Acta Cryst. (1975). A31, 836

Study of the New Wave Fields Appearing in Crystals Deformed by Large Thermal Gradients

BY F. BALIBAR, Y. EPELBOIN AND C. MALGRANGE

Laboratoire de Minéralogie–Cristallographie associé au CNRS, Université Pierre et Marie Curie, 4 *place Jussieu,* 75230 *Paris Cedex* 05, *France*

(*Received* 20 *March* 1975; *accepted* 30 *May* 1975)

The criterion for the validity of geometrical optics is quantitatively examined through a 'computer experiment' in the specific case of a crystal containing substantial deformations induced by a large thermal gradient. It is shown that the above-mentioned criterion is numerically correct. For larger deformations than those corresponding to the criterion the computed intensity shows that two wave fields appear where only one would normally have been expected, in good agreement with theoretical predictions. The intensity of the new wave field has been plotted as a function of deformation.

I. Introduction

The Ewald–Laue dynamical treatment of X-ray propagation in a triply periodic medium provides a complete understanding of how an X-ray wave propagates in a perfect crystal. The main concern of people working in the field at present is to give a correct account of the propagation of X-ray waves in crystals exhibiting defects of periodicity. For small deformations (slightly curved reflecting planes, for instance) different authors have shown that the Ewald–Laue theory can be extended (Penning & Polder, 1961; Kato, 1963); it is still possible to define ray trajectories for the energy propagation, the only difference from the perfect crystal case being that these trajectories are curved – they 'adjust' themselves to the curvature of the reflecting planes just as ordinary light rays 'adjust' themselves when they encounter a variation of index of refraction in the so-called geometrical optics treatment.

There is experimental evidence that this extension of the theory for small deformations no longer holds in the case of large deformations.

In the immediate vicinity of a dislocation line, for instance, the fine structure of images obtained by topography can no longer be interpreted along the lines of the previous extensions of the Ewald–Laue treatment, and one has to admit that, in the strongly-deformed regions, one wave field gives rise to two wave fields. This phenomenon is usually referred to as the creation of new wave fields in highly distorted regions.

In order to account for this, one needs to do more than merely to make an adjustment in the perfect-crystal treatment. Takagi (1962, 1969), and Taupin (1964) have produced more general theories which include the crystal deformations from the start. If one then calculates the intensity distribution on the exit surface of a crystal containing large deformations (a dislocation line) on the basis of such theories, one verifies that there is good agreement between the calculated intensity distribution and the experimental one (Balibar & Authier, 1967). One of us (Balibar, 1969) has shown that the difference between those theories and Penning's and Kato's is the same as the difference which exists between geometrical optics and 'wave optics'. This means that Takagi's and Taupin's theories take into account diffraction effects (*i.e.* those effects occuring in wave propagation when the propagation conditions change abruptly), while the other theories do not. In other words, we may say that Takagi's and Taupin's theories are general enough to contain Huyghens' principle. According to this principle, crystal deformations may be viewed as resulting in secondary sources excited by each incoming wave field and reemitting two wave fields inside the crystal. Therefore starting with one wave field, one is bound to obtain two wave fields while going through a region of strong distortions (Authier & Balibar, 1970). The so-called creation of new wave fields is the specific form that diffraction takes in the case of X-ray propagation: it will only occur when the deformations are large enough for diffraction effects to be observable. And it is possible to give a criterion (analogous to the well-known criterion for the validity of geometrical optics with ordinary light) for the appearance of such effects.

The purpose of this paper is to study the appearance of new wave fields with increasing deformation, for a specific kind of deformation (a temperature gradient), by means of computer experiments and to show that the previously given criterion is quantitatively exact.

II. Takagi's equations and the criterion for geometrical optics

According to Takagi's (1962, 1969) theory, the crystal wave in a distorted crystal may be expressed as a sum of Bloch waves:

$$D(\mathbf{r})=D_0'(\mathbf{r})\exp(-2\pi i\mathbf{k}_o\,.\,\mathbf{r})+D_h'(\mathbf{r})\exp(-2\pi i\mathbf{k}_h\,.\,\mathbf{r})\,,$$

$\mathbf{k}_0$ and $\mathbf{k}_h$ being two vectors of modulus $k=n/\lambda$ (where n is the index of refraction and λ the vacuum wavelength) and such that $\mathbf{k}_h=\mathbf{k}_0+\mathbf{h}$ (where $\mathbf{h}$ is the reciprocal-lattice vector characteristic of the considered reflexion); $\mathbf{k}_0$ can be chosen at will. Nevertheless we shall choose $\mathbf{k}_0$ so that it has the same tangential component as the incident wave vector.

$D_0'(\mathbf{r})$ and $D_h'(\mathbf{r})$ are then solutions of a system of linear partial differential equations:

$$\frac{\partial D_0'}{\partial s_0} = -i\pi k C \chi_{\bar{h}} D_h' \quad (1a)$$

$$\frac{\partial D_h'}{\partial s_h} = -i\pi k C \chi_h D_0' + 2i\pi k \beta_h' D_h', \quad (1b)$$

which is equivalent to the two second-order partial differential equations:

$$\frac{\partial^2}{\partial s_0 \partial s_h} D_0'(\mathbf{r}) - 2\pi i k \beta_h' \frac{\partial}{\partial s_0} D_0'(\mathbf{r}) + \pi^2 k^2 C^2 \chi_h \chi_{\bar{h}} D_0'(\mathbf{r}) = 0 \quad (2a)$$

$$\frac{\partial^2}{\partial s_0 \partial s_h} D_h'(\mathbf{r}) - 2\pi i k \beta_h' \frac{\partial}{\partial s_0} D_h'(\mathbf{r}) + (\pi^2 k^2 C^2 \chi_h \chi_{\bar{h}} - 2\pi i k \frac{\partial}{\partial s_0} \beta_h') D_h'(\mathbf{r}) = 0 \quad (2b)$$

where s_0 and s_h are coordinates along the refracted and reflected directions respectively; $\chi_h, \chi_{\bar{h}}$ are the Fourier coefficients of the dielectric susceptibility χ for a perfect crystal of the type under consideration; and $C = 1$ or $\cos 2\theta$ is the polarization factor.

The crystal deformation appears in equation (2) through the coefficient $\beta_h'(\mathbf{r})$

$$\beta_h'(\mathbf{r}) = \beta_h - \frac{1}{k} \frac{\partial}{\partial s_h} [\mathbf{h} \cdot \mathbf{u}(\mathbf{r})] \quad (3)$$

where $\mathbf{u}(\mathbf{r})$ is the displacement of an atom located at $\mathbf{r}$. In this equation, β_h is a constant coefficient, the value of which depends on the initial choice for $\mathbf{k}_0$. With our choice of $\mathbf{k}_0$

$$\beta_h = -\left[\Delta\theta \sin 2\theta - \frac{1}{2} \chi_0 \left(\frac{\gamma_0}{\gamma_h} - 1\right)\right], \quad (4)$$

$\Delta\theta$ being the departure of the incident wave from the exact Bragg law and

$$\gamma_0 = \cos(\mathbf{n}, \mathbf{s}_0)$$
$$\gamma_h = \cos(\mathbf{n}, \mathbf{s}_h)$$

(where $\mathbf{n}$ is normal to the crystal entrance surface).

Any partial differential equation such as (2*a*) or (2*b*) cannot be solved unless some appropriate initial conditions are given. These are provided by the physics of the problem; *i.e.*, in this case, the amplitude distribution of the incident wave on the enentrance surface and the gradient of this amplitude (Cauchy conditions). Examination of (2) makes it clear that the solutions of such equations will depend on $\beta_h'(\mathbf{r})$ and $\partial\beta_h'(\mathbf{r})/\partial s_0$.

It has been shown elsewhere (Authier & Balibar, 1970) that geometrical optics (*i.e.* the neglect of diffraction effects) is a valid approximation as long as

$$\left| -2\pi i k \frac{\partial}{\partial s_0} \beta_h' \right| \ll \pi^2 k^2 C^2 \chi_h \chi_{\bar{h}} \quad (5)$$

in the last term of equation (2*b*). The Penning & Polder results are then recovered – the energy is propagated in beams with curved trajectories. To each wave field there corresponds a trajectory and a given wave field can be followed throughout the crystal.

On the other hand, when condition (5) is not fulfilled, integration of (2) becomes more complicated and does not lead to the Penning & Polder (or Kato) results. It is then necessary to use the mathematical theory of distributions. As already hinted in the introduction, deformations can then be treated mathematically as equivalent distributions of ideal sources which are responsible for the creation of new wave fields. Therefore a wave field of type 1 gives rise to two wave fields, of types 1 and 2 respectively. Condition (5) gives the criterion for the validity of the geometrical optics; it states that the rate of change of β_h' or, as can be seen [from (3)], the second derivative of the component of the displacement $\mathbf{u}(\mathbf{r})$ must be less than a certain constant value, a value essentially determined by the reflexion considered.

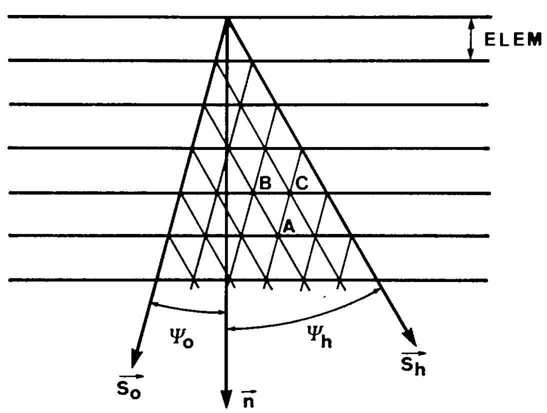

Fig. 1. Principle of the computation. The amplitudes of the wave fields at the point A depend on their values at points B and C.

III. Computer experiments

Takagi's equations, in the form of equation (1), can be numerically integrated on a computer. For this, we have used a new version of a program written, some years ago, by Authier, Malgrange & Tournarie (1968). The general technique of integration is described at length in the above reference. Suffice it to say here that the crystal is divided into elementary slabs parallel to the entrance surface and that the values of D_0' and D_h' at a given point A are obtained from their values at B and C, two points belonging to the preceeding slab and such that BA and CA are parallel to $\mathbf{s}_0$ and $\mathbf{s}_h$ respectively (Fig. 1):

$$\begin{bmatrix} D_0'(A) \\ D_h'(A) \end{bmatrix} = M \begin{bmatrix} D_0'(B) \\ D_h'(B) \\ D_0'(C) \\ D_h'(C) \end{bmatrix},$$

where M is a 2×4 matrix with coefficients depending on the nature of the crystal and its local deformations.

The deformations that have been considered in this work are those induced by a constant thermal gradient parallel to the entrance surface. This gradient produces a curvature of the atomic planes perpendicular to the entrance surface. These planes are chosen as reflecting planes; therefore the reflexion is symmetric.

As already mentioned [*cf.* equation (3)], the important parameter for our purpose is the rate of change of β_h'. To facilitate comparison with other studies, we shall use here the same parameter as that used by Penning,

$$\beta=-\frac{\lambda}{C\sqrt{\chi_h\chi_{\bar{h}}}}\frac{\partial^2}{\partial s_0\partial s_h}(\mathbf{h}\,.\,\mathbf{u})\,. \tag{6}$$

This β parameter is of opposite sign to that of Kato. This should not be confused with the β_h' appearing in Takagi's notations. These quantities are related through

$$\frac{\partial\beta_h'}{\partial s_0}=\beta C\sqrt{\chi_h\chi_{\bar{h}}}\cos\theta\,. \tag{7}$$

In our case (the symmetrical lane case), β reduces to

$$\beta=\frac{2\,\mathrm{tg}\,\theta}{\chi_h}[\mathbf{n}\,.\,\nabla_r\alpha T] \tag{8}$$

where $\mathbf{n}$ is normal to the entrance surface and α is the expansion coefficient.

(*a*) *Small temperature gradient*: *ray trajectories*

Let us first recall some theoretical results concerning the ray trajectories in the case of a small temperature gradient. Penning & Polder have shown, and this has been checked by previous computer experiments (Authier *et al.*, 1968), that for small values of β and for a symmetric case ($\gamma_0=\gamma_h$), the trajectories of the wave fields induced in the crystal by an incident vacuum plane wave are portions of hyperbolas.

Let x and z be two coordinates parallel and perpendicular respectively to the entrance surface, the origin being located at the intersection of the incident beam and the crystal entrance surface. The equation of the trajectories is then:

$$\left(\frac{\beta x}{\mathrm{tg}\,\theta}\pm\sqrt{1+\eta_i^2}\right)^2-(\beta z-\eta_i)^2=1$$

where η_i is a parameter which is related to the departure of the incident vacuum plane wave $(\Delta\theta)_i$ from Bragg's law:

$$\eta_i=\frac{(\Delta\theta)_i\sin 2\theta}{\sqrt{\chi_h\chi_{\bar{h}}}}\,. \tag{9}$$

The + and − signs correspond to wave fields 1 and 2 respectively. For different values of β, these curves form a set of hyperbolas with asymptotes parallel to $\mathbf{s}_0$ and $\mathbf{s}_h$; the curvature of each hyperbola at its top is determined by the value of β and increases with increasing β.

For a given value of β, any point on the corresponding hyperbola is characterized by the value of the parameter

$$\eta=\frac{\Delta\theta\sin 2\theta}{\sqrt{\chi_h\chi_{\bar{h}}}}\,,$$

where $\Delta\theta$ represents the departure of the considered wave field from the Bragg angle at the point considered. The parameter η is related to η_i, β and z (depth inside the crystal as measured from the entrance surface) through

$$\eta=\eta_i-\beta z\,. \tag{10}$$

The apex of the hyperbola corresponds to $\eta=0$. Let us note that, for a given β, the value of η_i determines which portion of the corresponding hyperbola is the wave-field trajectory (Fig. 2).

(*b*) *Large temperature gradient*: *creation of new wave fields*

For sufficiently large values of $\partial\beta_h'/\partial s_0$ [*i.e.* for large values of the parameter β in (8)], diffraction effects ap-

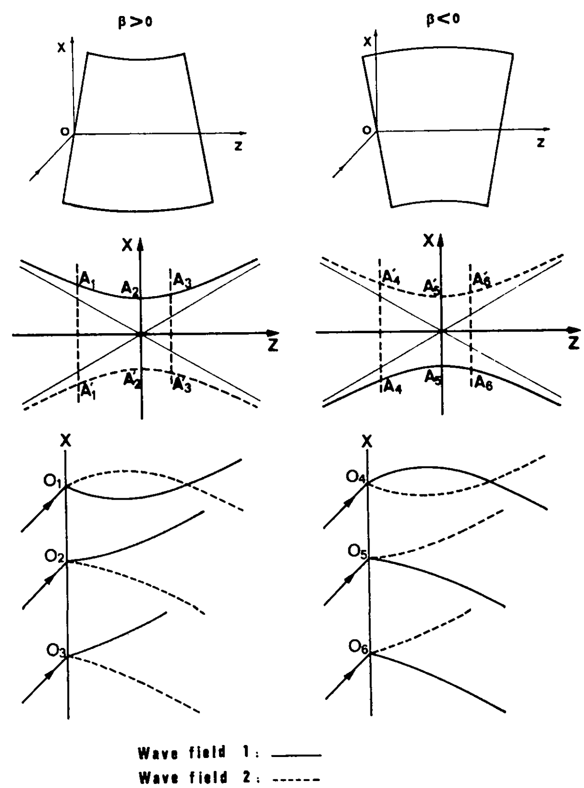

Fig. 2. Trajectories of the wave fields in a crystal slightly deformed by a thermal gradient. The different paths depend on the values of the deformation and on the departure of the incident wave at the entrance surface from the Bragg angle.

pear. From (5) and (7), we may conclude that this is bound to occur for

$$\beta > \pi/2\Lambda \tag{11}$$

where the Pendellösung wavelength Λ equals

$$\lambda \cos \theta / C \sqrt{\chi_h \chi_{\bar{h}}} \ .$$

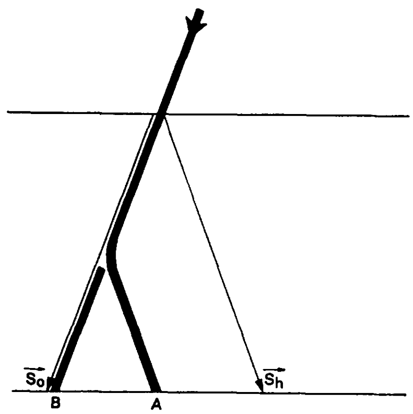

Fig. 3. Trajectories of the beams for a large deformation, where B is the new wave field and A the curved wave field.

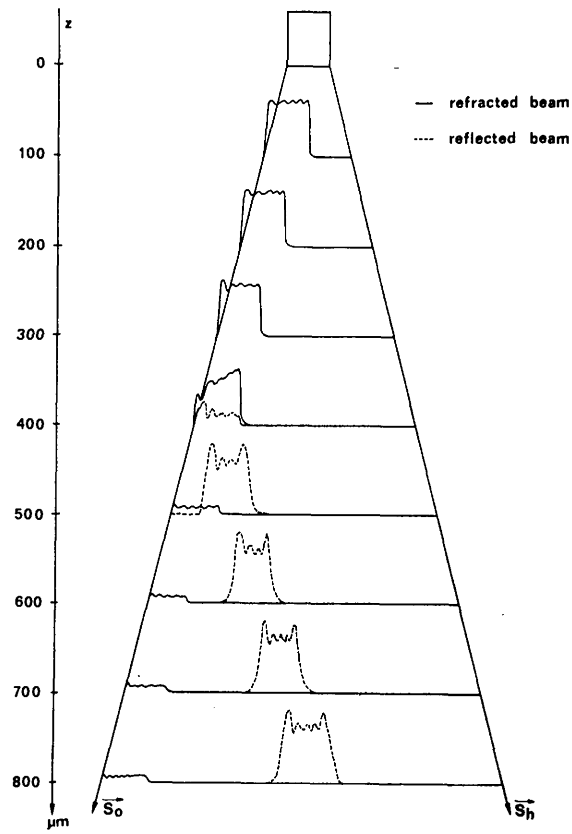

Fig. 4. Calculated intensity at different depths inside the crystal corresponding to: $\beta=20$ and $\eta_i=80$.

In our computer experiments we used $\lambda=0{\cdot}709$ Å, $\theta=10°\ 40'$, $C=1$ and $|\chi_h|=2\ .\ 10^{-6}$. These values correspond to reflexion 220 for a silicon crystal with Mo $K\alpha$ radiation. The corresponding value of Λ is 34·9 μm. We then expect to find the creation of new wave fields. In order to make this phenomenon most evident, we select *one* of the two 'normal' wave fields induced at the entrance surface by a vacuum plane wave characterized by a given value of η_i [by 'normal' we mean those wave fields which, for the η_i value considered, would propagate according to geometrical optics; in this case they would correspond to the hyperbolic trajectories (9)]. This is most easily achieved by choosing a large value of η_i, since consideration of the intensity formulae, as given by Malgrange (1967), shows that for such an η_i the intensity of one of the wave fields is nearly equal to zero.

Evidence of the creation of new wave fields for large values of β will therefore correspond to the observation of two wave fields at the exit surface while for small values of β we would observe only one wave field (Fig. 3).

As a matter of fact, closer examination of criterion (5) shows that diffraction effects are more important in the regions where the curvature of the 'normal' trajectory is large. In our case, the creation of new wave fields is expected to be most important when the apex of the hyperbola which represents the trajectory of the 'normal' wave field under consideration lies *inside* the crystal. This apex corresponds to a zero value of the parameter $\eta=\eta_i-\beta z$. Therefore for a given value of β, choosing η_i such that the apex of the hyperbola be midway between the entrance and the exit surface, represents a good compromise between the two conditions imposed on η_i.

IV. Results

Fig. 4 shows the results obtained for $\beta=20$ per 100μm and $\eta_i=80$. Under these conditions $\eta=0$ at a depth equal to 400 μm. Geometrical-optics results would give a curved wave field which, far from its apex, reduces to a reflected wave only. Fig. 4 shows that a new wave field appears in the zone where η is nearly equal to zero. As soon as we leave this particular zone, this wave field reduces to the refracted wave only.

In Fig. 5 we have plotted the relative intensity of the newly created wave field as a function of β, expressed in unit (100 μm)$^{-1}$. The intensity of the extra wave field equals 10% of the incident intensity as soon as $\beta=12{\cdot}5$ that is, for $\beta\simeq 3(\pi/2\Lambda)$ [see equation (11)]. From this we may conclude that criterion (5) (which was established on theoretical basis) is not only qualitatively correct but quantitatively exact.

Fig. 6 is a plot of log I_2/I_0 (where I_2 is the intensity of the newly created wave field and I_0 is the intensity of the incoming beam) as a function of $1/\beta$. This curve is a straight line with a slope of $-t$, which can be evaluated as $t\simeq 29$ [with a choice of unit of (100 μm)$^{-1}$].

We conclude that I_2 varies exponentially with $1/\beta$:

$$\frac{I_2}{I_0} = \exp(-t/\beta).$$

This conclusion can be related to some of Penning's (1966) theoretical results. Penning has shown that, in the specific case of a local reciprocal-lattice vector depending on only one coordinate z (depth in the crystal), the appearance of a new wave field is to be expected when the crystal deformations become very large. Penning also stated that this new wave field should appear in the region where the curvature of the ray trajectory is at its maximum and that, far from this region, the new wave field should be made up of only a transmitted wave of intensity I_2 such that $I_2/I_0 = \exp(-2S)$, where S is a parameter inversely proportional to our β.

Here we have the same kind of dependence of the intensity of the new wave field on deformation. Note that our result is more general than Penning's, since the local reciprocal-lattice vector depends not only on z but also on x. This exponential dependence of I_2 can be easily understood with reference to the results of the kinematical theory. (It is clear that for very large deformations the two theories should give the same results. As a matter of fact, for very large values of β [*e.g.* here for $\beta = 150$ units $(100\ \mu\text{m})^{-1}$]

$$I_2/I_0 = 1 - t/\beta,$$

which shows that the normal wave field intensity I_1 is equal to t/β. Since we have shown experimentally that this wave field is a reflected wave only, this means that the intensity of the reflected beam is inversely proportional to β. This result can be easily understood. The reflected intensity is kept at a non-vanishing level only for departures from the Bragg law of the order of the width of the rocking curve, which means $\Delta\eta \simeq 2$, corresponding [see (10)] to a depth Δz in the crystal of order $\simeq 2/\beta$. Owing to the strong curvature of the wave fields, the corresponding wave packet is made up of a large spectrum of wave vectors and therefore of η values. The intensity obtained is therefore an integrated intensity, which is, in the kinematical case, proportional to the volume where the reflexion occurs, that is in our case to $\Delta z \simeq 2/\beta$. The kinematical reflecting power is thus inversely proportional to β and the kinematical results are thus retrieved for large values of β.

From this result, we may infer that the direct image of a crystal imperfection, which is often interpreted as a kinematical effect, can in fact be taken into account through Takagi's equation.

As a conclusion we may say that the results presented here establish a bridge between the kinematical and the dynamical theories.

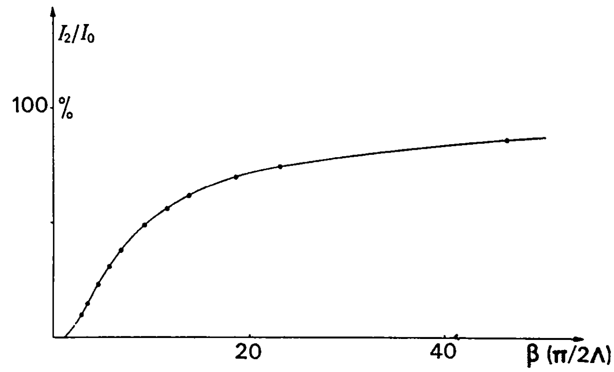

Fig. 5. Ratio of the intensity of the new wave field to the incident intensity as a function of β.

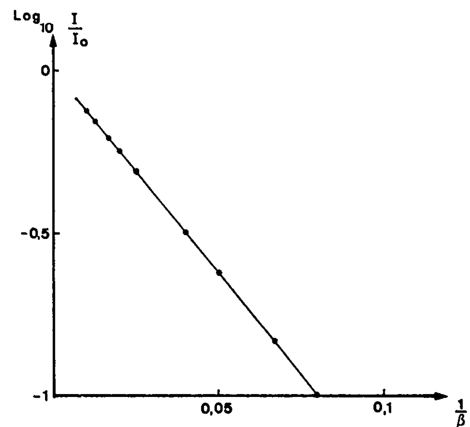

Fig. 6. Logarithm of the ratio of the new wave field to the incident intensity as a function of $1/\beta$ (100 μm).

References

AUTHIER, A. & BALIBAR, F. (1970). *Acta Cryst.* **A26**, 647–654.

AUTHIER, A., MALGRANGE, C. & TOURNARIE, M. (1968). *Acta Cryst.* **A24**, 126–136.

BALIBAR, F. (1969). *Acta Cryst.* **A25**, 650–658.

BALIBAR, F. & AUTHIER, A. (1967). *Phys. Stat. Sol.* **21**, 413–422.

KATO, N. (1963). *J. Phys. Soc. Japan*, **18**, 1785–1791.

MALGRANGE, C. (1967). *Etude de la Propagation des Rayons X dans des Cristaux Parfaits ou Légèrement Déformés.* Thèse d'Etat, Univ. de Paris.

PENNING, P. (1966). *Theory of X-ray diffraction in Unstrained and Lightly Strained Perfect crystals.* Thesis, Univ. of Delft.

PENNING, P. & POLDER, D. (1961). *Philips Res. Rep.* **16**, 419–440.

TAKAGI, S. (1962). *Acta Cryst.* **23**, 23–25.

TAKAGI, S. (1969). *J. Phys. Soc. Japan*, **26**, 1239–1253.

TAUPIN, D. (1964). *Bull. Soc. Fr. Minér. Crist.* **87**, 469–511.