

### General remarks

Statistical distribution of the twins, carried out on about thirty crystals from the same batch, has shown that twinning I is by far the most frequent of the three types; as is seen from X-ray intensities, crystals twinned according to this law have approximately equal volumes of the two components, thus suggesting the occurrence of multiple twinning; they moreover do not, in general, show either of the other two types of twinning: in the few cases where an individual twinned according to II or III is present, its volume is remarkably small. Twinning III is the least frequent and it generally occurs in those crystals which are also twinned according to II. A typical example of twinning II and III in the same crystal, where the volumes of the individuals happen to be approximately equal, is shown in Fig. 8.

At the boundary layers the molecules are packed in such a way that CH bonds are directed through oxygen

and nitrogen atoms of adjacent molecules; this feature is present in the crystal structures of this compound and of the other investigated biisoxazole isomers (Cannas & Marongiu, 1967; Biagini, Cannas & Marongiu, 1969); from the values of intermolecular contacts at the boundary layers, which are very close to those found in the mentioned crystal structures, it could be inferred that the energy at the twin boundaries should not be much different from that of the regular structure.

This work was supported by the Italian Consiglio Nazionale delle Ricerche.

### References

- BIAGINI, S., CANNAS, M. & MARONGIU, G. (1969). *Acta Cryst.* **B25**, 730.  
 CANNAS, M. & MARONGIU, G. (1967). *Z. Kristallogr.* **124**, 143.  
 CANNAS, M. & MARONGIU, G. (1968). *Z. Kristallogr.* **127**, 388.

*Acta Cryst.* (1972). **A28**, 501

## Polarization Phenomena in X-ray Diffraction

BY P. SKALICKY\* AND C. MALGRANGE

*Laboratoire Minéralogie Cristallographie, Université Paris VI, 9 quai St Bernard, Tour 16, Paris Vème, France*

(Received 11 November 1971 and in revised form 16 June 1972)

Because of the dielectric properties of most crystals in the X-ray frequency region, polarization phenomena such as birefringence and optical activity are not found with X-rays in simple transmission. It can however be deduced from the dynamical theory that such effects occur in crystal diffraction. An experiment is described which proves that under certain conditions all four branches of the dispersion surface can be excited by linearly polarized incident waves. This means that elliptically polarized X-rays can also be produced by crystal diffraction. Therefore in principle all polarization experiments that can be performed with visible light are also possible with X-rays.

### 1. Introduction

Polarization phenomena which are observed with visible light (such as birefringence and optical activity) are generally not found for X-rays. This is because of the dielectric properties of most materials in the X-ray frequency region. Little attention has however been paid to the fact that polarization phenomena similar to those observed with visible light occur in crystal diffraction. This can be demonstrated by considering the influence of polarization on dynamical diffraction effects. One such effect which is sensitive to the state of polarization of crystal waves is the occurrence of Pendellösung fringes in the diffraction patterns from wedge-shaped crystals, which was first observed by Kato & Lang (1959). An explanation of Pendellösung

fringes in terms of spherical wave theory was given by Kato (1961). Furthermore the effect of X-ray polarization on these fringes was investigated by two groups of authors: by Hattori, Kuriyama & Kato (1965) and by Hart & Lang (1965). They have shown that an unpolarized incident wave causes a fading of the fringes which can be explained as a superposition of two sets of fringes corresponding to waves with their polarization vectors parallel and perpendicular to the plane of incidence respectively. Besides that, Hart & Lang showed that there is only one set of fringes (and naturally no fading) when the incident wave is polarized perpendicular to the plane of incidence.

Still one important case remains to be investigated. This is the case of a linearly polarized incident beam with an electric vector making an angle different from 0 or 90° with the plane of incidence. From the theoretical results of Molière (1939) one can deduce that the diffraction of such a wave will give rise to elliptically

\* Present address: Institute of Applied Physics, Technische Hochschule, Vienna, Austria.

polarized waves and to a fading of Pendellösung fringes.

This has not previously been experimentally shown. The experiments required are rather difficult to perform since the intensity after diffraction by two crystals (one serving as a polarizer) which do not have the same plane of incidence, is extremely low.

In this paper we present experimental results which prove that an incident plane-polarized wave may after diffraction by a good crystal give elliptically polarized wavefields, *i.e.* that 'polarization mixing' does exist in the case of crystal diffraction. The term 'polarization mixing' was introduced by Ashkin & Kuriyama (1966) to denote a process where the final and the initial states of polarization are different.

In the first part, we recall Molière's results. Then, the variation of the phase and amplitude of elliptically polarized wavefields with the depth in the crystal and their interference is discussed. In the second part, the results of our experiment and some conclusions are given.

## 2. Theory

### (a) Polarization phenomena

The influence of the dielectric properties on the polarization of crystal waves in the case of diffraction has been very carefully studied and described in a paper by Molière (1939). His results will be briefly reviewed here.

According to Molière, the fundamental equations of the dynamical theory have the following form:

$$\left(1 - \frac{k^2}{K_m^2}\right) \mathbf{D}_m = \sum_q^N (C_{mq} \mathbf{D}_q)_{\perp \mathbf{K}_m}. \quad (1)$$

The  $C_{mq}$  generally have tensor character. They consist of the Fourier coefficients of the scalar dielectric susceptibility  $\chi_{m-q}$  and a resonance term  $A_{mq}$ , which can be split into a scattering and an absorption term:

$$C_{mq} = \chi_{m-q} + A_{mq}^{\text{scat}} + A_{mq}^{\text{abs}}. \quad (2)$$

Absorption is taken into account by the complex  $A_{mq}^{\text{abs}}$ . These  $A_{mq}^{\text{abs}}$  terms only become important near an absorption edge. They have tensor character and thus determine the tensor properties of  $C_{mq}$ . For X-ray frequencies far from an absorption edge,  $A_{mq}$  is negligibly small and the  $C_{mq}$  are equal to the Fourier coefficients of the dielectric susceptibility  $\chi$  which depend only on the difference of the indices  $m-q$ .

Following Molière, we introduce  $N$  different coordinate systems (one for each beam) for a 'mixed representation' of the tensor  $C_{mq}$ . The  $m$ th coordinate system has its  $z$  axis parallel to the wave vector  $\mathbf{K}_m$ . The amplitudes  $\mathbf{D}_m$  are perpendicular to the corresponding  $\mathbf{K}_m$ . Taking the index  $\perp \mathbf{K}_m$  on the right hand side of equation (1) into account, we obtain a two-dimensional reduced tensor  $C'_{mq}$ , which contains no  $z$  components. With the introduction of the excitation errors  $\varepsilon_m$  as usual:

$$2\varepsilon_m = \left(1 - \frac{k^2}{K_m^2}\right), \quad (3)$$

a system of two-dimensional matrix equations is obtained

$$\sum_q^N \{C_{mq} - 2\varepsilon_m \delta_{mq}\} \mathbf{D}_q = 0 \quad (4)$$

where  $\delta_{mq}$  is Kronecker's delta. This can alternatively be written as a  $2N$ -dimensional matrix equation

$$([\mathbf{C}] - 2[\varepsilon])\mathbf{D} = 0 \quad (5)$$

where  $[\varepsilon]$  contains the excitation errors and the two-dimensional unitary matrices  $E_2$ :

$$[\varepsilon] = \begin{pmatrix} \varepsilon_0 E_2 & 0 & 0 & \dots \\ 0 & \varepsilon_m E_2 & 0 & \dots \\ 0 & 0 & \varepsilon_q E_2 & \dots \\ \dots & \dots & \dots & \dots \end{pmatrix} \quad (6)$$

and  $N$  is the number of beams.

We now assume a two-beam case where only one reflexion  $m$  is excited. The matrix  $[\mathbf{C}]$  has then the following form:

$$[\mathbf{C}] = \begin{pmatrix} C_{00}^{x_0 x_0} & C_{00}^{x_0 y_0} & C_{0m}^{x_0 x_m} & C_{0m}^{x_0 y_m} \\ C_{00}^{y_0 x_0} & C_{00}^{y_0 y_0} & C_{0m}^{y_0 x_m} & C_{0m}^{y_0 y_m} \\ C_{m0}^{x_m x_0} & C_{m0}^{x_m y_0} & C_{mm}^{x_m x_m} & C_{mm}^{x_m y_m} \\ C_{m0}^{y_m x_0} & C_{m0}^{y_m y_0} & C_{mm}^{y_m x_m} & C_{mm}^{y_m y_m} \end{pmatrix}. \quad (7)$$

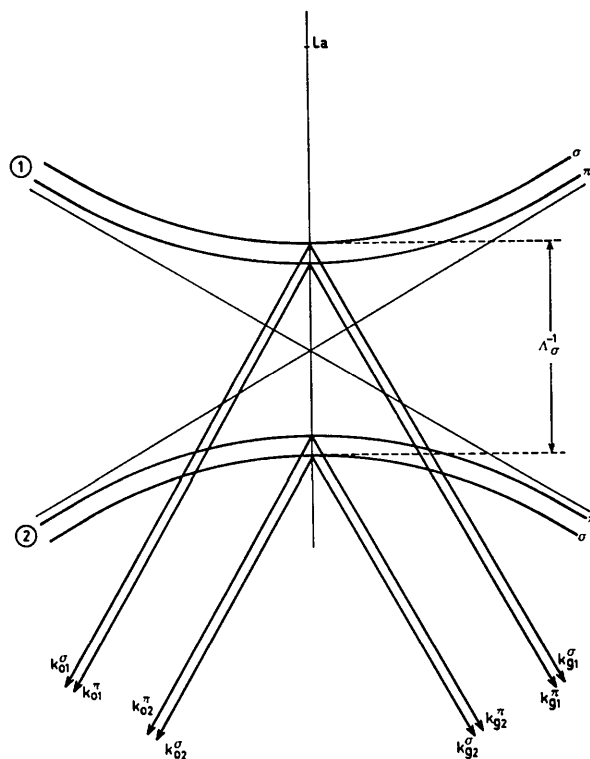


Fig. 1. Dispersion surface and wave vectors in the two-beam case.

Equation (5) can be regarded as an eigenvalue problem. The  $2N$  dimensional eigenvectors  $\mathbf{D}$  contain the components of the electric vectors of the crystal waves ( $D_0^{x_0}, D_0^{y_0}, D_m^{x_m}, D_m^{y_m}$ ).

The polarization of the crystal waves depends on the form of the eigenvectors  $\mathbf{D}$  and on the properties of the matrix  $[\mathbf{C}]$ . In the most general case, it is a non-hermitian matrix and the system of crystal waves is elliptically polarized with no definite relation between the axes and the eccentricities of the ellipses. If we assume that the crystal has a centre of symmetry and neglect absorption, the matrix  $[\mathbf{C}]$  is real and symmetric and the system of crystal waves is linearly polarized with mutually perpendicular planes of polarization. This is the case generally assumed in ordinary dynamical theory of diffraction.

In the two-beam case and with the coordinate system mentioned above, the matrix  $[\mathbf{C}]$  has the following form:

$$[\mathbf{C}] = \begin{pmatrix} \chi_0 & 0 & \chi_m & 0 \\ 0 & \chi_0 & 0 & \chi_m \cos 2\theta \\ \chi_m & 0 & \chi_0 & 0 \\ 0 & \chi_m \cos 2\theta & 0 & \chi_0 \end{pmatrix} \quad (8)$$

where  $\theta$  is the Bragg angle.

Equation (5) then gives the well known dispersion surface for the two-beam case with four branches; two for which the electric vector lies in the plane of incidence ( $\pi$  polarization) and two for which it is perpendicular to this plane ( $\sigma$  polarization). As mentioned already, in principle this does not hold near an absorption edge but can be used as a good approximation. In a crystal with a centre of symmetry the  $A_{mq}^{\text{scat}}$  are purely real and the  $A_{mq}^{\text{abs}}$  purely imaginary. For frequencies below the frequency of an absorption edge  $A_{mq}^{\text{abs}}$  becomes small and can be neglected. As was further shown by Molière and Laue (1960), all  $A_{mq}^{\text{scat}}$  become real and equal to each other if the X-ray wavelength

is sufficiently larger than the Bohr radius of the crystal atoms. Since in this case  $A_{mq}^{\text{scat}}$  depends on  $(m-q)$  only, we can write

$$C_{mq} = \chi_{m-q} + \Delta\chi_{m-q} \quad (8a)$$

where  $\Delta\chi_{m-q}$  represents a scalar correction term. If  $[\mathbf{C}]$  is a real, symmetrical tensor its eigenvectors represent crystal waves which are linearly polarized perpendicular to each other. The fundamental equations assume the form originally developed by Laue. This means that, because of the dielectric properties of the crystals, elliptically or circularly polarized crystal waves are not to be expected in practical cases.

The results of the calculations of the scattering probability  $C_{\lambda\lambda'}$  from one state of polarization into another by Hojo, Ohtsuki & Yanagawa (1966) are in principle equivalent to this. The value of  $C_{\lambda\lambda'}$  was calculated in terms of the wave functions which are used in the usual photo-electric excitation calculations. The wave function used for the ground state is the product of hydrogen-like eigenfunctions of electrons. These wave functions have proved to give good results in calculations of absorption coefficients for X-rays. Since it can be shown that  $C_{\lambda\lambda'}$  is zero as long as these wave functions are used, it was concluded by Hojo *et al.* that, to a very good approximation, waves with a polarization different from the initial one are not produced in the crystal. That means that the production of elliptically polarized X-rays is not possible owing to the dielectric properties of the crystals. It is however possible to obtain elliptically polarized X-rays by crystal diffraction as will be shown in the following.

#### (b) Polarization of the crystal waves in the two-beam case

For the two-beam case the dispersion surface is given by

$$\text{Det} \{[\mathbf{C}] - 2[\boldsymbol{\varepsilon}]\} = 0 \quad (9)$$

with  $[\mathbf{C}]$  from equation (7) or (8). This gives the well known four branch dispersion surface which lies in real  $\mathbf{k}$  space if absorption is neglected (Fig. 1). The excitation points for waves with their electric vectors perpendicular and parallel to the plane of incidence lie on the  $\sigma$  and  $\pi$  branches respectively. This has the consequence that the wave-vector difference between waves of type 1 and type 2 and therefore also the Pendellösung fringe distance depends on the state of polarization. If natural light (*i.e.* light where all states of polarization are equally present) is diffracted this will result in the superposition of two fringe systems with different spacing. This causes a periodic fading of the Pendellösung fringes which was first observed and explained by Hart & Lang (1965). An explanation in terms of spherical wave theory was given by Hattori *et al.* The amplitude of the fringes is modulated by the beat of the two patterns according to the factor

$$\cos \left\{ \pi \left( \frac{1}{A_\sigma} - \frac{1}{A_\pi} \right) t \right\} \quad (10)$$

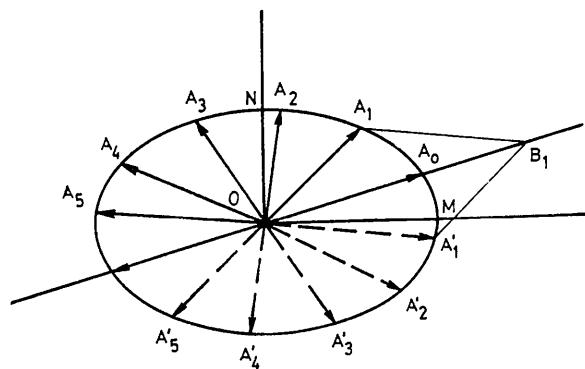


Fig. 2. Superposition of the amplitudes of two elliptically polarized waves with the same axes but different sense of turning. Full line: electric field vector of the left hand polarized wave. Dotted line: electric field vector of the right hand polarized wave. At the time  $t_0$  the amplitudes of both waves are  $\vec{OA}_0$ . At  $t_1$  they are  $\vec{OA}_1$  and  $\vec{OA}'_1$ , giving a resultant  $\vec{OB}_1$ .

where  $t$  is the crystal thickness and  $A_\sigma$  and  $A_\pi$  are the fringe distances for  $\sigma$  and  $\pi$  polarizations respectively. For the exact Bragg condition and the symmetrical Laue case, the wave-vector difference is given by

$$(\mathbf{k}_{g1} - \mathbf{k}_{g2})_{\sigma, \pi} = \frac{k}{\cos \theta} C |\chi_h| = \frac{1}{A_{\sigma, \pi}} \quad (11)$$

with the polarization factor  $C$  which is  $C=1$  for  $\sigma$  polarization and  $C=\cos 2\theta$  for the  $\pi$  polarization. The quantity  $\chi_h$  is the Fourier coefficient of the susceptibility which is directly related to the structure factor.

If the incident wave is already linearly polarized we must consider a possible interference between all crys-

tal waves which are now coherent. Let us assume that the electric vector is inclined at  $45^\circ$  with respect to the plane of incidence. If we consider only the diffracted waves we have four component plane waves with the wave vectors  $\mathbf{k}_{g1}^\sigma$ ,  $\mathbf{k}_{g1}^\pi$ ,  $\mathbf{k}_{g2}^\pi$ ,  $\mathbf{k}_{g2}^\sigma$ . Unlike in the case of natural light these waves with different states of polarization are not independent.

We shall suppose that the incident wave is a plane wave which is nearly the experimental case. For simplicity, we shall further assume that the deviation from the Bragg angle  $\Delta\theta$  is zero and that the reflecting planes are normal to the entrance surface of the crystal. If we consider the waves with index (1) only, we

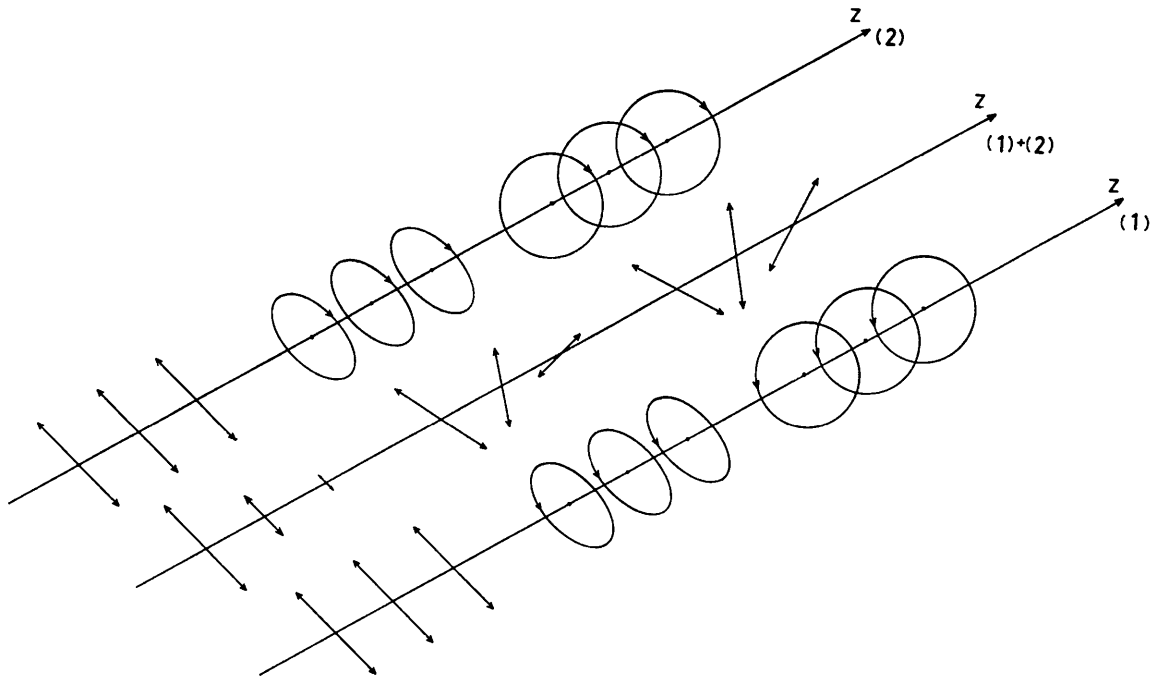


Fig. 3. Variation of the state of polarization with the depth  $z$  in the crystal. Right: wavefield (1) – Left: wavefield (2) – Centre: the resulting state of polarization where wavefields (1) and (2) overlap.

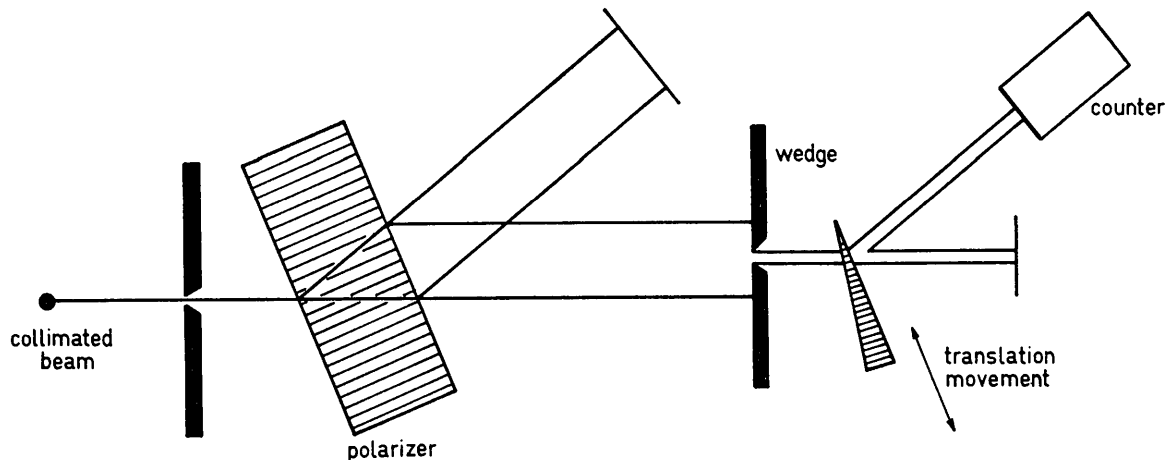


Fig. 4. Experimental arrangement.

find that the crystal diffraction introduces a phase difference of

$$2\pi(\mathbf{k}_{g1}^\sigma - \mathbf{k}_{g1}^\pi) \cdot \mathbf{r} = \pi \left( \frac{1}{A_\sigma} - \frac{1}{A_\pi} \right) \cdot t \quad (12)$$

between the  $\sigma$  and  $\pi$  components for a crystal of a thickness  $t$ . This means that the wavefield belonging to branch (1) of the dispersion surface will generally be elliptically polarized. The amplitude ratio is given by

$$\frac{E_1^\sigma}{E_1^\pi} = \frac{a_1^\sigma}{a_1^\pi} \exp \{ 2\pi i (\mathbf{k}_{g1}^\sigma - \mathbf{k}_{g1}^\pi) \cdot \mathbf{r} \} = \frac{a_1^\sigma}{a_1^\pi} \exp (i\delta) \quad (13)$$

where  $a_1^\sigma$ , and  $a_1^\pi$  are the amplitudes of the component plane waves. We shall now neglect absorption which gives

$$\frac{a_1^\sigma}{a_1^\pi} = \frac{a_2^\sigma}{a_2^\pi} = \cos 2\theta. \quad (13a)$$

A wave is of right-hand polarization for  $\delta > 0$ . The waves belonging to the (1) branches of the dispersion surface are therefore of right-hand polarization while the branch (2) waves form a left-handed polarized wave. This can immediately be seen from Fig. 1 where

$$\delta_2 = 2\pi(\mathbf{k}_{g2}^\sigma - \mathbf{k}_{g2}^\pi) \cdot \mathbf{r} < 0 \quad (14)$$

and we can notice that  $\delta_1 = -\delta_2$ . This remains true even for  $\Delta\theta \neq 0$ .

We can now introduce a polarization periodicity  $\Omega$ . It is given by

$$\frac{1}{\Omega} = \frac{1}{2}(1 - |\cos 2\theta|) \frac{1}{A_\sigma}. \quad (15)$$

The phase difference between the  $\sigma$  and  $\pi$  components can thus be expressed as

$$\exp (i\delta) = \exp (2\pi i t / \Omega). \quad (16)$$

This means that the same state of polarization is repeated with a depth periodicity of  $\Omega$ . Since  $|\delta_1| = |\delta_2|$ , the waves belonging to the (1) and (2) branches of the dispersion surface always have the same state of polarization. If  $\delta = \pi/2 + n\pi$  ( $n$ , integer) the wave is circularly polarized. Circular polarization occurs for a thickness of  $\Omega/4 + n\Omega/2$  or  $\delta = \pi/2$ . A crystal with such a thickness is therefore a quarter-wave plate for both wave fields (1) and (2) and could in principle be used to investigate elliptically polarized X-rays.

If the wave fields (1) and (2) overlap, they will interfere and give fringes at the exit surface. The contrast of the fringes depends on the state of polarization. It is strongest where both wave fields have linear polarization, and a fading is expected where both wave fields have circular polarization. It can easily be shown that an interference between two elliptically polarized waves with the same amplitude and eccentricity but with opposite sense of turning give a linearly polarized wave. The amplitude and orientation of this linearly polarized resultant wave depends on the phase difference between the two elliptically polarized waves. If at a

given time the two waves are both represented by  $\vec{OA}_0$  (Fig. 2), the resultant electric vector is  $2\vec{OA}_0 \cos \omega t$ . The value of the amplitude can therefore vary from  $2\vec{OM}$  to  $2\vec{ON}$  depending on the phase difference and thus on the thickness of the crystal. The period of this oscillation from  $2\vec{OM}$  to  $2\vec{ON}$  is equal to one half of the Pendellösung fringe distance. The greater the eccentricity of the ellipse, the greater is the difference between  $\vec{ON}$  and  $\vec{OM}$  and the greater the contrast of the fringes. Where the ellipse degenerates into a straight line, the contrast of the fringes is a maximum. In that limiting case the plane of polarization does not turn. On the other hand, the contrast is a minimum when the ellipse becomes a circle ( $\delta = \pi/2 + n\pi$ ). In this case the resultant amplitude is constant and the only the plane of polarization rotates. So a non-absorbing diffracting crystal with a thickness corresponding to a fading region rotates the plane of polarization of the incident radiation. The fringe region corresponds to a thickness for which both wave fields (1) and (2) are linearly polarized.

If the crystal is thick enough and the incident plane wave does not fulfil the Bragg condition exactly the two wave fields (1) and (2) will not overlap. They propagate in different directions which are given by the normals to the dispersion surface. At the exit surface the wave fields split into primary and diffracted waves which can thus be linearly, elliptically or circularly polarized depending on the crystal thickness. These results are summarized in Table 1 and illustrated in Fig. 3.

Table 1. *Crystal thickness and polarization*

Thickness	$\delta =  \mathbf{k}^\sigma - \mathbf{k}^\pi $	Polarization	Fading
0	0	linear	fringes
$\Omega/4$	$\pi/2$	circular	fading
$\Omega/2$	$\pi$	linear	fringes
$3\Omega/4$	$3\pi/2$	circular	fading

So far we have neglected absorption. Since the absorption is different for all four branches of the dispersion surface in the two-beam case, it affects the amplitude ratios and equation (13a) is no longer satisfied. The state of polarization of the crystal waves remains the same if we still call a wave circularly polarized for  $\delta = \pi/2$  even if  $|\mathbf{E}_x| \neq |\mathbf{E}_y|$ . It follows that the fading of the Pendellösung fringes is an indicator of the excitation of waves with their tie-points on all four branches of the dispersion surface. If the incident radiation is linearly polarized but neither in the  $\pi$  nor in the  $\sigma$  direction the fading is thus an indicator of the existence of elliptically polarized crystal waves.

### 3. Experimental

As mentioned already, Hart & Lang were the first to give an explanation for the difference in Pendellösung

fringe patterns obtained with unpolarized and with polarized radiation. They showed that the fading of the fringes observed with unpolarized incident radiation disappeared when  $\pi$ -polarized radiation was used. The experiment which is described here is however different from the one performed by Hart & Lang. As explained in § 2, we expect a periodic fading of the Pendellösung fringes not only for unpolarized but also for linearly polarized incident radiation if the electric vector is inclined to the plane of incidence. In this case, the fading cannot be explained by a superposition of two independent fringe patterns. An experiment of this kind was performed using a two-crystal arrangement similar to that of Cole, Chambers & Wood (1961) (Fig. 4). The Pendellösung fringes obtained by diffraction of linearly polarized X-rays by a wedge-shaped crystal were observed. The first crystal is a thick silicon crystal which is used as a Borrmann-type polarizer. With Mo  $K\alpha$  radiation and a 220 reflexion, the intensity ratio of radiation with  $\sigma$  and  $\pi$  polarizations is approximately 1:10 for the type (1) waves. The type (2) waves are strongly absorbed. Both the transmitted and the diffracted waves are therefore nearly completely polarized after the first crystal with their electric vectors parallel to the diffracting planes. The second crystal is wedge-shaped and mounted on a traversing mechanism. The intensity diffracted by the second crystal is recorded with a counter.

By a rotation of the first crystal around the centre line of the anomalously transmitted beam the E vector

can be made to take up any angle with respect to the plane of the drawing and thus to the plane of incidence at the second crystal. Since the transmitted beam has the same state of polarization as the diffracted beam, it is much more convenient to use this primary beam because it does not sensibly change its direction when the polarizer is turned.

If  $\varphi$  is the polarizer angle, we will call  $\varphi=0$  the 'parallel' arrangement. For this position of the polarizer the rocking curves of the second crystal are very sharp double-crystal rocking curves. Because the polarizer is thick, the Borrmann triangle is large and the slit before the second crystal will only receive radiation of very small angular divergence (Authier, 1961). This means that we have essentially a pseudo plane-wave case for the second crystal. When the polarizer is set at another angle (especially at  $\varphi=90^\circ$  or  $\varphi=270^\circ$ ) the rocking curves of the second crystal have the appearance of broad, triangular peaks. The intensity is of course drastically decreased. The average intensity in the experiment represented by the upper curve in Fig. 5 is approximately 600 counts/hour. In the 'parallel' arrangement all the radiation diffracted by the first crystal is also diffracted by the second one. In any other position,  $\varphi \neq 0^\circ$ , X-rays incident on the first crystal with a deviation  $\Delta\theta$  from the Bragg angle have an angle of incidence at the second crystal equal to  $\theta_B + \Delta\theta'$ . The deviation  $\Delta\theta'$  is a function of  $\Delta\theta$ ,  $\varphi$ , and of the angle  $\alpha$  of the X-rays with the plane of incidence of the second crystal, *i.e.* the plane of Fig. 4. As the

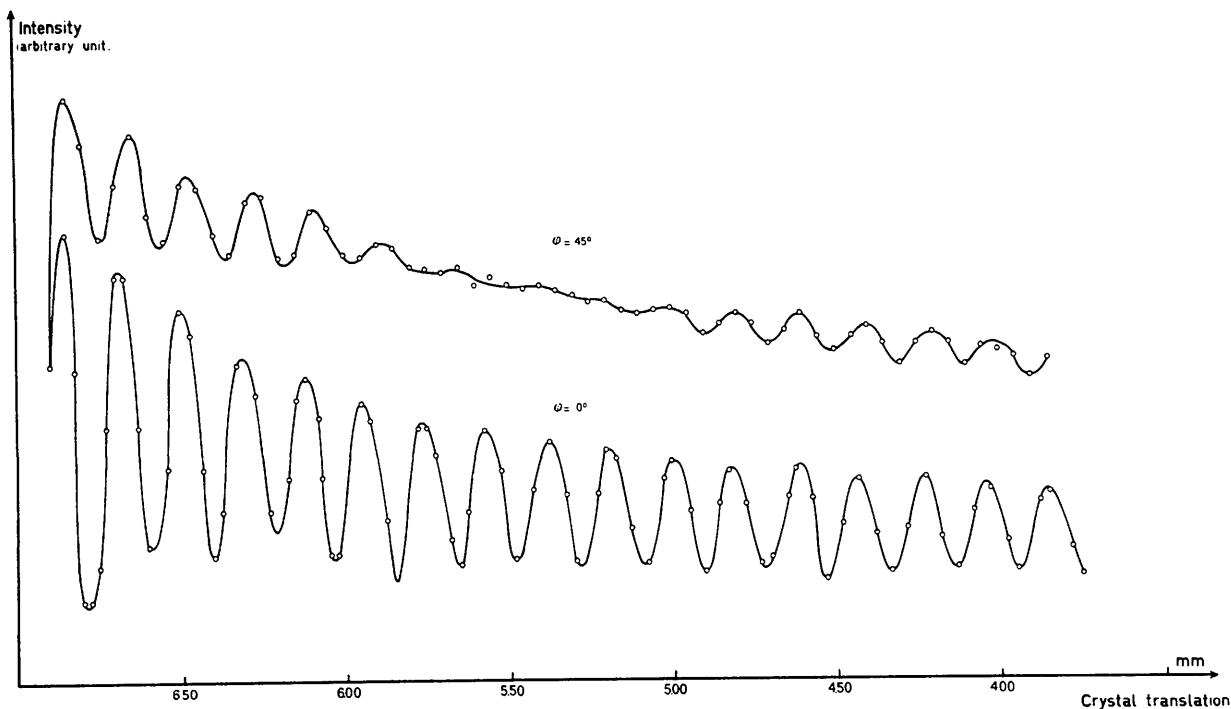


Fig. 5. Intensity versus position of the crystal for linearly polarized incident radiation. Lower curve: polarization perpendicular to the plane of incidence. Upper curve: polarization inclined at  $45^\circ$  with respect to the plane of incidence. Note that the fading of the fringes in the upper curve is *not* due to unpolarized incident radiation.

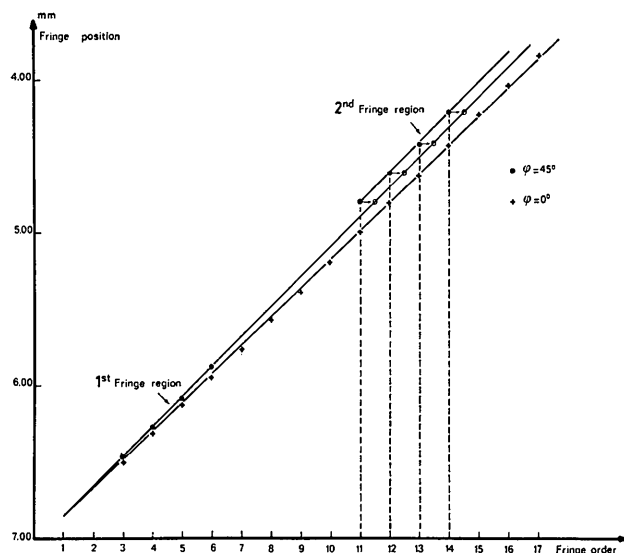


Fig. 6. Fringe position versus fringe order.  $\phi = 0^\circ$ ; points lie on a straight line.  $\bullet \phi = 45^\circ$ ; Points lie on a straight line only if half integers are assigned to the fringes in the second fringe region.

vertical divergence of the incident beam is about  $10^{-2}$ , X-rays diffracted by the second crystal have different angles  $\alpha$  and therefore different  $\Delta\theta'$ . The rocking curve is thus greatly broadened and the intensity measured after the second crystal at the maximum of the rocking curve can be considered as an integrated 'nearly plane-wave' intensity. Some preliminary experiments showed that the fringes for  $\phi \neq 0^\circ$  were well visible when the second crystal was moved on its translation movement.

Fig. 5 shows the results of measurements of the intensity diffracted by the second crystal as a function of its thickness for both  $\phi = 45^\circ$  and  $\phi = 0^\circ$ . The fading of the fringes and the shift of the fringe position are clearly visible. Fig. 6 illustrates the variation of fringe position as a function of the fringe order. As shown by Hattori *et al.* (1965), a nice alignment of the plotted points is obtained by assigning half integers to the fringes of the fringe regions of even order and integers to the fringes in the regions of odd order.

From these fringe patterns for different polarizer positions the following facts can therefore be deduced.

(i) In the  $\phi = 0^\circ$  position of the polarizer no fading effect is observed because only waves belonging to two branches of the dispersion surface are excited.

(ii) A fading of the fringes occurs both for unpolar-

ized radiation and for the  $\phi = 45^\circ$  position of the polarizer. This proves that in both cases all four branches of the dispersion surface are excited. As explained in § 2, the crystal waves will be elliptically polarized for linearly polarized incident radiation. If the crystal is thick and absorption is weak, the type (1) and (2) waves can be separated and circularly polarized X-rays can thus be obtained.

In conclusion we can recall that polarization mixing has been analysed theoretically by Molière (1939) and by Hojo, Ohtsuki & Yanagawa (1966) for the case of pure refraction, *i.e.* in a one-beam case. Molière showed that in some cases a polarization mixing could exist but he expected the effect to be small.

Hojo *et al.* studied the problem quantum mechanically and concluded that polarization mixing does not exist as long as hydrogen-like atomic wave functions are a good approximation. These results show that in any case polarization mixing is expected to be very weak in simple transmission. Our experiment shows however that in the two-beam case polarization mixing does exist. In principle therefore all polarization experiments which are possible with visible light can be performed with X-rays. Since however diffracting crystals must be used as polarizers and compensators it is in practice difficult to obtain sufficient intensity to find evidence for the proposed effects.

The authors wish to thank Professor Authier for his interest in this work and for many stimulating discussions. One of us (PS) also wants to acknowledge financial support from the French government and the Austrian Forschungsrat.

#### References

- ASHKIN, M. & KURIYAMA, M. (1966). *J. Phys. Soc. Japan*, **21**, 1549.  
 AUTHIER, A. (1961). *Bull. Soc. Fr. Minér. Crist.* **84**, 51.  
 COLE, H., CHAMBERS, F. W. & WOOD, C. G. (1961). *J. Appl. Phys.* **32**, 1942.  
 HART, M. & LANG, A. R. (1965). *Acta Cryst.* **19**, 73.  
 HATTORI, H., KURIYAMA, H. & KATO, N. (1965). *J. Phys. Soc. Japan*, **20**, 1047.  
 HOJO, A., OHTSUKI, Y. H. & YANAGAWA, S. (1966). *J. Phys. Soc. Japan*, **21**, 2082.  
 KATO, N. & LANG, A. R. (1959). *Acta Cryst.* **12**, 787.  
 KATO, N. (1961). *Acta Cryst.* **14**, 526, 627.  
 LAUE, M. VON (1960). *Röntgenstrahlinterferenzen*. Frankfurt: Akademische Verlagsgesellschaft.  
 MOLIERE, G. (1939). *Ann. Phys.* **35**, 272.