i\,\dfrac{\pi}{t}\exp i2\pi\,\underline{g}\cdot\underline{r}\;\mathcal{S} \\[2ex] \dfrac{d\mathcal{S}}{dz} = i\,\dfrac{\pi}{t}\exp -i2\pi\,\underline{g}\cdot\underline{r}\;\mathcal{T} \end{cases}$$

where

$1/t = 1/t_0 + i\,1/\tau_0$: extinction distance;

t_0 : anomalous absorption distance;

τ_0 : diffraction vector;

$\underline{g} = \underline{n} + \underline{s}$;

$\underline{n}$: reciprocal lattice vector corresponding to the considered reflection (only vectors $\underline{n}$ parallel to the surface will be considered); ($n = 1/d_n$ if d_n is the interplanar distance);

$\underline{s}$: vector describing the deviation of the reciprocal lattice point $\underline{n}$ from the reflection sphere;

s : component of $\underline{s}$ normal to the surface of the crystal, in the direction of the diffracted wave;

$\underline{r}$: position vector of the considered unit cell in the column, (we shall take the origin ~~of~~ in the interface);

z : component of r normal to the surface of the crystal;

$\underline{r} = \underline{r}_0 + z\,\underline{e}_z$ ($\underline{r}_0$ lies in the interface); $\underline{e}_z$ unit vector normal to interface).

The sandwich crystal producing the moiré pattern is considered as being derived from a single crystal by displacing the top half in a non-uniform manner with respect to the bottom half, the interface being parallel to the surface.

In this process the upper parts of the columns normal to the surface are slightly displaced with respect to the corresponding lower parts. This small displacement is described by a vector $\underline{u}(\underline{r}_0)$. The position vector $\underline{r}_0$ determines the intersection of the considered column with the interface.

Whilst $\mathcal{F}$ and φ, in the part of the column lying in the first section of the crystal, are solutions of the equations (1a)and (1b), where

$\exp i 2\pi \underline{g}\cdot\underline{r} = \exp i 2\pi(\underline{n}+\underline{s})\cdot\underline{r} = \exp i 2\pi s z$,

the same amplitudes in the second part will be solutions of the same system in which $\bar{g}.\bar{r}$ has to be replaced by $\bar{g}.(\bar{r} + \bar{u})$. The factor $\exp i 2\pi \underline{g}\cdot\underline{r}$ of the second member is changed into

$$\exp i 2\pi \underline{g}\cdot(\underline{r}+\underline{u}) = \exp i 2\pi(\underline{n}+\underline{s})\cdot(\underline{r}+\underline{u}) = \exp i\alpha \exp i 2\pi s z \qquad (2)$$

where $\alpha = 2\pi \underline{n}\cdot\underline{u}$

and $\underline{s}\cdot\underline{u}$ has been neglected.

Summarizing, we can state that $\mathcal{T}$ and $\mathcal{S}$ are solutions of the system

$$\begin{cases} \dfrac{d\mathcal{T}}{dz} = i\dfrac{\pi}{t}\left[1-(1-\exp i\alpha)\,\varepsilon(z-z_1)\right]\exp i2\pi sz\ \mathcal{S} & (3a) \\ \dfrac{d\mathcal{S}}{dz} = i\dfrac{\pi}{t}\left[1-(1-\exp -i\alpha)\,\varepsilon(z-z_1)\right]\exp -i2\pi sz\ \mathcal{T} & (3b) \end{cases}$$

where

$$\varepsilon(z-z_1) = \begin{cases} 0 & \text{for} \quad 0 \leq z < z_1 \\ 1 & \text{for} \quad z_1 < z \leq z_2 \end{cases}$$

z_1 and z_2 are the thicknesses of the two parts of the crystal ($z = 0$ corresponds to the entrance surface). We suppose here that the change in s and t from one part of the crystal to the other is negligible. We shall discuss this matter again in § 7.

§ 2. THE GEOMETRY OF THE MOIRE PATTERNS.

Suppose it is possible to define a vector $\bar{k}$, lying in the interface, by the relation $\underline{n}\cdot\underline{u}(\underline{r}_0) = \underline{k}\cdot\underline{r}_0$ (4)

One has $\underline{r}_0 = x\,\underline{e}_x + y\,\underline{e}_y$ if $\underline{e}_x$ and $\underline{e}_y$ are two unit vectors defining a reference system in the interface.

An adequate choice of the unit vectors consists in taking $\underline{e}_x$ parallel to, and $\underline{e}_y$ normal to, $\underline{k}$.

For $k = 1/\Lambda$ one has $\alpha = 2\pi x/\Lambda$ (5a,b)

All points of the surface with the same x coordinates, i.e. lying on the same straight line normal to $\bar{k}$, will have bright and dark field images of the same intensity, which have to be calculated by integrating the system (3) depending on $\exp i2\pi x/\Lambda$. This intensity depends on x. Moreover, the periodicity of $\exp i2\pi x/\Lambda$ induces the fringe character, with period Λ, of the bright and dark field interference image of the crystal sandwich. The image is observed as a moiré pattern : bright and dark fringes normal to $\bar{k}$, the spacing between two successive bright (dark) fringes being $1/k = \Lambda$.

We shall now derive an expression for $\underline{k}$. We suppose that the axes of the unit cell of the first part of the crystal are $\bar{a}_1$, $\bar{a}_2$, $\bar{a}_3$, ($\bar{a}_1$ and $\bar{a}_2$ in the interface, $\bar{a}_3$ normal to the interface), and that the axes of the second part are $\bar{a}_1 + \underline{\Delta}\bar{a}_1$, $\bar{a}_2 + \underline{\Delta}\bar{a}_2$, $\bar{a}_3$ with $\underline{\Delta a_1}/\underline{a_1}$, $\underline{\Delta a_2}/\underline{a_2} \ll 1$.

We consider a column intersecting the interface at the point $\underline{r}_0 = p\,\underline{a}_1 + q\,\underline{a}_2$. One has then for $\underline{u}$: $\underline{u} = p\,\underline{\Delta a}_1 + q\,\underline{\Delta a}_2$ and for $\underline{u}\cdot\underline{n}$:

$$\underline{u}\cdot\underline{n} = p(\underline{n}\cdot\underline{\Delta a}_1) + q(\underline{n}\cdot\underline{\Delta a}_2) = \underline{r}_0\cdot\left[(\underline{n}\cdot\underline{\Delta a}_1)\underline{A}_1 + (\underline{n}\cdot\underline{\Delta a}_2)\underline{A}_2\right]$$

where $\underline{A}_1$, $\underline{A}_2$ and $\underline{A}_3$ are the reciprocal lattice vectors corresponding to the lattice vectors $\underline{a}_1$, $\underline{a}_2$ and $\underline{a}_3$. It is now clear from the definition (4) that

$$\underline{k} = (\underline{n}\cdot\underline{\Delta a}_1)\underline{A}_1 + (\underline{n}\cdot\underline{\Delta a}_2)\underline{A}_2 \tag{6}$$

It is possible to give a simple geometric interpretation for the vector $\underline{k}$. The changes $\underline{\Delta A}_1$ and $\underline{\Delta A}_2$ of the reciprocal lattice vectors $\underline{A}_1$ and $\underline{A}_2$ are to be found from the equations

$$\underline{a}_i\cdot\underline{A}_j = \delta_{ij} \tag{7a}$$

which relate the unit cell of the lattice to the unit cell of the reciprocal lattice.

It follows then :

$$\underline{A}_j\cdot\underline{\Delta a}_i = -(\underline{a}_i + \underline{\Delta a}_i)\cdot\underline{\Delta A}_j\,, \text{ or} \tag{7b}$$

$$\underline{A}_j\cdot\underline{\Delta a}_i = -\underline{a}_i\cdot\underline{\Delta A}_j\,, \text{ if } \underline{\Delta a}_i\cdot\underline{\Delta A}_j \text{ is neglected.} \tag{7c}$$

The vector $\underline{n} = h_1\underline{A}_1 + h_2\underline{A}_2$ of the reciprocal lattice is changed into $\underline{n}' = \underline{n} + \underline{\Delta n}$, with $\underline{\Delta n} = h_1\,\underline{\Delta A}_1 + h_2\,\underline{\Delta A}_2$

It is easy to show that

$$\underline{\Delta n} = -\underline{k} \tag{8}$$

We have

$$\underline{k}\cdot\underline{a}_i = \underline{n}\cdot\underline{\Delta a}_i = \left(\sum_{j=1}^{2} h_j\underline{A}_j\right)\cdot\underline{\Delta a}_i = -\sum_{j=1}^{2} h_j\,\underline{a}_i\cdot\underline{\Delta A}_j = -\underline{a}_i\cdot\left(\sum_{j=1}^{2} h_j\,\underline{\Delta A}_j\right) = -\underline{a}_i\cdot\underline{\Delta n} \quad (i=1,2)$$

from which follows immediately the relation to be proved.

As a consequence, a two beam moiré pattern for a given reflection will consist of alternative bright and dark straight fringes normal to $\underline{\Delta n}$, the vector difference of the considered reciprocal lattice vectors in both components of the crystal sandwich. The distance between two successive bright (or dark) fringes will be given by the inverse of the length Δn of the vector $\underline{\Delta n}$.

Let us consider, first, a simple rotation moiré pattern. This arises when the two components of the crystal sandwich are rotated over an angle with respect to each other around an axis normal to the interface.

In this case, the fringes form with $\underline{n}$ an angle $\varepsilon/2$ in general negligibly small, whilst for Λ, the interspacing of the fringes, one has

$$\Lambda = \frac{1}{\Delta n} = \frac{1}{2\sin\frac{\varepsilon}{2} n}, \text{ or } \Lambda \simeq \frac{d_n}{\varepsilon} \qquad (9\,a,b)$$

(d_n : interplanar distance of the lattice planes defined by $\underline{n}$).

Let us consider next what we propose to call a "compression" moiré pattern. The latter occurs if $\underline{\Delta n} = \varepsilon \underline{n}$, which can be interpreted as a compression of the lattice planes normal to $\underline{n}$? More explicitely :

$$\varepsilon = \frac{d_n - d_{n'}}{d_{n'}} \qquad (10)$$

The fringes are normal to the reciprocal lattice vector $\underline{n}$, whilst their interspacing is still given by the same simple relation (9b), where for ε the adequate expression (10) has to be substituted.

The general case can be thought as a combination of the two simple cases described. As shown in fig. 1, $\underline{\Delta n}$ can be decomposed into two components $(\underline{\Delta n})_r$ and $(\underline{\Delta n})_c$, the first expressing a rotation over a small angle ε_r, the second being the result of a slight compression ε_c. If the ε's are small, so that higher powers of ε can be neglected, one finds for β, the angle formed by the fringes and the diffraction vector :

$$tg\,\beta \simeq \frac{(\Delta n)_c}{(\Delta n)_r} = \frac{\varepsilon_c}{\varepsilon_r} \qquad (11a)$$

For the interfringe spacing one has then

$$\frac{1}{\Lambda} \simeq \sqrt{\frac{1}{\Lambda_r^2} + \frac{1}{\Lambda_c^2}}, \text{ since } (\Delta n)^2 \simeq [(\Delta n)_r]^2 + [(\Delta n)_c]^2 \qquad (11b)$$

The column approximation used in the next paragraph for the calculations of the intensities fails to be a good approximation if $z \cdot 2\theta$ (z = total thickness θ Bragg angle) is not negligibly small compared to Λ. Since Λ is proportional to the inverse of Δn, this will occur for larger values of ε. The neglecting of $\underline{\Delta a_i} \cdot \underline{\Delta A_j}$ in (7c) is then questionable and the proof that $\underline{k}$ and $-\underline{\Delta n}$ are identical breaks down. However, from the arguments developed in § 7, it will follow that the vector governing the geometry of the moiré pattern is, also for large values of ε, the reciprocal lattices vector difference $\underline{\Delta n}$.

This matt r will again be discussed in § 7.

To close this paragraph, we remark that it is possible to imagine a situation more general than the one considered above, giving rise to a moiré pattern. If to the displacement $\underline{u}$ is added an arbitrary other displacement $\underline{v}$, such that $\underline{v}.\underline{n}=0$, a moiré fringe system will be produced. However, for any other reciprocal lattice vector $\underline{m}$ not parallel to $\underline{n}$, $\underline{m}.\underline{v}$ may be different from zero and a moiré would no longer be observed. In order for a moiré pattern with straight fringes to occur, $\underline{u}$ should of course be such that it is possible to define a vector $\underline{k}$ by the relation (4).

§ 3 THE INTENSITY PROFILE OF THE MOIRE FRINGES

The substitution $\mathcal{T} = \exp i\pi s z\, T$ and $\mathcal{S} = \exp -i\pi s z\, S$ which does not alter intensities $|\mathcal{T}|^2$ nor $|\mathcal{S}|^2$, and the boundary conditions $\mathcal{T}(0)=1$, $\mathcal{S}(0)=0$, changes sytem (3) into the new system

$$\begin{cases} \dfrac{dT}{dz} + i\pi s\, T = i\dfrac{\pi}{t}\left[1-(1-\exp i\alpha)\,\varepsilon(z-z_1)\right] S & (12a) \\[2ex] \dfrac{dS}{dz} - i\pi s\, S = i\dfrac{\pi}{t}\left[1-(1-\exp -i\alpha)\,\varepsilon(z-z_1)\right] T & (12b) \end{cases}$$

From (12a) one obtains

$$(D-i\pi s)(D+i\pi s)\,T = i\frac{\pi}{t}\left[1-(1-\exp i\alpha)\,\varepsilon(z-z_1)\right](D-i\pi s)\,S - i\frac{\pi}{t}(1-\exp i\alpha)\,\delta(z-z_1)\,S$$

using the relation $\dfrac{d}{dz}\varepsilon(z) = \delta(z)$, where δ : Dirac delta function.
Applying now (12b), yields the system

$$\begin{cases} \dfrac{d^2T}{dz^2} + (\pi\sigma)^2 T = -i\dfrac{\pi}{t}(1-\exp i\alpha)\,\delta(z-z_1)\,S & (13a) \\[2ex] \dfrac{d^2S}{dz^2} + (\pi\sigma)^2 S = -i\dfrac{\pi}{t}(1-\exp -i\alpha)\,\delta(z-z_1)\,T & (13b) \\[2ex] \sigma = \sqrt{1+(st)^2}/t\,, & (13c) \end{cases}$$

the second equation is derived in the same way as the first.

For $z<z_1$, the amplitudes T_0 and S_0 are solutions of the homogeneous system to which system (13) reduces for $z<z_1$.
One has then

$$\begin{cases} T_0 = A\cos\pi\sigma z + B\sin\pi\sigma z & (14a) \\ S_0 = C\cos\pi\sigma z + D\sin\pi\sigma z & (14b) \end{cases}$$

The constants A, B, C and D are obtained by introducing (14) into the system (12), where $\varepsilon(z-z_1)=0$ for $z<z_1$ and by expressing the boundary conditions $T_0(0)=1$, $S_0(0)=0$.
After performing these calculations, one obtains

$$\begin{cases} T_0 = \cos\pi\sigma z - i\,\frac{s}{\sigma}\sin\pi\sigma z & (15a) \\ S_0 = \dfrac{i\sin\pi\sigma z}{\sigma t} & (15b) \end{cases}$$

which yield the well-known expressions for the "two-beam" approximation of the amplitude of the wave transmitted and diffracted by a perfect crystal (Bethe, 1928; Mc Gillary, 1940; Heidenreich, 1949; Kato, 1952,1953; Whelan and Hirsch, 1957; Howie and Whelan, 1961).

The general solution of an equation of the type

$$\frac{d^2y}{dx^2} + k^2 y = f(x) \qquad (16)$$

can be written as

$$y = y_0 + \frac{1}{k}\int^{x} \sin k(x-t)\, f(t)\, dt \qquad (17)$$

where y_0 is the general solution of the homogeneous differential equation obtained from (16).

If this procedure is applied to (13a) one obtains

$$T = T_0 - i\frac{1}{\sigma t}(1-\exp i\alpha)\int_0^z \sin\pi\sigma(z-t)\,\delta(t-z_1)\,S(t)\,dt\,,$$

where for T_0 the expression (15a) has to be taken, since the integral vanishes for $z<z_1$.
Finally

$$T = T_0 - i\frac{1}{\sigma t}(1-\exp i\alpha)\sin\pi\sigma(z-z_1)\,S_0(z_1) \quad (z\geq z_1) \qquad (18a)$$

At the surface $z=z_1+z_2$, taking (15) and (5b) into account, the formula (18c) becomes

$$T = \cos\pi\sigma(z_1+z_2) - i\frac{s}{\sigma}\sin\pi\sigma(z_1+z_2) + \frac{1}{(\sigma t)^2}\left(1-\exp i2\pi\frac{x}{\Lambda}\right)\sin\pi\sigma z_1\sin\pi\sigma z_2 \qquad (18b)$$

In a similar way we have

$$S = S_0 - i\frac{1}{\sigma t}(1-\exp -i\alpha)\int_0^z \sin\pi\sigma(z-t)\,\delta(t-z_1)\,T(t)\,dt$$

or at the surface

$$S = S_0 - i\frac{1}{\sigma t}(1-\exp -i\alpha)\sin\pi\sigma(z-z_1)\,T_0(z_1) \qquad (19a)$$

$$S = \frac{i}{\sigma t}\left\{\sin\pi\sigma(z_1+z_2) - \left(1-\exp -i2\pi\frac{x}{\Lambda}\right)\sin\pi\sigma z_2\left(\cos\pi\sigma z_1 - i\frac{s}{\sigma}\sin\pi\sigma z_1\right)\right. \qquad (19b)$$

The same result may also easily be obtained by noticing that for $z > z_1$, the amplitudes T_1 and S_1 satisfy the same homogeneous system of second-order differential equations as T_0 and S_0.

As a consequence, one gets

$$\begin{cases} T_1 = A' \cos \pi\sigma z + B' \sin \pi\sigma z \\ S_1 = C' \cos \pi\sigma z + D' \sin \pi\sigma z \end{cases}$$

The constants A', B', C', D' are now determined by substituting these expressions into the system (12) (with $\varepsilon = 1$) and by expressing that

$$T_1(z_1) = T_0(z_1) \quad , \quad S_1(z_1) = S_0(z_1)$$

This method avoids the use of the δ-function and the Green function, which have only been introduced for reasons of mathematical elegance.

§ 4. DISCUSSION OF THE FRINGE PROFILE

For an adequate discussion of the fringe profile, it is usefull to derive from the expressions (18) for T, the intensity $I_\pi(x) = |T|^2$ in a form showing directly the properties of the profile.

Formula (18) can be rewritten as follows :

$$T = a + b \exp i 2\pi \frac{x}{\Lambda} \tag{20a}$$

with

$$a = \cos \pi\sigma(z_1 + z_2) + \frac{\sin \pi\sigma z_1 \sin \pi\sigma z_2}{(\sigma t)^2} - i \frac{\delta}{\sigma} \sin \pi\sigma(z_1 + z_2) \tag{20b}$$

or, after calculations,

$$a = T_1 T_2 \tag{20c}$$

where

$$T_i = \cos \pi\sigma z_i - i \frac{\delta}{\sigma} \sin \pi\sigma z_i \tag{20d}$$

$\exp i \pi\sigma z_i \, T_i$ is the amplitude of the wave transmitted by a crystal with thickness z_i,

and

$$b = \frac{i \sin \pi\sigma z_1}{\sigma t} \cdot \frac{i \sin \pi\sigma z_2}{\sigma t} = S_1 S_2 \tag{20e}$$

where $S_i = \frac{i \sin \pi\sigma z_i}{\sigma t} \exp - i \pi z_i \, S_i$ is the amplitude of the wave diffracted by a crystal with thickness z_i.

If the anomalous absorption is neglected, one has

$$T_i = \sqrt{I_\pi^{(i)}} \exp i \Phi_i \tag{21a}$$

$$I_\pi^{(i)} = 1 - \frac{\sin^2 \pi\sigma z_i}{(\sigma t_0)^2} \tag{21b}$$

$$S_i = \sqrt{I_S^{(i)}} \, e^{\pm i \pi/2} \quad , \quad I_S^{(i)} = \frac{\sin^2 \pi\sigma z_i}{(\sigma t_0)^2} \tag{21c, d}$$

(the $\pm$ signs correspond to the cases $\sin \pi\sigma z_i \gtrless 0$).

If (21) is taken into account one obtains from (20)

$$I_\pi = I_\pi^{(1)} I_\pi^{(2)} + I_s^{(1)} I_s^{(2)} + 2\left(I_s^{(1)} I_\pi^{(1)} I_s^{(2)} I_\pi^{(2)} \right)^{1/2} \cos \frac{2\pi}{\Lambda}(x - x_0) \tag{22}$$

where x_0 is determined by the relations

$$\exp i \frac{2\pi}{\Lambda} x_0 = \pm \exp i (\Phi_1 + \Phi_2) \tag{23a}$$

or explicitely

$$\left(I_\pi^{(1)} I_\pi^{(2)} \right)^{1/2} \cos \frac{2\pi}{\Lambda} x_0 = \pm \left[\cos \pi\sigma (z_1 + z_2) + \frac{\sin \pi\sigma z_1 \sin \pi\sigma z_2}{(\sigma t_0)^2} \right] \tag{23b}$$

$$\left(I_\pi^{(1)} I_\pi^{(2)} \right)^{1/2} \sin \frac{2\pi}{\Lambda} x_0 = \pm \left[-\frac{1}{\sigma} \sin \pi\sigma (z_1 + z_2) \right] \tag{23c}$$

$$\left(\pm \text{ if } \sin \pi\sigma z_1 \sin \pi\sigma z_2 \lessgtr 0 \right)$$

and so also

$$\operatorname{tg} \frac{2\pi}{\Lambda} x_0 = \frac{-1/\sigma \sin \pi\sigma (z_1 + z_2)}{\cos \pi\sigma (z_1 + z_2) + \frac{\sin \pi\sigma z_1 \sin \pi\sigma z_2}{(\sigma t_0)^2}} \tag{23d}$$

We shall now proceed to a discussion of some important aspects of the moiré intensity distribution given by (22), assuming the anomalous absorption to be negligible.

1) <u>The position of the dark and bright fringes</u>

The centres of the dark fringes are at $x = x_0 + k\Lambda$ (k : integer or zero), whilst the centres of the bright fringes are given by $x = x_0 + (k + \frac{1}{2})\Lambda$ (k : integer or zero).

Formula (23) shows that the actual position of the fringes depends in a complicated way on the thicknesses z_1 and z_2 and on the crystal orientation s.

2) <u>The contrast between dark and bright fringes</u>

The difference C_π between the maximum value of I_π

$$I_\pi^{(max)} = \left[\left(I_\pi^{(1)} I_\pi^{(2)} \right)^{1/2} + \left(I_s^{(1)} I_s^{(2)} \right)^{1/2} \right]^2 \tag{24a}$$

and the minimum value of I_π

$$I_\pi^{(min)} = \left[\left(I_\pi^{(1)} I_\pi^{(2)} \right)^{1/2} - \left(I_s^{(1)} I_s^{(2)} \right)^{1/2} \right]^2 \tag{24b}$$

is:

$$C_\pi = 4 \left(I_s^{(1)} I_\pi^{(1)} I_s^{(2)} I_\pi^{(2)} \right)^{1/2} \tag{25a}$$

or explicitely

$$C_\pi = \left[4\frac{\sin^2\pi\sigma z_2}{(\sigma t_0)^2}\left(1-\frac{\sin^2\pi\sigma z_2}{(\sigma t_0)^2}\right)\right]^{1/2}\cdot\left[4\frac{\sin^2\pi\sigma z_2}{(\sigma t_0)^2}\left(1-\frac{\sin^2\pi\sigma z_2}{(\sigma t_0)^2}\right)\right]^{1/2} \qquad (25b)$$

This last formula indicates how the contrast dark-bright of a moiré pattern depends on the thicknesses of both parts of the crystal and on the crystal orientation.

If (z_1, z_2) and (or) s changes, the position of the fringes and their contrast are altered in a way difficult to predict, since formulae (23) and (25) express a rather complicated dependence on these parameters. The direction of the fringes and their spacing are influenced neither by a variation of thicknesses nor by a variation of orientation (of course, only small changes in orientation are considered, such that the same reflection is always active).

We shall now consider in some detail the behaviour of the moiré for variations of the parameters z_1, z_2 and s which result (a) from the presence of a step , and (b) from tilting the crystal.

3) <u>Influence of a step on the surface.</u>

Since $I_S^{(i)}$, $I_\pi^{(i)}$ $(i=1,2)$, C_π and x_0 are periodic in z_2 with period the extinction distance, a step with height $\Delta z_2 = k\, t_0'$ ($t_0' = t_0/\sqrt{1+(s t_0)^2}$, k : integer) will influence neither the position of the fringes and their profile nor their contrast. If the step is not a multiple of the extinction distance, these features may change according to formulae (23), (22) and (25).

Let us first consider the pure dynamical case, s = o. For s = o, formula (18) becomes

$$T(s=0) = \exp i\pi\frac{x}{\Lambda}\left[\cos\pi\frac{z_1+z_2}{t_0}\cos\pi\frac{x}{\Lambda} - i\cos\pi\frac{z_1-z_2}{t_0}\sin\pi\frac{x}{\Lambda}\right].$$

For the fringe profile itself, we obtain then

$$I_\pi(s=0) = \cos^2\pi\frac{z_1+z_2}{t_0}\cos^2\pi\frac{x}{\Lambda} + \cos^2\pi\frac{z_1-z_2}{t_0}\sin^2\pi\frac{x}{\Lambda} \qquad (26)$$

If the increase Δz_2 of z_2 is introduced in (26), it is easily seen that the positions of the fringes are not perturbed. This corresponds to the fact that the second member of (23,c) vanishes for s = o. The intensities of the fringes, however, will be changed, and for adequate step heights it is even possible that maxima become minima and vice versa. For values of Δz_2 such that

$$\cos^2\pi\frac{z_1+(z_2+\Delta z_2)}{t_0} = \cos^2\pi\frac{z_1-(z_2+\Delta z_2)}{t_0}$$ the moiré pattern stops at the step.

Summarising : in the pure dynamical case, the fringes , when crossing a step may behave in quite different ways : (1) there is no change in position and little or no change in intensity; (2) there is no change in position but an appreciable change in intensity; (3) they stop at the step; (4) the positions of the dark and the bright fringes are interchanged which is accompanied by an appreciable change in intensity; (5) the interchange is not accompanied by an appreciable change in intensity. The actual behaviour will depend on the thicknesses z_1 and z_2 and the stepheight Δz_2.

To make this argument clearer, we shall give a simple example : suppose $z_1 = 1/4\, t_0 + k t_0$, $z_2 = 1/4\, t_0 + k' t_0$ (k, k' : integer or zero). Then $I_\pi = \sin^2 \pi \frac{x}{\Lambda}$ is the value of the intensity before crossing the step. The maximum value of I_π is 1 for $x = (l + \frac{1}{2})\Lambda$ and the minimum value is 0 for $x = l\Lambda$ (l, l' : integer or zero).

After crossing a step Δz_2 one has

$$I_\pi = \sin^2 \frac{\pi \Delta z_2}{t_0} \cos^2 \pi \frac{x}{\Lambda} + \cos^2 \frac{\pi \Delta z_2}{t_0} \sin^2 \pi \frac{x}{\Lambda}$$

For different values of Δz_2, all five possibilities may occur, as is shown in table I.

TABLE I

	max.	min.	
$\Delta z_2 = 1/6\, t_0$	$x = \Lambda/2$, $I = 3/4$	$x = 0$, $I = 1/4$	possibility 2
$\Delta z_2 = 1/4\, t_0$	constant intensity 1/2		possibility 3
$\Delta z_2 = 1/3\, t_0$	$x = 0$, $I = 3/4$	$x = \Lambda/2$, $I = 1/4$	possibility 4
$\Delta z_2 = 1/2\, t_0$	$x = 0$; $I = 1$	$x = \Lambda/2$, $I = 0$	possibility 5
$\Delta z_2 = t_0$	$x = \Lambda/2$, $I = 1$	$x = 0$, $I = 0$	possibility 1

If $s \neq 0$, the situation becomes more complicated. The five possibilities may occur again, but there is also a shift in the position of the fringes.

The displacement Δx_0 of the fringes should be calculated from equations (23) which express the dependence of x_0 on z_2 .

One has

$$\exp i \frac{2\pi}{\Lambda} \Delta x_0 = \pm \exp i \Delta\phi_2 \quad (\pm \text{ if } \sin \pi\sigma z_2 \sin \pi\sigma (z_2 + \Delta z_2) \gtrless 0) \qquad (27a)$$

$$\text{with } \Delta\phi_2 = \phi_2 (z_2 + \Delta z_2) - \phi (z_2) \qquad (27b)$$

After some calculations one also obtains

$$\operatorname{tg}\frac{2\pi}{\Lambda}\Delta x_0 = -\operatorname{tg}\pi\sigma\Delta z_2 \frac{s/\sigma}{1-\frac{1}{(\sigma t_0)^2}\,\frac{\sin\pi\sigma z_2\sin\pi\sigma(z_2+\Delta z_2)}{\cos\pi\sigma\Delta z_2}} \qquad (28)$$

This expression yields for Δx_0 two different values, their difference being $\Lambda/2$. To decide which of these values is the correct one, relations (27) (21a,b) and (20b) should be considered.

Since the distance of a bright to a dark fringe is also $\Lambda/2$, a value Δx_0 satisfying (28) represents the separation of a dark fringe at the one side of the step and a dark or a bright fringe at the other side.

The displacement Δx_0 depends on z_2, s and the stepheight Δz_2. The first factor of the second member of (28) $\operatorname{tg}\pi\sigma\Delta z_2 = \operatorname{tg}\pi\frac{\Delta z_2}{t_0}\sqrt{1+(st_0)^2}$, describes a simple dependence on s and Δz_2; z_2 itself does not influence that factor. If the second factor were 1, the formula (28) would yield $\left|\frac{\Delta x_0}{\Lambda}\right| = \frac{1}{\pi}\frac{\Delta z_2}{t_0}\sqrt{1+(st_0)^2}$. This simple result is in fact altered by a second factor, which differs not much from 1 if s/σ is sufficiently close to 1, i.e. if $(st_0)^2 \gg 1$. For values of s not satisfying this condition, the second factor may have values significantly different from 1. The simple formula for $\left|\frac{\Delta x_0}{\Lambda}\right|$, valid for the kinematical case, has to be corrected by a factor which describes the dynamical character. It does not only depend on s an Δz_2 but also on the crystal thickness z_2. From (28) and the foregoing argument, it follows that changing s into $\sigma = \sqrt{1+(st_0)^2}/t_0$ does not completely take into account the dynamical corrections.

4) <u>Effect of tilting the crystal</u>

If the crystal is tilted, $|s|$ varies from o, for the purely dynamical case, towards higher values of $|s|$. For s-values such that $(st_0)^2 \gg 1$, the kinematical approximation, to be discussed in the next paragraph, may be applie resulting in a simplification of the description of the behaviour of the moiré If s changes, $I_m^{(i)}, I_s^{(i)}$ $(i=1,2)$ and x_0, which are functions of this parameter, will vary. This means that the details of the moiré fringe pattern will be modified in a continuous manner during tilting. Whilst the direction and the interspacing of the fringes remain unaltered, they are shifted and the contrast between dark and bright fringes and their intensities vary. Let us examine this behaviour in somewhat more detail.

If the crystal is tilted out of the Bragg condition s = o, the displacement of the fringe is governed by the formulae (23).

In this context, we prefer to rewrite (23,d) in the form

$$\operatorname{tg} 2\pi \frac{x_0}{\Lambda} = -\operatorname{tg} \pi\sigma(z_1+z_2) \frac{s/\sigma}{1 + \frac{\sin \pi\sigma z_1 \sin \pi\sigma z_2}{(\sigma t_0)^2 \cos \pi\sigma(z_1+z_2)}} \qquad (29a)$$

For $(s t_0)^2 \gg 1$, i.e. for the kinematical case, the position of the fringe is given by the simple relation

$$\frac{x_0}{\Lambda} = -\frac{z_1+z_2}{2t_0}\sqrt{1+(s t_0)^2} \simeq -\frac{z_1+z_2}{2} s \qquad (29b)$$

For smaller values of $s t_0$, i.e. for the more dynamical case, the dependence of x_0 on s is more complicated, since the second factor of the second member of (29a) may adopt values significantly different from 1. In particular, the fringe position for a certain s will not only depend on $z_1 + z_2$, the total thickness, but on z_1 and z_2 separately.

During tilting of the crystal, the moiré pattern not only moves, but the contrast between its dark and bright fringes varies. The variation is given by formula (25b), and in order to be able to make predictions about contrast behaviour during tilting, C should be studied as a function of s.

It seems difficult to achieve, within the framework of this paper, a complete discussion of this behaviour, as C depends on the two parameters z_1 and z_2. We shall restrict ourselves to some remarks and some numerical examples. We may write

$$C_T = (C_1 C_2)^{1/2} \qquad (30a)$$

with $$C_i = 4 I_S^{(i)} I_T^{(i)} = 4 \frac{\sin^2 \pi\sigma z_i}{(\sigma t_0)^2}\left[1 - \frac{\sin^2 \pi\sigma z_i}{(\sigma t_0)^2}\right] \qquad (30b)$$

The factor C_i will acquire the absolute maximum value 1 for values of $\sigma t_0 = \sqrt{1+(s t_0)^2}$ for which

$$I_S^{(i)} = \frac{\sin^2 \pi(\sigma t_0)\frac{z_i}{t_0}}{(\sigma t_0)^2} = \frac{1}{2}$$

For values of s for which $I_S^{(i)}$ differs not too much from 1/2, C_i will retain appreciable values.

If $|s|$ varies from zero to infinity, $I_S^{(i)}$ fluctuates. It has minimum values zero for s-values given by $(z_i/t_0)\sigma t_0 = k$.(k : integer) and adopts maximum values for $(z_i/t_0)\sigma t_0 = \gamma_k$ if γ_k are the roots of the transcedental equation $\operatorname{tg} x = x$. Successive maxima decrease approximately as $1/(\sigma t_0)^2$.

Except for extremely thin crystals, for which $z_i/t_0 < \sim 1/4$, $I_s^{(i)}$ will acquire the value 1/2 for one or two $|s|$ -values in the first interval. By a s-interval is meant an interval defined by two successive zeros of $I_s^{(i)}$. If in the second interval the maximum of $I_s^{(i)}$ is still larger than 1/2, two further s-values occur for which $I_s^{(i)} = 1/2$. This will happen for crystals which are not too thin. For thicker crystals, it is even possible that for s-values of the third, fourth,..... interval. However, for these crystals the "non-absorption" dynamical theory cannot be relied on, anomalous absorption effects becoming important (see § 6).

Apart from the important maxima already discussed, it is clear from (30,b) that C_i oscillates, being zero where $I_s^{(i)}$ is zero, and being maximum where $I_s^{(i)}$ is maximum. However, in a s-interval where $I_s^{(i)} = 1/2$ for two s-values, the s for which $I_s^{(i)}$ is maximum, will yield for C_i not a maximum but a minimum.

Let us consider now, first, the easiest case to discuss, the symmetrical case $z_1 = z_2$ (= z). Then $C_T = C_1$ or C_2, and the qualitative analysis of C_i just performed, describes then what should be observed in this special case. For certain small s_1 - values $(|s| < \frac{1}{t_0})$ a deeply contrasted moiré (C = 1) is observed. If the crystal is now tilted towards higher s_1 -values, the contrast will remain appreciable for a certain range, depending on the thickness z, to decrease then quickly to zero. The moiré disappears during tilting. If the tilting is continued, two qualitatively different behaviours are possible. In principle, the contrast will increase again and will reach in the second s-interval one or two maxima. If these are again of the order one, an observably contrasted moiré will be revealed again. After disappearing rather quickly, the pattern reappears. This type of behaviour should be expected for crystals which are not too thin (z_i/t_0 sufficiently large) The pattern may disappear and reappear several times, the contrast decreasing however mostly rather drastically from the third interval on.

The second possibility occurs if the maximum of the contrast in the second interval is small, due to the fact that the corresponding maximum of $I_s^{(i)} (= I_s^{(i)})$ is not important. The maximum intensity of the moiré (centre of the bright fringe) for the symmetrical case $I_T^{(max)} = (I_T^{(i)} + I_s^{(i)})^2$ is one. For the s-values corresponding to the maximum of the second, third.... interval the moiré intensity is given by $I_+ = 1 - C \sin^2 \frac{\pi}{\Lambda}(x - x_0)$ where C is small ($C \lesssim 1/4$).

We believe that such a pattern would be hardly visible, or at least will be easily overlooked. A well defined moiré pattern should be revealed for one or two small s -values, but should disappear rather slowly during tilting, and should not reappear, or at least should reappear vaguely.

This type of behaviour should occur for rather thin crystals, let us say for which $z < \sim 0{,}4\, t_0$. It should be noticed that a crystal of a given substance and thickness may behave differently for different reflections, since the extinction distance t_0 depends on the reflection considered, and the quantity involved is z/t_0. One should also keep in mind that a certain crystal thickness may be considered as being thin for one crystal substance but not for another material, since t_0 varies from substance to substance. During tilting of the crystal, equation (29,a) describes how the fringe positions varies continuously. It should, however, be remarked that after passing an orientation for which the moiré disappears, the dark fringe reappears as bright and vice versa.

In fig. 2 are plotted some C_T-curves for the symmetrical case :

- Curve (a) is for $z_1 = z_2 = 1/3\, t_0$;
  C will be zero for $s t_0 = \pm\sqrt{(3k)^2 - 1}$ (k = 1,2 ....). In the first s-interval ($0, 2\sqrt{2}/t_0$), $I_s^{(i)}$ will vary from 3/4 to 0 in a monotonous way, such that only for one s-value $s \simeq 1/t_0$, $I_s^{(i)}$ will reach the critical value 1/2, which yields for C the absolute maximum value 1. In the second interval ($2\sqrt{2}/t_0, \sqrt{35}/t_0$), $I_s^{(i)}$ has a maximum value $\sim 0{,}05$ for $s \simeq 4{,}2/t_0$ which leads to a second maximum for the contrast C, $C_{max} = 0{,}2$. It is questionable whether this second maximum will be observable or not.
- Curve (b) is for $z_1 = z_2 = 2/3\, t_0$;
  C = o for $s t_0 = \pm\sqrt{(3k/2)^2 - 1}$ (k : 1,2 .....)
  In the first s-interval ($0, \sqrt{5}/2\, t_0$), $I_s^{(i)}$ decreases monotonously from 3/4 to o, so that for one s-value of this interval, $I_s^{(i)}$ becomes 1/2, and as a consequence C = 1. In the second interval ($\sqrt{5}/2\, t_0, 3\sqrt{2}/t_0$) the maximum of $I_s^{(i)}$ is only 0,2 to which corresponds however the appreciable value C = $\sim$0,65 which should surely be observable.
- Curve (c) is for $z_1 = z_2 = \frac{7}{6}\, t_0$;
  C = o for $s t_0 = \pm\sqrt{(6k/7)^2 - 1}$ (k : 2, 3, 4 .....).
  In the first s-interval ($0, \sqrt{95}/7t_0$), $I_s^{(i)}$ increases first monotonously from 1/4 to the maximum $\sim$0,63, to decrease then again to o. In this interval $I_s^{(i)}$ will then be equal to 1/2 for two different s-values, which means that C will become 1 for two different values of s. Furthermore, the successive maxima for C have non negligible values.

- Curve (d) is for $z_1 = z_2 = 5/3\, t_0$;
  $C = 0$ for $s t_0 = \pm\sqrt{(3k/5)^2 - 1}$ (k : 2, 3 .....)
  In the first and second interval , C reaches the maximum value one, in the following s-intervals the values of the maximum decrease rather slowly, so that it seems that at least for the first of these series an observable moiré pattern is present.

We shall now discuss the asymmetrical case $z_1 \quad z_2$. A general discussion being impossible, we will restrict ourselves to some general remarks and some numerical examples.

The contrast $C_T = \sqrt{C_1 C_2}$ is now the product of two factors $\sqrt{C_1}$ and $\sqrt{C_2}$, the variation of each having been discussed above. C has only values of the order 1, if there exists s-values for which C_1 and C_2 are both close to one. From the examples discussed above, we may conclude that in general the s-intervals for which C_1 and C_2 are of the order 1, are rather broad. It seems reasonable to accept that nearly always both intervals overlap. This means that we can usually predict also for the asymmetrical case crystal orientations s for which moiré patterns show up with deep, well observable contrast, mostly for small values of s. It may however be expected that the further maxima for C will be smaller or even much smaller than for C_1 and C_2. Moreover, the successive C-maxima do not necessarily decrease monotonously, but they decrease in general in a modulated way.

In fig. 3 the contrast C_T and the bright fringe maximum of the bright field moiré pattern are plotted as a function of s, for $z_1 = 2/3\, t_0$, $z_2 = 7/6\, t_0$ [curves (a) and (c)] . With the aid of these two curves, the fringe profile for a given s can be constructed.

C is zero for $s t_0 = \pm\sqrt{(6k/7)^2 - 1}$ (k : 2, 3.....)and $s t_0 = \pm\sqrt{(3k/2)^2 - 1}$ (k : 1, 2 ...). In the first s-interval $(0, \sqrt{5}/2\, t_0)$ C obtains once (for $s \simeq 0{,}9/t_0$) the maximum value 1; in the second interval ($\sqrt{5}/2\, t_0, \sqrt{95}/7 t_0$) the C-values remain negligible small, but in the third interval ($\sqrt{95}/7 t_0, 2\sqrt{2}/t_0$) the contrast rises again to an appreciable value (2/3). The fluctuations in the following intervals are unimportant. The main difference between the moiré for $s \simeq 0{,}45/t_0$ and $s \simeq 1{,}55/t_0$ is that, whilst for the first the minimum intensity of the dark fringe is zero, this is not the case for the second, the minimum being now 1/3. In both cases the intensity of the bright fringes is one.

Curves (b) and (d) of figure 3 represent the very asymmetrical situation $z_1 = 1/3\ t_o$, $z_2 = 5/3\ t_o$.

The C_T-curves may be considered as the C_T curve for $z_1 = z_2 = 5/3\ t_o$, (see figure 2,d), modulated by the C_T-curve for $z_1 = z_2 = 1/3\ t_o$ (see fig. 2,a). C is zero for $st_o = \pm\sqrt{(3k/5)^2 - 1}$ (k : 2, 3 ....) and $s t_o = \pm\sqrt{(3k)^2 - 1}$ (k : 1,2...) In the first and second s-interval exists an s-value, respectively 0,25 / t_o and 1,15 / t_o to which corresponds maximum values of C_T respectively equal to 0,9 and 1 (in both cases the dark fringe minimum is approximately zero). In the third s-interval, an important maximum still occurs; the corresponding moiré exhibits, however, fringes which are no longer completely dark (intensity $\sim$ 0,3)(the bright fringes have the intensity 1). From the fourth interval on the maxima of the contrast are small.

From formulae (21b,d) and (23) it follows that $I_s^{(i)}$ and $I_\pi^{(i)}$ are even functions of s, whilst x_o is odd in s. If the crystal is tilted, starting from the orientations s = o, in both senses over the same amount, the intensity profile in both cases will be the same, the resulting displacements of the fringes however equal but opposed.

§ 5. THE KINEMATICAL APPROXIMATION

The kinematical approximation applies if $1/(s t_o)^2 \ll 1$

Then $\sigma = \dfrac{\sqrt{1 + (s t_o)^2}}{t_o} \simeq |s|$, $1/(\sigma t_o)^2 = 1/(s t_o)^2$

Introducing these approximations into expressions (21), (22) and (23) determining I_π and neglecting higher powers of $1/(s t_o)^2$, we obtain a kinematical approximation.

$$I_T = 1 - I_S$$

$$I_S = \frac{4}{(s t_o)^2}\left\{\sin^2 \pi s \frac{z_1 + z_2}{2} \cos^2 \pi s \frac{z_1 - z_2}{2} - \sin \pi s z_1 \sin \pi s z_2 \sin^2 \pi\left[\frac{x}{\Lambda} + \frac{s(z_1 + z_2)}{2}\right]\right\} \quad (31a)$$

This formula (31a) is not immediately comparable with that obtained by Rang (loc cit), which is deduced for the special case $z_1 = z_2 = z$. Then (31) reduces to

$$I_S = 4\ I_o \cos^2 \pi\left(\frac{x}{\Lambda} + s z\right) \quad (31b)$$

where $I_o = \sin^2 \pi s z/(s t_o)^2$ is the intensity which should be diffracted by one half of the crystal in the absence of the other half.

This expression is still different from the one obtained by Rang, but reduces to it for s= o. This corresponds to the fact that the theory as developed by Rang is a kinematical one for s=o. It is believed, however, that putting s = o in a kinematical theory is difficult to justify. In any case, the dependence of the moiré profile on crystal thicknesses, in particular its behaviour when crossing a step and during tilting of the crystal, is neither predicted nor described by such simplified kinematical treatment. The method of the "optical analogon" which has been frequently used (see e.g. O. Rang (loc cit)), does not take into account either the influence of the thicknesses or the finite value of s, and obviously not the dynamical effects. Results obtained by this method should be confirmed by theoretical calculations.

Formula (31b) has to be compared to the expression obtained from (22) and (23) for which $z_1 = z_2 = z$, $I_S^{(1)} = I_S^{(2)} = I_0$, $I_\pi^{(1)} = I_\pi^{(2)} = 1 - I_0$, $I_S = 1 - I_\pi$

The correct expression for s = o is

$$I_S = 4\, I_0 (1 - I_0) \cos^2 \pi \frac{x}{\Lambda} \qquad (31c)$$

which is obtained from the formula of Rang by substituting $I_0(1 - I_0)$ for I_0.

For the kinematical approximation, the contrast C is given by $4 \frac{\sin \pi s z_1 \sin \pi s z_2}{(s t_0)^2}$ This expression may in favourable circumstances for values of $s t_0$ as large as 3, 4, 5 .... still have a value not negligible with respect to one. Under the conditions of a kinematical approximation moiré fringes may occur with observable contrast. As a consequence, it makes sense to look for the simplifications introduced by the kinematical approximation for the behaviour of the moiré when crossing a step or during the tilting of the crystal.

The formula (28), which describes the displacement of the dark (bright) fringes when crossing a step with height h, is extremely simplified and yields now

$$\frac{\Delta x_0}{\Lambda} = -\frac{sh}{2} + \begin{cases} 0 & \text{if } \sin \pi s z_2 \sin \pi s (z_2 + h) > 0 \\ \frac{1}{2} & \text{if } \sin \pi s z_2 \sin \pi s (z_2 + h) < 0 \end{cases} \qquad (32)$$

In principle, formula (32) should enable the measurement of by measuring s and $\Delta x_0 / \Lambda$.

The moiré contrast vanishes for s-values satisfying the relations $sz_i = k$ (k : integer). In principle, the measurements of these values should enable the measurement of the crystal thicknesses z_1 and z_2. However, this method should be very difficult or even impossible to apply, since the moiré patterns are likely to be very poorly defined for s-values for which the kinematical approximation is valid.

§ 6. "ANOMALOUS" ABSORPTION EFFECTS

1. Introduction

The discussion of § 4 and 5 is valid if the crystal is thin enough compared to the absorption length, for the effects associated with the anomalous absorption (due to loss of electrons by inelastic scattering) to be neglected. It is well known that these effects are taken into account (Molière, 1939; Yoshida, 1957; Howie and Whelan, 1961) if $1/t$ is put equal to $(1/t)(1+i\alpha)$ where α may be considered as a number small enough for the higher power of to be negligible (Hashimoto et al, 1960; Howie and Whelan, 1960a,b); is called the absorption length.

Expression (22) for the transmitted intensity still holds if the adequate expressions for I_s and I_n are introduced. We have

$$\sigma = \frac{1}{t_0}(1+i\alpha)\left[1+(st_0)^2(1-i\alpha)^2\right]^{1/2} = \frac{\sqrt{1+(st_0)^2}}{t_0} + i\,\frac{1}{\tau_0\sqrt{1+(st_0)^2}} = \beta + i\gamma$$

$$|\sigma|^2 = \frac{1+(st_0)^2}{t_0^2}\ ; \quad \frac{s}{\sigma} = \frac{st_0}{\sqrt{1+(st_0)^2}}\left[1 - i\,\frac{\alpha}{1+(st_0)^2}\right]$$

From

$$T^{(i)} = \cos\pi(\beta+i\gamma)z_i - i\,\frac{s}{\sigma}\sin\pi(\beta+i\gamma)z_i$$

it follows, since

$$I_T^{(i)} = |T^{(i)}|^2 e^{-\mu z_i}$$

(μ : mean absorption coefficient

that $$I_T^{(i)} = \left[\left(\mathrm{ch}\,\frac{\pi z_i}{\tau_0\sqrt{1+(st_0)^2}} + \frac{st_0}{\sqrt{1+(st_0)^2}}\,\mathrm{sh}\,\frac{\pi z_i}{\tau_0\sqrt{1+(st_0)^2}}\right)^2 - \frac{\sin^2\frac{\pi z_i}{t_0}\sqrt{1+(st_0)^2}}{1+(st_0)^2}\right]\exp-\mu z_i$$

if the term $-\alpha\,\frac{st_0}{\sqrt{1+(st_0)^2}}\,\frac{1}{1+(st_0)^2}\sin 2\pi\frac{z_i}{t_0}\sqrt{1+(st_0)^2}$ is neglected. This expression enables to discuss quantitatively the conditions under which the anomalous absorption becomes important.

Negligible anomalous absorption means that the first term of (33) is ~ 1 : for instance, smaller than 1,1. Accepting this suggestion, the anomalous absorption effect may be neglected for crystals such that

$$ch \frac{\pi z_i}{\tau_0} < \sim 1{,}05 \quad \text{or} \quad z_i < \sim 0{,}1\,\tau_0$$

We shall call these crystals thin.

The analytical expression (33) shows clearly that $I_T^{(i)}$ is different for two s-values equal but opposite in sign, and this effect, known in the theory of X-ray diffraction as the Borrmann effect (Borrmann, 1941, 1950; Zachariesen, 1945; Von Laue, 1949; Hirsch, 1952), is only important if the crystal thickness is sufficiently large(s > o means that the reciprocal lattice node defined by lies in the interior of the reflecting sphere)

Crystals somewhat thicker than $0{,}1\,\tau_0$ will be called of "intermediate thickness". For such crystals, the first term of (33) minus one becomes of the same order as the second term. Roughly stated, the results for the non-anomalous absorption are still qualitatively correct, but quantitatively more or less influenced by these absorption effects. If we put forward as a condition that $-1{,}05 < ch \frac{\pi z_i}{\tau_0} < \sim 2$, we obtain $\sim 0{,}1\,\tau_0 < z_i < \sim 0{,}4\,\tau_0$

If the first term of (33) deviates significantly from 1 for s = o, it is obvious that this deviation decreases with s, which means that for increasing s-values the crystal behaves more and more as a thin crystal.

For thick crystals, the first term is the most important one, it determine the character of the variation of $I_T^{(i)}$ with s. For very thick crystals it is even sensible to neglect the second term. It is then easily seen from (33) that $I_T^{(i)}$ will decrease monotonously from a maximum value to zero, and in particular that this maximum does not correspond to s = o but to a value of s depending on the thickness z_i, the extinction distance t_0 and the absorption length τ_0. Moreover, $I_T^{(i)}$ is assymetrical in s.

For the intensity of the diffracted wave, we obtain, in a similar way

$$S_i = \frac{i \sin \pi (p + i\gamma) z_i}{\sigma t}$$

Since

$$I_S^{(i)} = |S_i|^2 \exp - \mu z_i$$

one has

$$I_S^{(i)} = \left(\frac{sh^2 \frac{\pi z_i}{\tau_0 \sqrt{1+(st_0)^2}}}{1+(st_0)^2} + \frac{\sin^2 \frac{\pi z_i}{t_0} \sqrt{1+(st_0)^2}}{1+(st_0)^2} \right) \exp - \mu z_i \qquad (34)$$

Here again we have to consider :

(a) Thin crystals

It is a good approximation to neglect the hyperbolic function and consequently the non-absorption theory applies.

(b) Crystals of intermediate thickness

The general tendency of the variation of $I_s^{(i)}$ with s will not be altered qualitatively, i.e. s-values for which $I_s^{(i)}$ has well-defined maxima and minima are still present. These s-values will be only slightly different from the values predicted by the non-absorption theory. The minimum value of $I_s^{(i)}$ however, which should be zero if anomalous absorption is not taken into account, have no longer negligibly small values, except for sufficiently large s-values.

(c) Thick crystals

The variation of $I_s^{(i)}$ is qualitatively determined by the variation of the hyperbolic function, to which relatively small oscillations, due to the goniometric functions, have to be superposed. For very thick crystals, these can be neglected, yielding for $I_s^{(i)}$ a monotonous decreasing variation, symmetrical in s, the maximum occuring for s = o.

2. Fringe profile of the dark field moiré pattern.

In § 4 the diffracted intensity I_s is not discussed. This discussion was obviously superfluous, since I_s and I_T are complementary, i.e. $I_s = 1 - I_T$ if the anomalous absorption is not considered. If this last effect is taken into account, this is no longer true. Therefore we shall derive from expression (19b) for the amplitude of the diffracted wave, an expression for I_s , in a manner similar to the one used for the derivitaion of the expression for the transmitted wave intensity.

We obtain

$$S = a' + b' \exp - 2\pi \frac{x}{\Lambda} \qquad (35a)$$

with

$$a' = \frac{i}{\sigma t}\left\{ \sin \pi\sigma(z_1 + z_2) - \sin \pi\sigma z_2 \left(\cos \pi\sigma z_1 - i \frac{s}{\sigma} \sin \pi\sigma z_1\right)\right. \qquad (35b)$$

$$b' = \frac{i}{\sigma t} \sin \pi\sigma z_2 \left(\cos \pi\sigma z_1 - i \frac{s}{\sigma} \sin \pi\sigma z_1\right) \qquad (35c)$$

or, after some calculations :

$$a' = S_1 T_2^{(-)} , \quad b' = S_2 T_1 \qquad (35d, e)$$

where

$$T_i^{(-)}(s) = T_i(s) \qquad (35f)$$

One has finally :

$$I_S = I_S^{(1)} I_T^{(-2)} + I_T^{(1)} I_S^{(2)} - 2\left(I_S^{(1)} I_T^{(1)} I_S^{(2)} I_T^{(-2)} \right)^{1/2} \cos \frac{2\pi}{\Lambda}(x - x_0') \qquad (36)$$

$I_T^{(1)}$ is the expression for the intensity of the wave transmitted by a crystal with thickness z_1 for the orientation s, whilst $I_T^{(-1)}$ is then defined by the relation

$$I_T^{(-1)}(s) = I_T^{(1)}(-s) \qquad (37)$$

For the contrast bright-dark fringe of the dark field moiré patterns, one has

$$C_S = \left(C_1 C_2^{(-)} \right)^{1/2}, \text{ if } C_2^{(1)}(s) = C_2(-s) \qquad (38a,b)$$

C_1 and C_2 are defined by

$$C_i = 4 I_S^{(1)} I_T^{(1)}$$

We have already pointeddout that $I_S^{(1)}(s)$ is an even function of s, whilst $I_T^{(1)}(s) \neq I_T^{(1)}(-s)$. A similar effect exists for the dark and bright field moiré patterns for the symmetrical case $z_1 = z_2$. This follows from expressions (36), (33) and (34) for the dark field moiré and from expressions (22) ~~(32)~~ (33) and (34) for the bright field moiré.

From formulae (22) and (36) it follows that the geometrical properties of the moiré pattern, i.e. the direction and separation of the fringes, are not influenced by the anomalous absorption. The position of the fringes, and more important, the actual profile of the fringes, in particular their contrast, may depend strongly on the anomalous absorption.

3. Influence on the position of the fringes

From relations (20) it follows that the position x_0 of the bright field moiré fringes ~~are~~ is defined by

$$\exp i 2\pi \frac{x_0}{\Lambda} = \exp i \left(\phi_T^{(1)} + \phi_T^{(2)} - \phi_S^{(1)} - \phi_S^{(-2)} \right) \qquad (39a)$$

where $\phi_T^{(1)}$ ($\phi_S^{(1)}$) are the phase angles of T_1 (S_1) and $\phi^{(-1)}(s) = \phi^{(1)}(-s)$

In the same way it follows that from relations (35) that the position x_0' of the dark field moiré fringes is determined by

$$\exp i 2\pi \frac{x_0'}{\Lambda} = \exp i \left(\phi_T^{(1)} - \phi_T^{(-2)} - \phi_S^{(1)} - \phi_S^{(2)} \right) \qquad (39b)$$

If anomalous absorption is neglected, $x_0 = x_0'$ (mod Λ), since

$$\phi_T^{(-2)} = -\phi_T^{(2)} \quad \phi_S^{(1)} + \phi_S^{(-2)} = \phi_S^{(1)} - \phi_S^{(2)} \; (\text{mod } 2\pi)$$

The bright fringes of the bright field pattern correspond of course to the dark fringes of the dark field pattern.

If anomalous absorption is important, these last relations are no longer true, since then $T_i(-s) = [T_i(s)]^*$ and the phase angles of S are no longer zero or π. This means that in general $x_0 \neq x_0'$, i.e. the positions of the fringes of both types of patterns differ, slightly for crystals of intermediate thickness, to a more pronounced degree for thicker foils.

The displacement of the fringes if crossing a step on the surface or during tilting of the crystal will also be significantly altered for thicker specimens. The formulae (28) and (29a) should be replaced by more complicated ones, depending now on τ_0, and which should be calculated with the aid of the relations (39).

4. Position of the interface

Let us first examine in some detail the influence of absorption on the dependence of the contrast from the position of the interface.

For a thin crystal $C_T (= C_S)$ is periodic in z_1 (or z_2) with period (fixed s) since $I_S^{(i)}$ $(i = 1, 2)$ have this periodicity. Positions of the interface differing by a multiple of $t_0' = 1/\sigma = t_0/\sqrt{1+(s t_0)^2}$ give rise to the same moiré pattern. If the interface is now thought to be displaced continuously from o to z, total thickness, C will vary periodically having minimum values zero for $z_1 = k t_0'$, $z - z_1 = k' t_0'$ (k, k' : integers) and maximum values and eventually subsidary minimum values for positions which depend on the total thickness. For $s t_0 = 0$, $C_T (= C_S) = \left| \sin 2\pi \frac{z_1}{t_0} + \sin 2\pi \frac{z_2}{|t_0|} \right|$ and its periodicity is halved and is equal to $1/2 t_0$.

For a thick crystal C_T and C_S are of course still varying with the positions z_1 of the interface, althoughthe variation is no longer periodic, due to the presence of the hyperbolic functions in the adequate expressions for $I_S^{(i)}$ and $I_T^{(i)}$ $(i = 1, 2)$. For a symmetrical or nearly symmetrical position of the interface, and small s-values the variation of the contrast is mainly determined by the variation of the hyperbolic functions. This results in a rather slowly monotonous variation, on which oscillations due to the goniometric functions have to be superposed. These are small compared with the mean of the contrast.

However, if the interface is moved closer to one of both surfaces, the character of the variation changes, since one of both crystal parts becomes thinner. Analytically this means that if z_2 decreases, the factor $(4 I_S^{(2)} I_T^{(2)})^{1/2}$ will behave as in the case of a thin crystal. A thick crystal with an interface very asymmetrically situated is of mixed type : one of both halves is thick, whilst the other is of intermediate thickness or thin. Strong fluctuations are now expected. The variation, however, is not the same as for a thin or intermediate crystal, since the variation of the factor $(4 I_S^{(1)} I_T^{(1)})^{1/2}$ corresponding to the thick part of the crystal remains slow.

Fig. 4 gives the plot of C_T versus z_1 (or z_2) for and shows quantitatively the properties described above. The fully drawn curve of fig. 4 is obtained if anomalous absorption is taken into account, whilst the broken line (if periodically repeated) pictures the case of non absorption. It is easily seen that the anomalous absorption effects change little the values of z_1 for which the contrast is maximum and minimum, but may have strong influence on the relative values of the minima and maxima. If the position of the interface is nearly symmetrical, the fluctuations of the contrast with interface positions are not important, and this effect will be more pronounced for thicker crystals.

From the definition (30,a) it follows that C_T is symmetrical in z_1 and z_2, and this also if anomalous absorption is important, i.e. the positions of the interface symmetrical with respect to the middle of the crystal sandwich give rise to the same contrast for the bright field moire pattern. From (38) it follows that the dark field moiré contrast has this property only if the anomalous absorption is negligible, since otherwise $C_1(-1) C_2(-1) \neq C_2(1) C_1(-1)$ as a consequence of $C_1(-1) \neq C_1(1)$.

5. <u>Tilting of the crystal</u>.

If absorption is neglected, the contrast bright-dark fringes is the same for bright and dark field patterns. From (25a) and (38) follows $C_T = C_S$ since $I_T^{-1} = I_T$. This last equality does no longer hold if anomalous absorption is taken into account.

We can conclude that this effect will tend to differentiate the contrast for dark and bright field moiré patterns. We have $C_T(-s) \neq C_T(s)$, since $C_1(-s) \neq C_1(s)$, whilst $C_S(-s) = C_S(s)$ if $z_1 = z_2$ (the symmetrical case), but also $C_S(-s) \neq C_S(s)$ if $z_1 \neq z_2$ (the asymmetrical case).

Let us now examine in some detail, with the aid of some numerical examples, how anomalous absorption influences the variation of the fringe profile with crystal orientation s.

We consider first the symmetrical case $z_1 = z_2 = t_0, \alpha = 0,1$, i.e. the case of a rather thin crystal.

In fig. 5 the curve showing the variation of C_T (or C_S) the anomalous absorption neglected, is plotted [curve (a); the bright fringe maximum for the bright field moiré is equal to 1; the bright and the dark field moiré are complementary] .

In fig. 7 are given the contrast of the bright field moiré and its bright fringe maximum, the contrast of the dark field moiré and its dark fringe minimum [curves (a), (b), (d) (d)] .

Comparison of both figures suggests that the influence of the anomalous absorption on the positions of the maxima and minima of the contrast is negligible; for the ratio of the first and the second maximum its influence amounts only to a rather small correction decreasing somewhat the relative importance of the second maximum; the asymmetry of C_T in s is the most significant of the effects, reducing rather strongly for $s < o$ the intensities of the bright fringes of the bright field moiré.

Possibly except for this last effect, no great harm would be done by neglecting the anomalous absorption, and this will be certainly the case for thinner crystals.

Now we shall consider the symmetrical case $z_1 = z_2 = 2\,t_0, \alpha = 0,1$, i.e. the case of a crystal of intermediate thickness. The curves of figures 5 (curve b) and 8 (curves (a), (b), (c), (d)) have the same significance as for the preceding example. These curves demonstrate that the anomalous absorption affects the details of the variation of the contrast for values of s varying approximately between -2 and + 2. The most striking differences between these cases are :

- the ratio of the maxima of the first interval to the maxima of the further intervals of the bright field contrast are rather strongly influenced. The non-absorption result predicts for the ratio of the maximum of second interval to the maxima of the first interval the value ~0.9, i.e. there is little or no difference in the aspect of the moiré for both crystal orientations, since in both cases the bright fringe maximum is one. If the anomalous absorption is taken into account, this theoretical value is reduced to its real value ~0,55. In more detail we have :

  for the moiré in the first interval : bright fringe maximum intensity 1
  dark fringe minimum intensity 0;

  for the moiré in the second interval : bright fringe maximum intensity 0,8
  dark fringe minimum intensity 0,26

  We can conclude that the anomalous absorption differentiates the aspects of the moiré for the two considered crystal orientations.
- for the non-absorption treatment , the contrast becomes zero, i.e. the moiré pattern disappears for orientations $st_0 = \pm\sqrt{(k/2)^2 - 1}$ (k = 2, 3 ...)
  If absorption is now considered, the s-values for which the contrast is minimum are very approximately the same, but the corresponding values for the contrast are no longer zero, although, for the thickness considered here, only the value of the first minimum is non-negligible.
- the s-values for which minima and maxima of the contrast occurs may be considered as being not noticeably influenced, possibly for the maximum of the first interval, its displacement being in any case very small.
- the asymmetry in s of the bright field moiré remains however the most striking feature (for sufficiently small s-values).

Let us now consider the case of a somewhat thicker crystal, $z_1 = z_2 = st_0$, $\alpha = 0,1$. Figures 6 (curve a) and 9 (curves a, b, c, d) show again the curves to be considered.

Clearly, the same influence of the anomalous absorption as for the preceding example is revealed by those curves, yet the discussed effects are now more important as a consequence of the increased thickness.

It is not difficult to predict how the situation should evoluate if the crystal thickness is more and more increased. The effects discussed for the examples would become still more pronounced.

Finally, the curves of interest, figure 6 (b,c) , fig. 10 (a),(b),(c), (d), have been computed for the asymmetrical case $z_1 = t_0$, $z_2 = 3t_0$, $\alpha = 0{,}1$. The anomalous absorption effects, showing up for the other examples discussed, are again present. Even for this rather thick specimen, with asymmetrically situated interface, the correction, for the s-values determining the successiv maxima and minima of the moiré contrast, amounts only to a few per cent. The relative magnitudes of these maxima and minima are however strongly influenced. The first minimum occurs for $st_0 \simeq 0{,}9$. The non-absorption theory predicts that the moiré should disappear for this crystal orientation during slow tilting of the crystal. The fluctuating character of the moiré contrast should be easily detectable. Let us examine if this is still true when anomalous absorption is taken into consideration.

a) <u>The bright field moiré pattern</u>

For $st_0 \simeq 0{,}55$, the contrast is maximum, the moiré pattern is characterized by (the factor taking into account the normal absorption will be left out)

$I_{\pi}^{(max)} = 5{,}4$, $I_{\pi}^{(min)} = 1{,}4$, $I_{\pi}^{(min)} / I_{\pi}^{(max)} = 0{,}26$

If st increases, the bright fringe maximum and the contrast decreases and a minimum for the latter is reached for $st_0 \simeq 0{,}9$. The moiré pattern is now described by $I_{\pi}^{(max)} = 4{,}5$, $I_{\pi}^{(min)} = 1{,}8$, $I_{\pi}^{(min)} / I_{\pi}^{(max)} = 0{,}4$ (to compare with the value 1 obtained if absorption is neglected).

This means that the bright fringe intensity decreases, whilst the dark fringe intensity increases during tilting from orientation $st_0 = 0{,}55$ on towards orientation $st_0 \simeq 0{,}9$, both however in a not very pronounced way. The patterns for both orientations are not very different. We are inclined to believe that only careful observation should réveal the fluctuating character of the moiré contrast. The pattern can roughly be considered as disappearing slowly and nearly monotonously during continued tilting. We can conclude that anomalous absorption affects significantly the behaviour of the moiré pattern, notwithstanding the fact that maxima and minima of the contrast still occur for nearly the same s-values.

b) The dark field moiré

We have

for $s t_0 \simeq 0,55$ $\quad \left.\begin{array}{l} I_S^{(max)} = 4,2 \\ I_S^{(min)} = 1,4 \end{array}\right\} \; I_S^{(min)} / I_S^{(max)} = 0,33$

for $s t_0 \simeq 0,9$ $\quad \left.\begin{array}{l} I_S^{(max)} = 1,55 \\ I_S^{(min)} = 0 \end{array}\right\} \; I_S^{(min)} / I_S^{(max)} \simeq 0$

These data suggest that the variation of the dark field pattern is more similar to the type of variation predicted by non-absorption treatment than it was the case for the bright field moiré. In particular, for $s t_0 \simeq 0,9$ the moiré pattern does not disappear completely, but the bright fringe intensity is strongly reduced and the dark fringe becomes completely dark, which should easily be observable in the electron microscope.

It seems that the anomalous absorption will affect more strongly the qualitative behaviour of the bright field moiré pattern than the behaviour of the dark field moiré pattern.

As for the other examples, the asymmetry in s is very important. In the cases considered hitherto ($z_1 = z_2$), the bright field pattern only was asymmetrical, whereas now both types of patterns become asymmetrical.

§ 7. GENERALIZATION

Expressions (20) and (35) for T and S depend only on the amplitudes of the waves transmitted and diffracted by each component of the crystal sandwich separately. This suggests that it should be possible to derive these formulae by a reasoning involving only these waves.

If $\underline{K}_0$ is the wave vector of the incident beam, corrected for refractio and $\underline{g}$ the diffraction vector, the transmitted and diffracted beams are represented by

$$T(z) \exp i 2\pi \underline{K}_0 \cdot \underline{r} = T(z) \exp i \pi s z \exp i 2\pi \underline{K}_0 \cdot \underline{r} \qquad (40a)$$

$$S(z) \exp i 2\pi \underline{K}_{\underline{g}} \cdot \underline{r} = S(z) \exp -i \pi s z \exp i 2\pi (\underline{K}_0 + \underline{g}) \cdot \underline{r} \qquad (40b)$$

On the other hand, assuming the two-beam approximation is valid, we have for the electron wave function :

$$\Psi = \Psi_0 \exp i 2\pi \underline{K}_0 \cdot \underline{r} + \Psi_{\underline{n}} \exp i 2\pi \underline{K}_{\underline{n}} \cdot \underline{r} \qquad (41)$$

where $\underline{K}_{\underline{n}} = \underline{K}_0 + \underline{n}$, $\underline{n}$ is the reciprocal lattice vector considered.

Since $\underline{g} = \underline{n} + \underline{s}$, we have the relations

$$\psi_0 = \mathcal{E}(z) = \exp i\pi s z\, T(z), \quad \psi_{\underline{n}} = \exp i 2\pi \underline{s}\cdot\underline{r}\, \mathcal{S}(z) = \exp i\pi s z\, S(z) \qquad (42a, b)$$

where T and S are given by (20,b,f).

From now on we shall use the wave vector representation (41) instead of the diffraction vector representing (40).

Fig. 11 represents the A_3 -plane [$(h_1 k_2 o)$- plane] of the reciprocal lattice with the vectors n and n' defining the reflecting planes in the first and second components of the crystal sandwich, (a'a) and (b b') are the intersections of the corresponding Brillouin zones. O is the origin of the reciprocal lattice, whilst C is the projection of the centre of the reflection sphere. As proposed and demonstrated experimentally by Bassett et al. (1958), the bright (dark) field moiré pattern arises by the interference of two transmitted (diffracted) beams (in the two-beam case).

These are :

1) The "doubly" transmitted wave, i.e. the incident beam is first transmitted through the first part of the crystal (or the first crystal), falls on the second part (or second crystal) and it is transmitted again.

At the interface the amplitude of the wave is $T_1(s, z_1, t_1) \exp i\pi s z_1$. If x is the distance of C to the Brillouin zone, s is given by the relation $s = n\lambda x$ (λ: wave length of electron beam) (Whelan and Hirsch, loc cit). A wave with amplitude 1 falling on the second part and transmitted further, will arrive at the back surface with an amplitude $T_2(s', z_2, t_2) \exp i\pi s' z_2$, where $s' = n'\lambda x'$ (x': distance from C to the Brillouin zone (bb')). We have introduced for t two possibly different values t_1 and t_2 , in odsr to cover also the case of two superposed crystals of different crystalline material, or of two superposed crystals of the same substance but with different reflections active.

The doubly transmitted wave is then represented by

$$\exp i\pi (s z_1 + s' z_2)\, T_1(s, z_1, t_1)\, T_2(s', z_2, t_2) \exp i 2\pi \underline{K}_0 \cdot \underline{r}$$

where the wave vector $\underline{K}_0$ has $\vec{CO}$ as component parallel to the A_3 -plane.

2) The "doubly" diffracted wave, i.e. the incident wave is first diffracted by the first part (or first crystal) and then diffracted again by the second part (or second crystal).

At the interface, the amplitude of the wave diffracted by the first part is

$$S_1(s, z_1, t_1) \exp i\pi s z_1$$

For the diffraction of this wave considered as incident on the second part, N must be taken as the origin of the reciprocal space, the active reciprocal lattice point being N", defined by $\overrightarrow{NN''} = -\underline{n}'$ the corresponding Brillouin zone being (cc'). The wave vector $\underline{K}_0'$ of this wave has now $\overrightarrow{CN''} = \overrightarrow{CO} - \underline{\Delta n}$ as lateral component, i.e. $\underline{K}_0' = \underline{K}_0 - \underline{\Delta n}$

If the indident amplitude on the second part is 1, the amplitude of the diffracted beam at the back surface is $S_2(-s'', z_2, t_2) \exp i\pi s'' z_2$ with $s'' = n'\lambda x''$ (x" : distance from C to the Brillouin zone c'c). The doubly diffracted beam is then represented by

$$\exp i\pi(s z_1 + s'' z_2)\, S_1(s, z_1, t_1)\, S_2(-s'', z_2, t_2) \exp -i 2\pi \underline{\Delta n}\cdot\underline{r} \exp i 2\pi \underline{K}_0\cdot\underline{r}$$

The interference of these two waves yields a total transmitted wave with amplitude ψ_0 given by

$$\psi_0 = \exp i\pi(s z_1 + s' z_2)\, T_1(s, z_1, t_1)\, T_2(s', z_2, t_2)$$
$$+ \exp i\pi(s z_1 + s'' z_2)\, S_1(s, z_1, t_1)\, S_2(-s'', z_2, t_2) \exp -i 2\pi \underline{\Delta n}\cdot\underline{r} \qquad (43)$$

In a similar way the formulae (35) for S can be generalized. The diffracted wave arises by the superposition of two diffracted waves with slightly different wave vectors.

(i) the first is obtained if the incident wave is transmitted by the first component of the crystal sandwich, and afterwards diffracted by the second component.

The wave vector of this wave has a lateral component $\overrightarrow{CN'} = \overrightarrow{CN} + \underline{\Delta n}$, the wave vector itself then being $\underline{K}_n + \underline{\Delta n}$, with $\underline{K}_n = \underline{K}_0 + \underline{n}$

Its amplitude is :

$$T_1(s, z_1, t_1)\, S_2'(s', z_2, t_2) \exp i\pi(s z_1 + s' z_2)$$

(ii) The second wave arises if the incident wave is first diffracted by the first part of the crystal and then transmitted by the second part.
The wave vector is $\underline{K}_{\underline{n}}$, and the amplitude

$$S_1(s, z_1, t_1)\, T_2(-s'', z_2, t_2) \exp i\pi(s z_1 + s'' z_2)$$

The total diffracted wave has the amplitude

$$\psi_{\underline{n}} = \exp i\pi(s z_1 + s'' z_2)\, S_1(s, z_1, t_1)\, T_2(-s'', z_2, t_2)$$
$$+ \exp i\pi(s z_1 + s' z_2)\, T_1(s, z_1, t_1)\, S_2(s', z_2, t_2) \exp i 2\pi \underline{\Delta n}\cdot\underline{r} \qquad (44)$$

The formulae (43) and (44) for ψ_0 and $\psi_{\underline{n}}$ reduce as a matter of fact to the corresponding ones (20) and (35) (if the relations (42) are taken into account) obtained by using the column approximation, if

1°) $t_1 = t_2$, 2°) $s = s' = s''$, 3°) $\underline{\Delta n} = -\underline{k}$ ($\underline{\Delta n}\cdot\underline{r} = x/\Lambda$)

The column approximation could still be developed if the difference in extinction distance, absorptionlength and interference failure s are taken into account in the construction of system (3). However, the mathematical development becomes much more elaborate.

The changes of $\underline{k}$ into $-\underline{\Delta n}$ cannot be taken into account by an extension of the hypothesis underlying the column approximation.

In order to permit the discussion of the possible differences between the generalized result and the result obtained by the column approximation, we shall derive expressions for s, s' and s".

If $\overrightarrow{OC} = \underline{\rho}$, we have

$$s = n\lambda(\underline{\rho}\cdot\underline{n}/n - n/2) \qquad (45a)$$

$$s' = n'\lambda(\underline{\rho}\cdot\underline{n}'/n' - n'/2) \qquad (45b)$$

$$s'' = n'\lambda[(\bar{\rho} - \bar{n})\cdot\underline{n}'/n' - n'/2] \qquad (45c)$$

Let us apply these formulae first to the case of a rotation moiré pattern defined by its rotation angle ε . We can suppose that in the two-beam case Now n = n' and $\Delta n = 2\sin n\,\varepsilon/2\,n$

We have then

$$s - s' = -\lambda\,\underline{\rho}\cdot\underline{\Delta n} \simeq -\lambda\,\frac{\underline{n}\cdot\underline{\Delta n}}{2} = \frac{\lambda}{4}\left(2\sin n\frac{\varepsilon}{2}n\right)^2 = \frac{\lambda}{4\Lambda^2}$$

if (9) is taken into account.

Even for a moiré with interspacing Λ only 10 Å (close to the resolving power of the electron microscope) s-s' is of the order 10^{-4}. This means that $(s-s')t_0$ should be only a few times 10^{-2}. From the discussion of the variation of the intensities I_T and I_S as a function of st_0, we can conclude that this difference in s can be neglected.

In a similar way we obtain

$$s-s'' = \left(\frac{\underline{\rho}\cdot\underline{n} + \underline{n}\cdot\underline{n}' - \underline{\rho}\cdot\underline{n}'}{n} - n\right) n\lambda = \frac{\lambda\, \underline{n}\cdot\underline{\Delta n}}{2} = -\frac{\lambda}{4\Lambda^2}$$

and it is seen that also this s-difference can be neglected.

For the compression moiré defined by $\varepsilon = \Delta n/n$ the situation is not so favourable. We have now, since $\underline{n}$ and $\underline{n}'$ are parallel and $\Delta n = 1/\Lambda$

$$s-s' = \lambda\left\{n\left(\frac{\underline{\rho}\cdot\underline{n}}{n} - \frac{n}{2}\right) - n'\left(\frac{\underline{\rho}\cdot\underline{n}'}{n'} - \frac{n'}{2}\right)\right\} = \frac{\lambda n}{2}\Delta n = \frac{\lambda n}{2}\frac{1}{\Lambda}$$

and in the same way we obtain

$$s-s'' = -\frac{\lambda n}{2}\frac{1}{\Lambda}$$

If $\Lambda > 100$ Å these differences result in differences for st_0 less than a few times 10^{-2} and can again be neglected. For still more closely spaced moiré fringes, these s-differences become more important and should influence quantitatively the result. We believe, however, that the qualitative behaviour described in § 5 remains valid, the correction being probably that the maximum values of the contrast are somewhat different and occur for somewhat other values of s than predicted if these differences are not taken into account. The closely spaced moiré pattern occurs however mostly for two superposed crystals of different material, for which the difference in extinction distance (and eventually the absorption length) plays a more important role. This will be discussed below.

Following the column approximation, the geometry of the moiré pattern is given by the vector $\underline{k}$. We know now that the correct vector is $-\underline{\Delta n}$. It has already been proved that both vectors are approximately equal. We shall derive an expression for the error $\underline{\Gamma} = -\underline{\Delta n} - \underline{k}$ for the rotation and for the compression moiré. If (7b) is taken into account, one gets from (6) that

$$\underline{k}\cdot\underline{a}_i = -\underline{a}_i\cdot\underline{\Delta n} - \underline{\Gamma}\cdot\underline{a}_i = \underline{n}\cdot\underline{\Delta a}_i = \left(\sum_{j=1}^{2} h_j \underline{A}_j\right)\cdot\underline{\Delta a}_i = -\left(\sum_{j=1}^{2} h_j \underline{\Delta A}_j\right)\cdot(\underline{a}_i + \underline{\Delta a}_i)$$

or

$$= -\underline{\Delta n}\cdot\underline{a}_i - \underline{\Delta a}_i\cdot\underline{\Delta n} \quad (i = 1, 2)$$

$$\underline{\Gamma}\cdot\underline{a}_i = \underline{\Delta a}_i\cdot\underline{\Delta n} \quad (i = 1, 2),$$

or

$$\underline{\Gamma} = \sum_{i=1}^{2}\left(\underline{\Delta a}_i\cdot\underline{\Delta n}\right)\underline{A}_i$$

For a rotation moiré

$\underline{\Delta a}_i \cdot \underline{\Delta n} = (2\sin\frac{\varepsilon}{2})^2 \underline{a}_i \cdot \underline{n} = h_i (2\sin\varepsilon/2)^2$, and as a consequence

$\Gamma = (2\sin\varepsilon/2)^2 \underline{n}$, whilst $\Delta n = 2\sin\varepsilon/2\, n$ (the angle formed by $\underline{\Delta n}$ and $\underline{n}$ is $90° - \frac{\varepsilon}{2} \simeq 90°$)

From this it follows, since $2\sin\varepsilon/2 < 0,1$, that neglecting $\underline{\Gamma}$ is unimportant. For a compression moiré $\underline{\Delta n} = \varepsilon \underline{n}$, and Γ will again be γn, where γ is of the order of magnitude of ε^2. We can conclude that it is unimportant to take $\underline{k}$ or $-\underline{\Delta n}$, the latter being the correct vector to use and also the easiest to deal with.

As a general conclusion, it can be stated for crystals of the same material that the column approximation reveals itself as a very good approximation, only perhaps for very closely spaced moiré of the compression or mixed type, a small quantitative correction might be necessary.

Let us now consider the systems formed by two superposed crystals of different crystalline material.

The contrast of the resulting moiré pattern is described, as discussed in § 5, by

$$C_{T(S)} = \left(C_1 C_2^{(\pm)}\right)^{1/2} \text{ with } C_1 = I_T^{(2)}(s)\, I_S^{(1)}(s) \text{ and } C_2^{(\pm)} = I_T^{(1)}(-s') \, I_S^{(2)}(-s'')$$

However, now differences in extinction distance (and possibly in the absorption length) must be taken into account. For larger values of s, $\sigma \simeq |s|$ i.e. the kinematical approximation applies, and the influence of t_0 should drop out. However, the examples discussed in § 5 suggest that the well contrasted moiré patterns occur for values of s too small for this approximation to be valid.

We believe that qualitatively the properties of the moiré pattern as discussed in § 5 are unaltered, since a strongly contrasted moiré pattern (C $\simeq$ 1) will still occur for crystal orientations s for which $I_T^{(1)}(s, z_1, t_1) \simeq I_S^{(1)}(s, z_1, t_1) \simeq \frac{1}{2}$ and at the same time $I_T^{(2)}(s', z_2, t_2) \simeq I_S^{(2)}(-s'', z_2, t_2) \simeq \frac{1}{2}$ (bright field moiré) ($s \simeq s' \simeq s''$ is a good approximation). The actual s-darkes for which these and subsidiary maximum contrasts occur will depend, of course, on the crystal thicknesses, but also on both extinction distances (and for thicker crystals on both absorption lengths).

A system formed by two crystals of the same crystalline material but superposed in such a way that two different reciprocal lattice points to which correspond different extinction distances, come close together, is obviously equivalent to the system formed by two crystals of a different kind.

It can be proved that if in formulae (43) and (44), S_1 and S_2 are defined by (20 f)(σ and τ real or complex), a constant term, i.e. which does not depend on crystal thicknesses and crystal orientation, should be added in the exponent to $2\pi \Delta\nu \, \tau = 2\pi x/\Lambda$. This term is zero if both components of the crystal sandwich are of the same substance and if $\bar{n}$ and $\bar{n}'$ correspond to the same reciprocal lattice point. Otherwise, the term is the eventual difference of the phase angles of the corresponding Fourier expansion coefficients of the electronic potential. . If different from zero, this constant term will only result in a change in origin for x and is consequently of no importance for our discussions.

§ 8 REMARKS

The principles of the theory developed in the preceding paragraph are the same as those introduced by Hashimoto et al (loc cit). The details of the mathematical description are however different. Hashimoto et al. consider in the first crystal four waves : two transmitted and two diffracted ones, whilst we describe only one transmitted and one diffracted beam. Each of these waves has an amplitude depending on z. This means that each wave is equivalent to a wave packet, consisting in the considered case of two waves only with slightly different wave vectors. When entering through the interface in the second crystal, each wave gives rise to four waves. The result is the presence of sixteen waves in the second component of the crystal sandwich. Our method now corresponds to describing from the onset each group of four waves as a single one with z-depending amplitudes. So, only four waves have to be considered in the second crystal, and no calculation is needed, as shown in the preceding paragraph, to express their amplitudes if the well-known expressions for the amplitude of the waves transmitted and diffracted by a perfect crystal are used.

Our treatment implies that surface-reflected waves may be neglected, so that the crystal waves are determined by the boundary conditions at the entrance surface, and are independant of those at the back surface. Only then may the boundary conditions of the interfaces of overlapping crystals be applied successively and independently. The validity of this approximation for high energy electrons in electron diffraction has been discussed by Bethe(1928), Thompson and Cochran (1939), Kato (1952), and Whelan and Hirsch (1957).

It is possible to bring the final results obtained by Hashimoto et al. into the form of formulae (22) and (36). This is achieved by algebraic calculations which are rather long, and will for that reason not be given here.

An important advantage of our method is that it would be much easier to describe quantitatively the moiré patterns resulting from more complicated situations.

Assume that there are two strongly diffracted beams. The method of Hashimoto et al. would then introduce the presence in the second crystal of thirty-six waves, against nine if our description is used, each with an amplitude which can be written down without calculations provided the amplitudes of the wave transmitted or diffracted by a perfect crystal are knwon for this "three-beam" case.

The generalization of our method to systems formed by the superposition of more than two crystals will not introduce major mathematical difficulties. For systems of three crystals, the method of Hashimoto should have to deal with sixty-four waves present in the crystal, against eight if our procedure is used, again with amplitudes which need no calculations.

Another interesting situation, easy to deal with by our method, arises if a stacking fault is present in one of the crystals.

These three generalizations will be examined inddetail in separate papers, to be published elsewhere.

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CAPTIONS FOR FIGURES

Fig. 1 : Decomposition of the vector difference $\underline{\Delta n}$ in a "rotation" and a "compression " component.

Fig. 2 : Variation of the contrast C_T of the bright and dark fringes of the bright field moiré pattern (anomalous absorption neglected) - z_1 and z_2 are the thicknesses of both components of the crystal sandwich - t_o : extinction distance

a) $z_1=z_2=1/3t_o$; b) $z_1=z_2=2/3t_o$; c) $z_1=z_2= 7/6t_o$; d) $z_1=z_2= 5/3t_o$.

Fig. 3 : Variation of the contrast C_T of the dark and the bright fringes of the bright field moiré pattern and the variation of the maximum intensity $I_T^{(max)}$ of the bright fringes of the bright field moiré pattern (anomalous absorption neglected) - z_1 and z_2 : thicknesses of both components of the crystal sandwich - t_o : extinction distance

a) C_T for $z_1 = 2/3t_o$, $z_2=7/6t_o$ b) C_T for $z_1=1/3t_o, z_2=5/3t_o$

c) $I_T^{(max)}$ for $z_1=2/3t_o, z_2=7/6t_o$ d) $I_T^{(max)}$ for $z_1=1/3t_o, z_2=5/3t_o$.

Fig. 4 : Dependence of the contrast C_T of the dark and bright fringes of the bright field moiré on the position of the interface between the two components of the crystal sandwich - z_1 and z_2 : thicknesses of both components of the crystal sandwich, t_o : extinction distance . τ_o : absorption length $\alpha = t_o/\tau_o = 0,1 - 1 = 0 - z_1 + z_2 = 5t_o = 1/2\,\tau_o$

broken line : anomalous absorption neglected,

solid line : anomalous absorption considered.

Fig. 5 : Variation of the contrast C_T of the dark and bright fringes of the bright field moiré pattern - z_1 and z_2 : the thicknesses of both components of the crystal sandwich - t_o : extinction distance (anomalous absorption neglected).

a) $z_1 = z_2 = t_o$ b) $z_1 = z_2 = 2t_o$

Fig. 6 : Variation of the contrast C_T of the dark and bright fringes of the bright field moiré pattern, and the variation of the maximum intensity $I_T^{(max)}$ of the bright fringes of the bright field moiré - z_1 and z_2 : thicknesses of both components of the crystal sandwich- t_o : extinction distance (anomalous absorption neglected).

a) C_T for $z_1=z_2=3t_o$ b) C_T for $z_1=t_o, z_2=3t_o$

c) $I_T^{(max)}$ for $z_1=t_o$, $z_2= 3t_o$.

Fig;7 : Variation of the contrast C_T(C_S) of the dark and bright fringes of the bright (dark) field moiré pattern and the variation of the maximum (minimum) intensity $I_T^{(max)}$ ($I_S^{(min)}$) of the bright (dark) fringes of the bright (dark) field moiré pattern - z_1 and z_2 : thicknesses of both components of the crystal sandwich - t_o : extinction distance - τ_o : absorption length; - $\alpha = t_o/\tau_o = 0,1$ - $z_1 = z_2 = t_o$

a) C_T b) $I_T^{(max)}$ c) C_S d) $I_S^{(min)}$

Fig. 8 : Variation of the contrast C_T (C_S) of the dark and bright fringes of of the bright (dark) field moiré pattern and the variation of the maximum (minimum) intensity $I_T^{(max)}$ ($I_S^{(min)}$) of the bright (dark) fringes of the bright (dark) field moiré pattern - z_1 and z_2 : thicknesses of both components of the crystal sandwich - t_o : extinction distance τ_o : absorption length - $\alpha = t_o/\tau_o = 0,1$ - $z_1 = z_2 = 2t_o$

a) C_T b) $I_T^{(max)}$ c) C_S d) $I_S^{(min)}$

Fig. 9 : Variation of the contrast C_T (C_S) of the dark and bright fringes of the bright (dark) field moiré pattern and the variation of the maximum (minimum) intensity $I_T^{(max)}$ ($I_S^{(min)}$) of the bright (dark) fringes of the bright (dark) field moiré pattern - z_1 and z_2 : thicknesses of both components of the crystal sandwich - t_o : extinction distancee - τ_o : absorption length - $\alpha = t_o/\tau_o = 0,1$ - $z_1 = z_2 = 3t_o$

a) C_T b) $I_T^{(max)}$ c) C_S d) $I_S^{(min)}$

Fig. 10 : Variation of the contrast C_T (C_S) of the dark and bright fringes of the bright (dark) field moiré pattern and the variation of the maximum (minimum) intensity $I_T^{(max)}$ ($I_S^{(min)}$) of the bright (dark) fringes of the bright (dark) field moiré pattern - z_1 and z_2: thicknesses of both components of the crystal sandwich - t_o : extinction distance - τ_o : absorption length - $\alpha = t_o/\tau_o = 0,1$ - $z_1 = t_o$, $z_2 = 3t_o$

a) C_T b) $I_T^{(max)}$ c) C_S d) $I_S^{(min)}$

Fig. 11 : The A_3 plane of the reciprocal lattice showing the components of the wave vectors responsible for the moiré pattern occurence.

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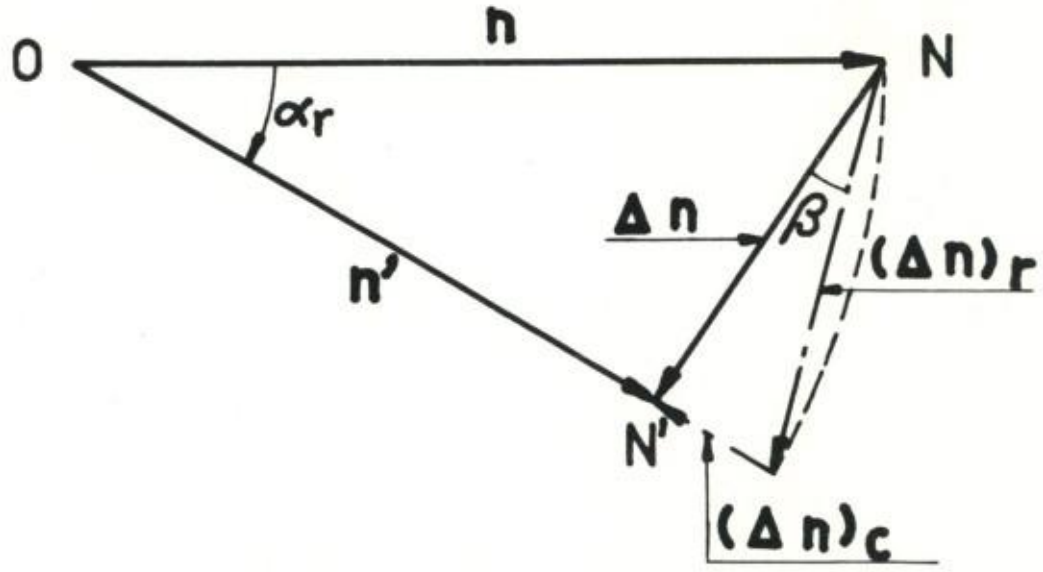


fig. 1

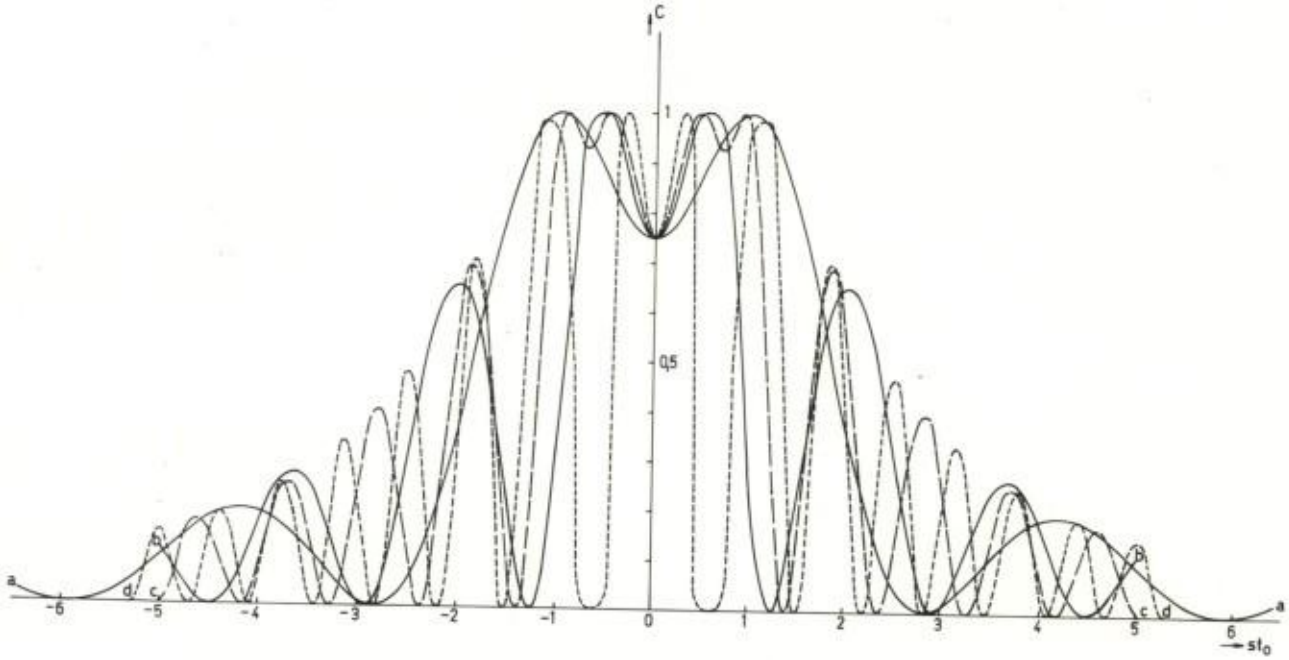
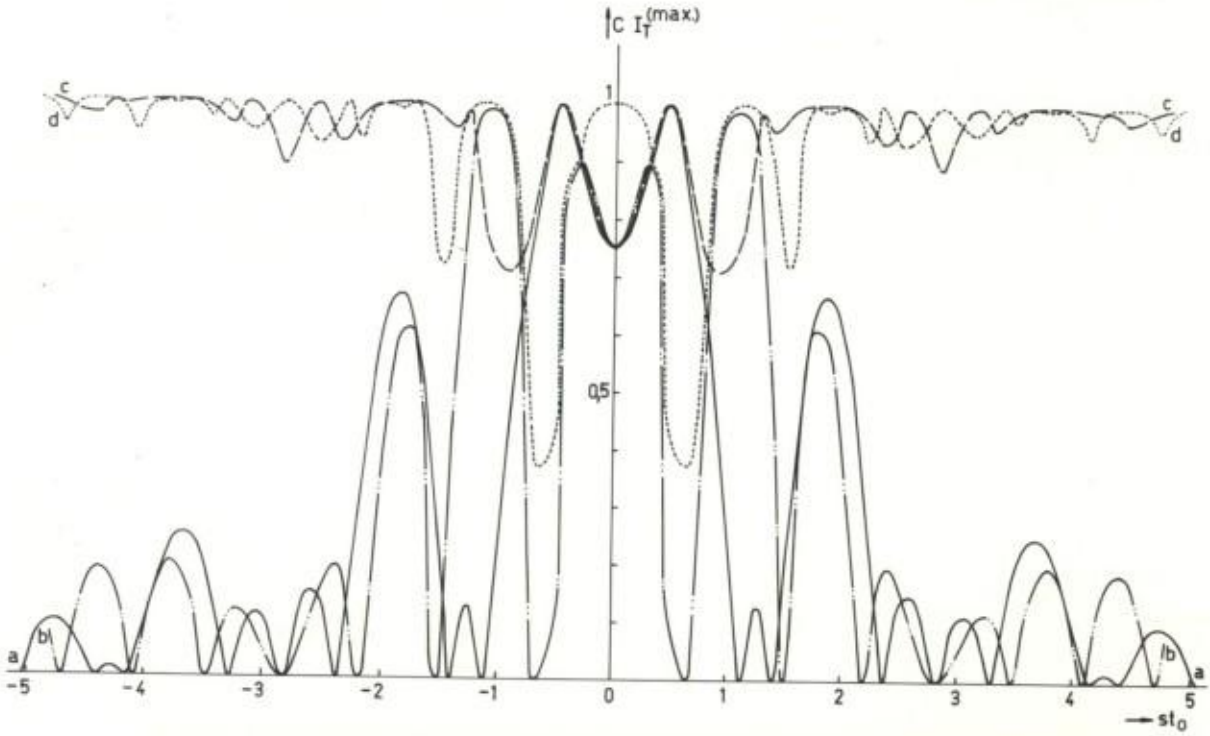


Fig. 2



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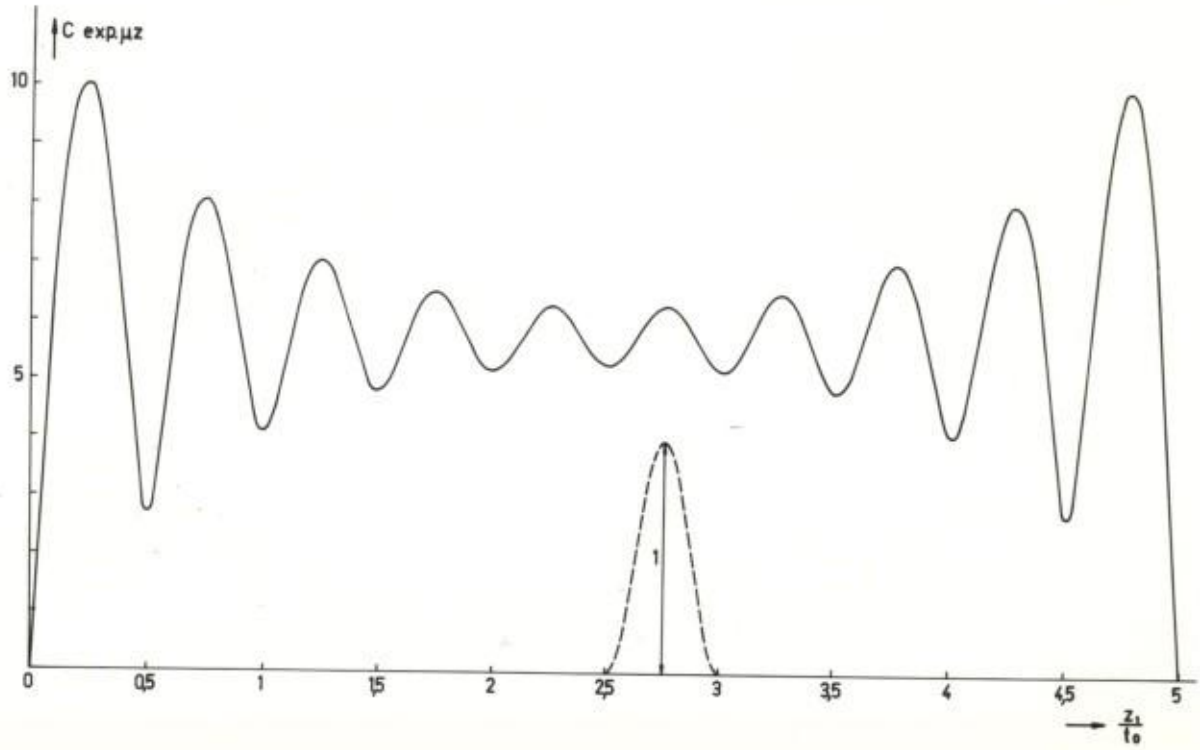


Fig. 4

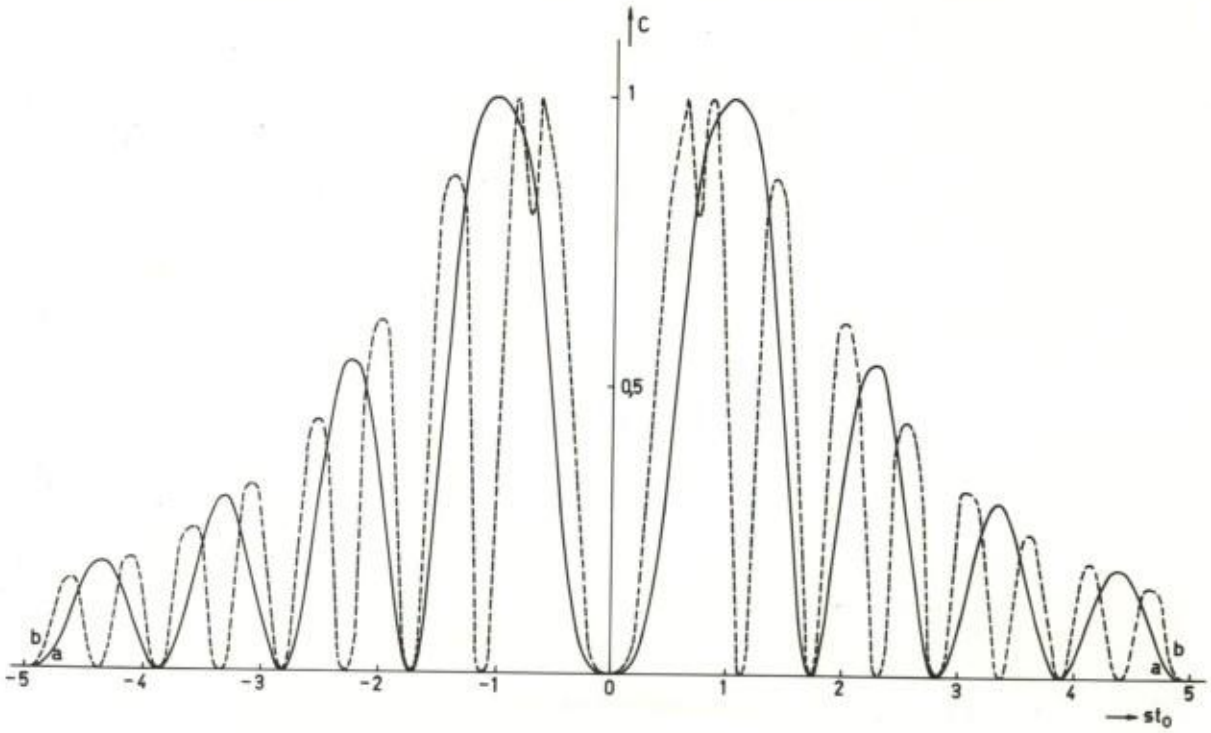


fig. 5

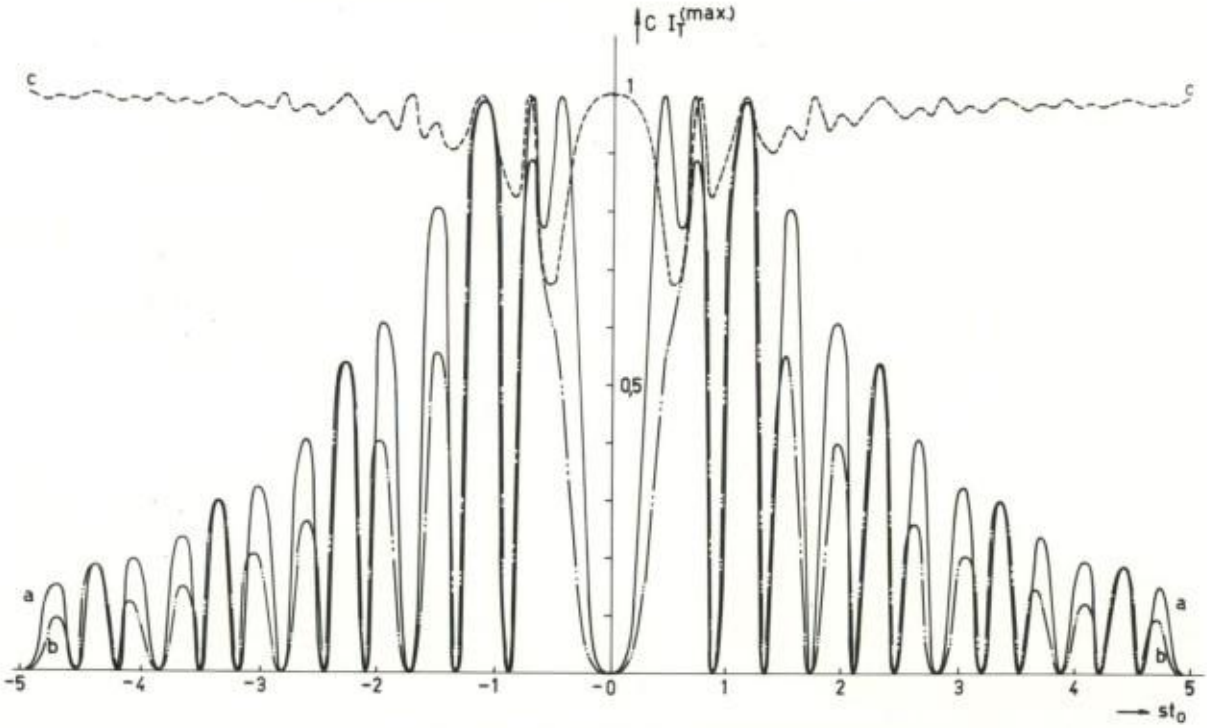


fig. 6

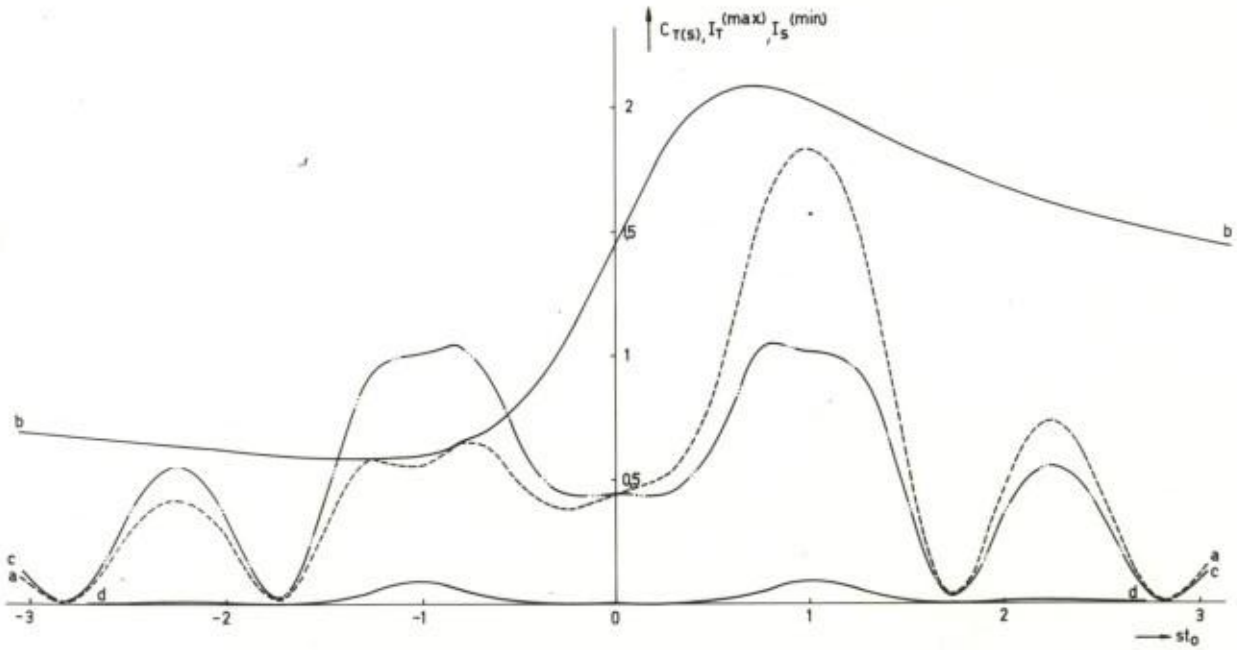


fig. 7

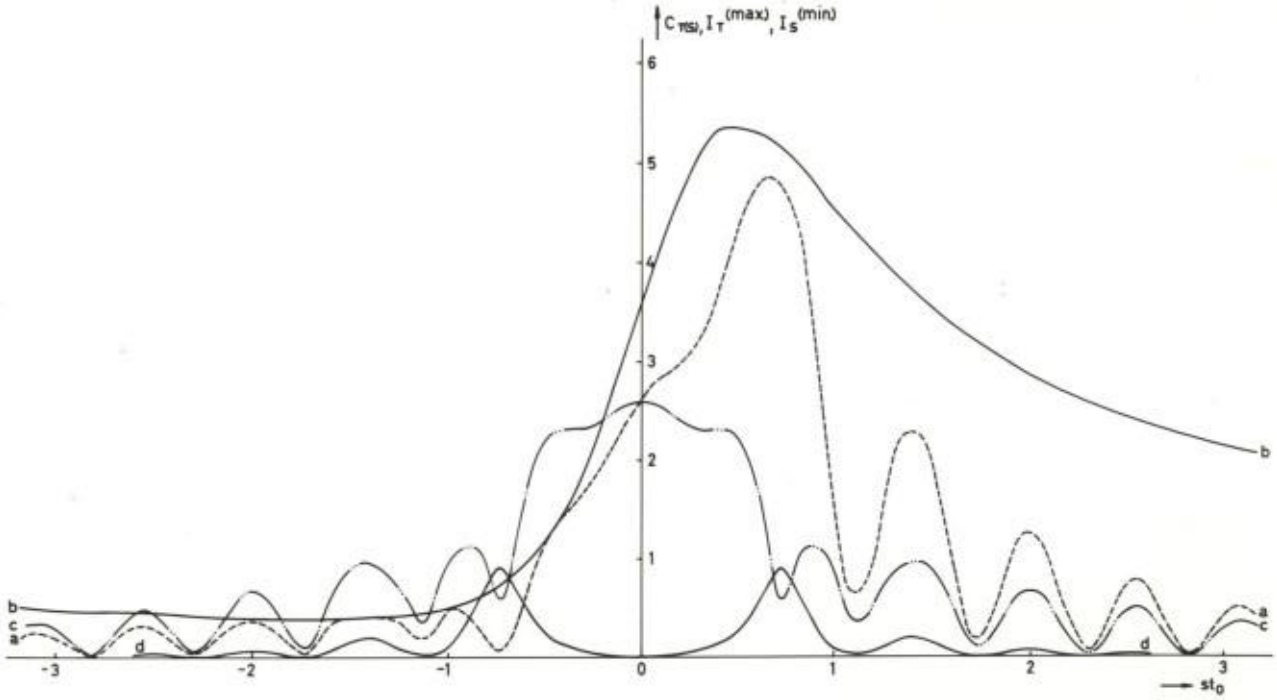
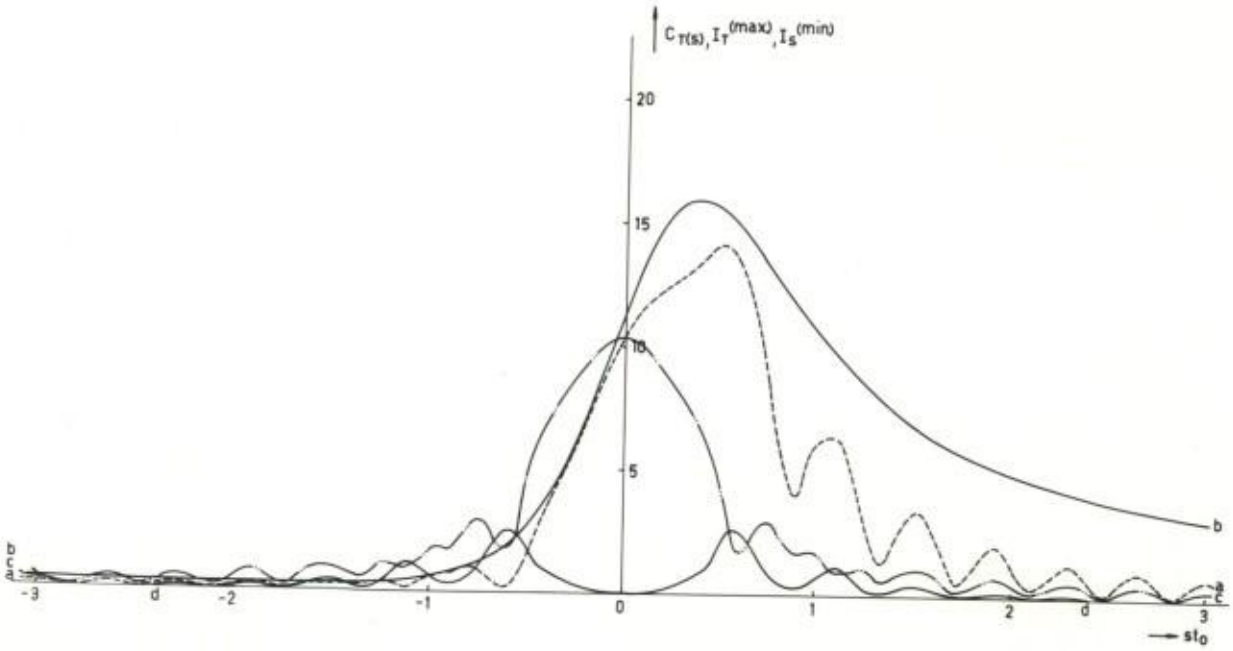


fig 8



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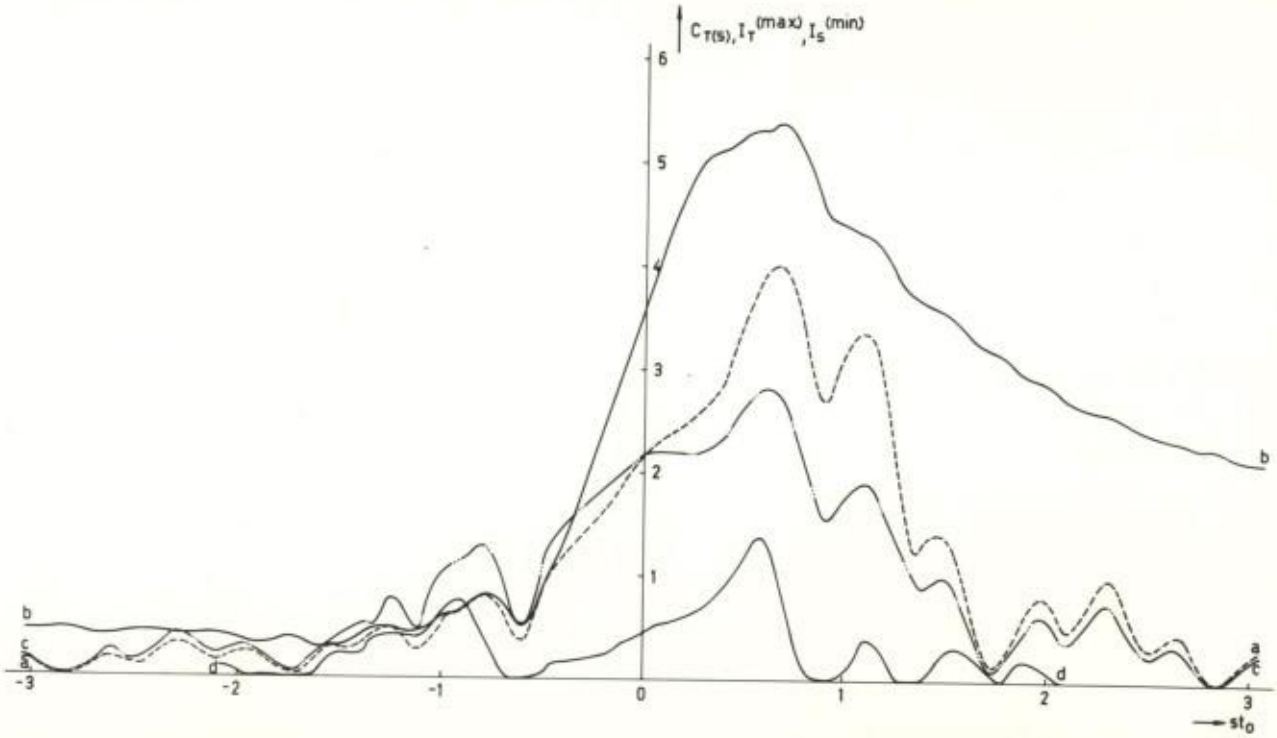

fig. 10

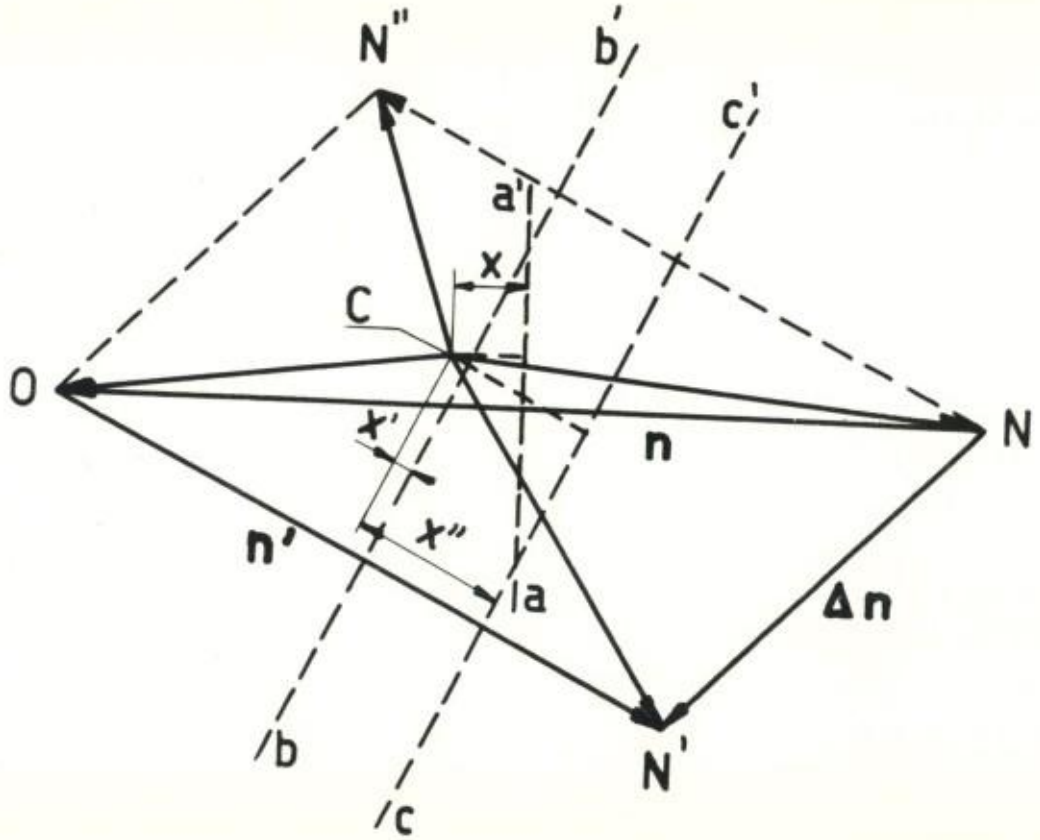

fig 11