Table 1. *Theoretical and experimental (in parentheses) values of* $\langle\alpha_{\mathbf{h}}\rangle$ *and* $\sigma_{\alpha_{\mathbf{h}}}$ *from p samples of q events distributed according to the Von Mises distribution* $M[\theta; 0, G]$

p, q	50, 5000	20, 2000	10, 10	5, 5
G	1·5	3	1·5	1·5
$\langle\alpha\rangle$	4451	4864	9·69	4·45
	(4479)	(4848)	(10·0)	(4.18)
σ	53·2	36·2	2·38	1·68
	(59·8)	(36·0)	(2·23)	(1·61)

Equation (39) is shown for some selected cases in Fig. 1. The distribution $P(x)$ was invoked by Hull & Irwin in order to justify their weight

$$w'_h = \psi\, e^{-x^2} \int_0^x \exp t^2\, \mathrm{d}t. \qquad (40)$$

In particular they supposed that (40) (the dotted line in Fig. 1) roughly corresponds to $P(x)$. Our results show that:

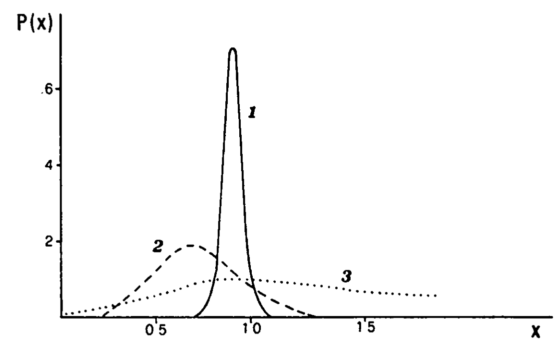

Fig. 1. The distribution $P(x)$ given by (43) is shown for selected values of parameters m and σ''. Line 1: $m = 0{\cdot}95$, $\sigma'' = 0{\cdot}03$. Line 1 represents the distribution of x for a sample of ten complex vectors $2\, e^{i\theta_j}$ distributed according to $M[\theta_j; 0, 2]$. Line 2: $m = 0{\cdot}84$, $\sigma'' = 0{\cdot}13$. Line 2 represents the distribution of x for a sample of ten complex vectors $e^{i\theta_j}$ distributed according to $M[\theta_j; 0, 1]$. Line 3: The distribution (6).

(1) $P(x)$ depends on the two parameters m and σ'' while no parameter is in (40);

(2) the distribution (40) does not agree with $P(x)$. In particular the maximum of $P(x)$ is usually not at 1.

8. Conclusions

The asymptotic distribution of the resultant of the complex vectors $G_j \exp(i\theta_j)$, $j = 1, \ldots, r$, where $\theta_j = \theta_{\mathbf{k}_j} + \theta_{\mathbf{h}-\mathbf{k}_j}$ is distributed according to $M[\theta_j; \varphi_{\mathbf{h}}, G_j]$, is calculated. The statistical results suggest that the phase of the resultant is distributed around $\varphi_{\mathbf{h}}$ approximately according to a Von Mises distribution with concentration parameter equal to $\langle\alpha_{\mathbf{h}}\rangle$, while the modulus of the resultant is normally distributed around $\langle\alpha_{\mathbf{h}}\rangle$ [given by (34) and (36) for non-centrosymmetric and centrosymmetric structures, respectively].

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Polarization Phenomena of X-rays in the Bragg Case

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Abstract

Using a double crystal diffractometer with an additional crystal between the two crystals used as X-ray-optical polarizer and analyzer the phase relation between mutually perpendicularly polarized wave fields is examined in the Bragg case. The additional crystal is a (110)-surface oriented silicon crystal adjusted for the symmetric 220 Cu $K\alpha_1$ Bragg case. In the case of coherent excitation of both σ- and π-polarized wave fields in the silicon crystal it is experimentally shown that a unique phase relation

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exists. In a further experiment it is demonstrated that in the case of incoherent excitation the rocking curve is the sum of the σ- and π-polarized intensities without any phase relation. Owing to the different half width of these two curves, step-shaped flanks of the rocking curve are found at excitation with unpolarized radiation.

Introduction

In the case of simple transmission, polarization phenomena are generally not observed with X-rays. Such phenomena, however, may occur in connection with crystal diffraction because of the different behaviour of differently polarized X-rays (Hart, 1978). From the dynamical theory of X-ray diffraction (Laue, 1960; Batterman & Cole, 1964) it can be concluded that in the two-beam Bragg case four wave fields are generated in the crystal by excitation with unpolarized radiation. By excitation with linearly polarized waves two wave fields exist only. Out of the range of the so-called Darwin's total reflection the phase shift between the vectors of the dielectric displacement $\mathbf{D}_{0i}$ and $\mathbf{D}_{Hi}$ $(i = 1, 2)$ does not change. 0 designates the vector of the direction of the incident wave and H designates that of the diffracted one. The sum of the vectors of the dielectric displacement represents the so-called wave field. But inside this range the phase shift changes from zero to π. Because the rocking curves of the σ- and π-polarized wave fields differ in half width there is a range where a phase difference between the mutually perpendicularly polarized wave fields occurs and it is expected that polarization phenomena may be observed in the Bragg case of X-ray diffraction. Hence, changing the polarization state of the diffracted X-rays could possibly be dependent on the angle of incidence at coherent excitation of both σ- and π-polarized wave fields with highly collimated X-radiation. σ means that the electric field vector is normal to the reflection plane and π that the electric field vector lies in this plane. The proof of the phase shift between the mutually perpendicularly polarized waves is the aim of this paper. First, however, the differences and the corresponding polarization states of the diffracted X-ray beam are calculated for the performed experiments.

Theoretical considerations

The rocking curves of both σ- and π-polarized wave fields were calculated using the formalism given by Zachariasen (1945) and are shown in Fig. 1. The asymmetric shape of the rocking curves is caused by absorption (Borrmann effect). In the range of Darwin's total reflection the phase difference between the waves $\mathbf{D}_0^\sigma$ and $\mathbf{D}_H^\sigma$ and $\mathbf{D}_0^\pi$ and $\mathbf{D}_H^\pi$, respectively, changes from zero to π. This phase difference is designated as η and is plotted against the angle of incidence θ in Fig. 1(b) (James, 1963). η^σ and η^π can be distinguished because of the different half width of the corresponding rocking curves. Therefore, a phase shift between $\mathbf{D}_H^\sigma$ and $\mathbf{D}_H^\pi$ exists. This phase shift is given by $\Delta\eta = \eta^\sigma - \eta^\pi$. The dielectric displacements $\mathbf{D}_H^\sigma$ and $\mathbf{D}_H^\pi$ were calculated at varying angle of incidence θ according to the dynamical theory of X-ray diffraction. If the phase difference $\Delta\eta$ and the relevant amounts of the dielectric displacements of the mutually perpendicularly polarized waves are taken into account, the resulting vector $\mathbf{D}_H$ is given by

$$\mathbf{D}_H = \mathbf{D}_H^\sigma \cos \omega t + \mathbf{D}_H^\pi \cos(\omega t - \Delta\eta), \qquad (1)$$

where t is the time and $\omega = 2\pi kc$, c is the vacuum light velocity and $k = |\mathbf{k}|$ is the wave number of the incident wave. The projection of this vector of the diffracted wave gives the following equation in the plane perpendicular to the direction of propagation

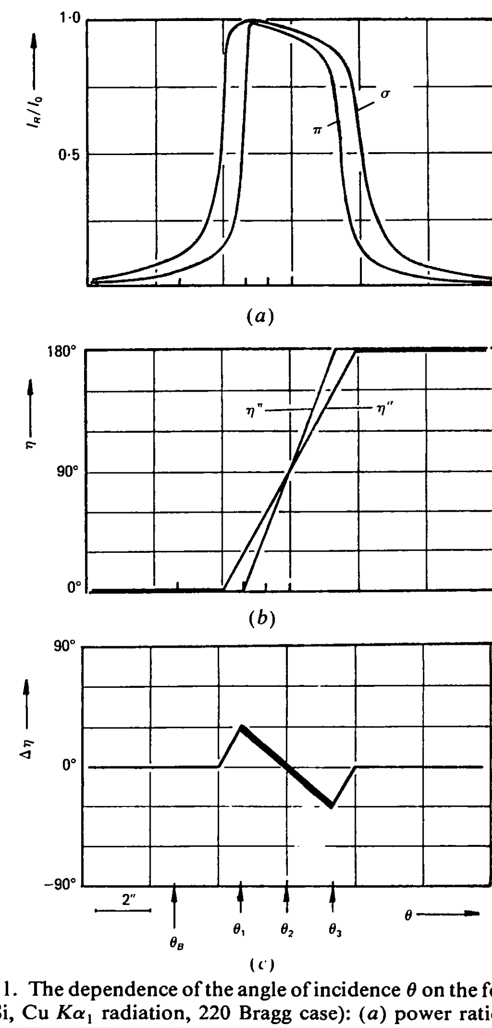

Fig. 1. The dependence of the angle of incidence θ on the following (Si, Cu $K\alpha_1$ radiation, 220 Bragg case): (a) power ratio I_R/I_0; (b) phase difference between $\mathbf{D}_0^\sigma$ and $\mathbf{D}_H^\sigma(\eta^\sigma)$ and between $\mathbf{D}_0^\pi$ and $\mathbf{D}_H^\pi(\eta^\pi)$; ($c$) phase difference between the mutually perpendicularly polarized components of the diffracted beam, $\Delta\eta = \eta^\sigma - \eta^\pi$.

(z axis):

$$\frac{x^2}{(D_H^\sigma)^2}-\frac{2xy}{D_H^\sigma D_H^\pi}\cos\Delta\eta+\frac{y^2}{(D_H^\pi)^2}=\sin^2\Delta\eta. \quad (2)$$

This is the general equation of an ellipse. For $\Delta\eta = n\pi$ in particular, the corresponding curve is a straight line, $y = \pm(D_H^\pi/D_H^\sigma)x$, *i.e.* the polarization state of a linearly polarized wave. The upper sign is valid for even numbers and the lower sign for odd numbers. For $\Delta\eta=(n+\frac{1}{2})\pi$ and $D_H^\sigma = D_H^\pi$ the circular polarization state is obtained. If n is an even number a right-handed circularly polarized wave occurs and if n is an odd number a left-handed circularly polarized wave exists in the crystal. The polarization states of the diffracted wave of the angular positions θ_1, θ_2 and θ_3 marked in Fig. 1 were calculated as described above and are shown in Fig. 2. For simplicity the calculation was done for $D_H^\sigma = D_H^\pi$, which can be realized in the experiment by a suitable orientation of the polarization plane of the linearly polarized wave with respect to the reflection plane of the Bragg reflection. It can be seen that the polarization state of the diffracted wave depends on the angle of incidence. An experimental verification of these results is very difficult because the phase shift changes from zero to π in an angular range of some seconds of arc. Only in the thick-line part of the curve $\Delta\eta(\theta)$ in Fig. 1(c) can the phase relation be experimentally examined because the amplitudes of $\mathbf{D}_H^\sigma$ and $\mathbf{D}_H^\pi$ are of the same order of magnitude in this range.

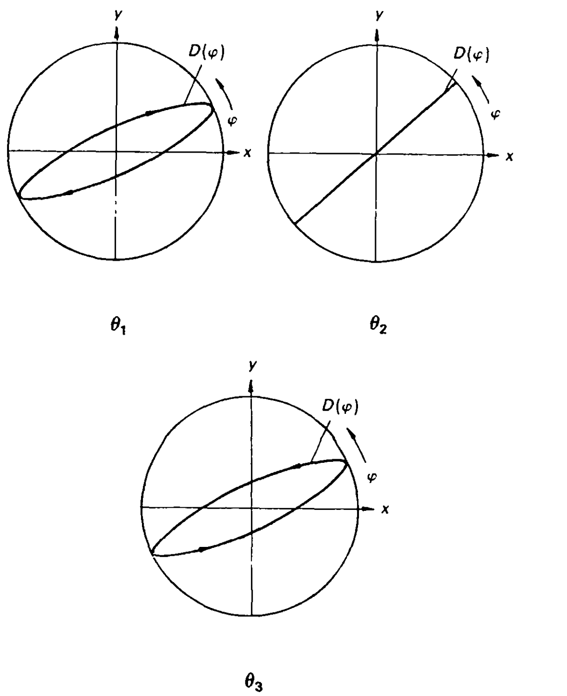

Fig. 2. The function $D(\varphi)$ of the angular positions θ_1, θ_2 and θ_3. The direction of the diffracted wave is normal to the plane of the paper.

The influence of the polarization state on the rocking curve can also be studied if the wave fields are incoherently excited by a highly collimated and unpolarized X-ray beam. On the assumption that both components are equally strongly excited the rocking curve is the sum of the rocking curves of the σ- and the π-polarized wave fields. The calculated curve convoluted with the experimental window function is represented in Fig. 6 as a broken line. On the flanks of this curve small steps can clearly be seen. These steps can be recorded experimentally by excitation with a highly collimated beam only.

Experiments

The scheme of the experimental arrangement is shown in Fig. 3. A (111)-surface oriented silicon crystal plate adjusted for the symmetric 220 Bragg case serves as the 'phase shifter' for the incident linearly polarized X-ray beam.

The thickness of this crystal plate is about 200 μm. The adjustment was made so that the reflection plane of the polarizer was inclined by 45° with respect to the reflection plane of the phase shifter. Thus the σ- and π-polarized wave fields are coherently excited. Furthermore, the excited crystal area was limited by a corresponding slit. The vertical divergence of the beam diffracted by the polarizer was limited to 216″ by a Soller slit. The angle of incidence of the phase shifter could be changed with an accuracy of 0·05″. With this double-crystal arrangement the reflection curve of the phase shifter was measured. The result is represented in Fig. 4. The half width of this curve

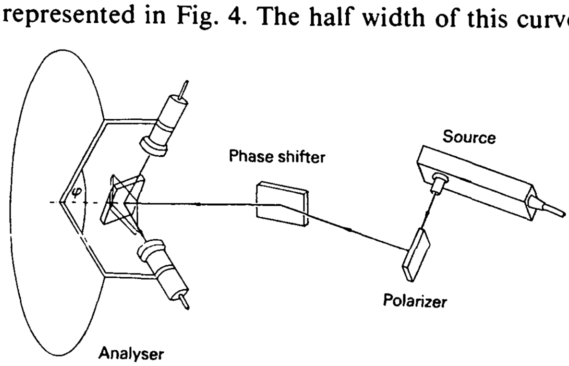

Fig. 3. The experimental arrangement.

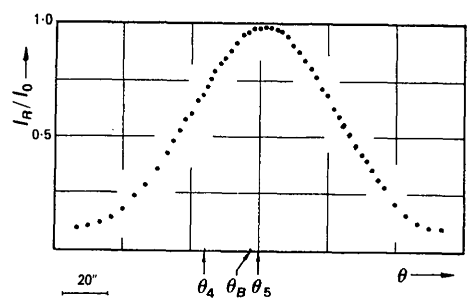

Fig. 4. Measured reflection curve for the 220 Bragg case, σ- and π-polarized components are coherently excited.

is 65″ because of the influence of the vertical divergence of the beam diffracted by the polarizer. Since the reflection plane of the phase shifter is inclined by 45° with respect to the reflection plane of the polarizer the vertical divergence is transferred by a factor of cos 45° to the horizontal divergence with respect to the phase shifter. Therefore, the crystal function was not directly measured but the convolution of it with the angular intensity distribution of the incident beam, which is very broad. This is the main problem in the experimental proof of the predictions. With a conventional X-ray tube and a single-crystal polarizer the divergence and the wavelength dispersion are very different in the horizontal and vertical planes with respect to the reflection plane of the polarizer. Better conditions can be expected with synchrotron radiation which is linearly polarized and highly collimated.

To analyse the polarization state of the wave diffracted by the phase shifter a further reflection was used. It was the symmetric 333 Bragg case. The germanium crystal plate of the analyser could be rotated about an axis parallel to the incident beam. The rotation was measured by the angle φ which is given by $\tan\varphi = y/x = D^{\pi}_{H}/D^{\sigma}_{H}$. This device rendered it possible to determine the polarization state of the incident beam by measuring its components. In the case of a non-coplanar ray path the integral intensities had to be measured because of the influence of the angular dispersion and the beam divergence (Brümmer, Eisenschmidt & Nieber, 1982). Using a highly sensitive angular setting (Brümmer, Eisenschmidt & Höche, 1982), it is possible to vary the angle of incidence for measurements of the entire reflection curve at a fixed angle φ. For the crystal positions θ_4 and θ_5 marked in Fig. 4, the dependence of the integral intensity on the angle φ was determined. The results are shown in Fig. 5. The angle φ was changed in steps of 10° over an interval of 180°.

In order to investigate the influence of the polarization on the rocking curve a further experiment was performed. A double-crystal diffractometer was adjusted so that the two silicon crystals were in the $(n, -n)$ position and the 220 Cu $K\alpha_1$ Bragg case was excited at each crystal. The angular position of the first crystal was very asymmetric. Thus, the radiation was highly collimated. The rocking curve measured by this arrangement is shown in Fig. 6. The horizontal divergence of the X-ray beam was about 0·35″. On the flanks of the measured curve the steps can clearly be seen.

The use of the analyser made it possible to separate the polarization components. The analyser was adjusted so that the σ- or the π-polarized component

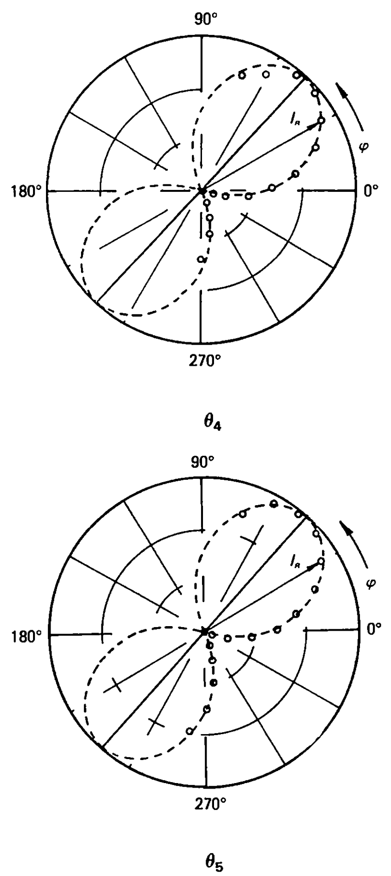

Fig. 5. Measured polarization states of the angular positions θ_4 and θ_5.

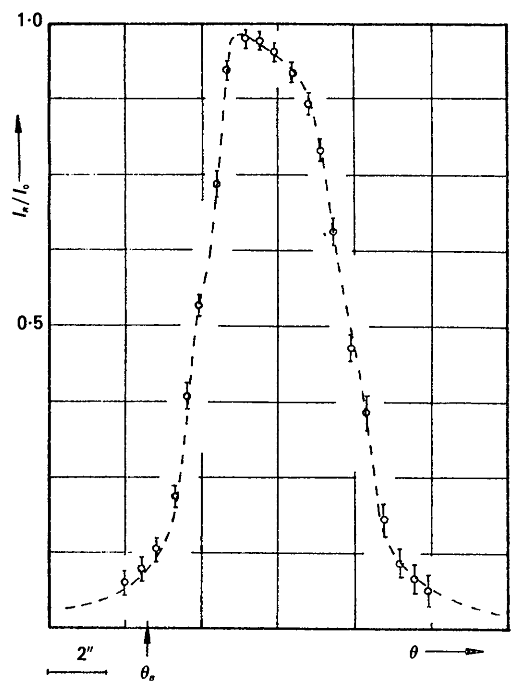

Fig. 6. 220 rocking curve excited by unpolarized and highly collimated radiation.

could be measured only. At a fixed position of the analyser the rocking curve of the second crystal was measured as in the previous experiment. The resulting rocking curves of the σ- and π-polarized waves are represented in Fig. 7. The broken lines show the theoretical curves.

Results

For both crystal positions θ_4 and θ_5 the polarization state was determined as described above. The background-corrected integral intensities I_R are shown in Fig. 5. For the comparison of the experimentally determined polarization states with the theoretically determined ones the calculated polarization states must be convoluted with the $\cos^2 \varphi'$ function of the analyser according to the law of Malus. The convoluted curves are also shown in Fig. 5 (broken lines). Both polarization states agree with the polarization state of the incident X-ray beam. Because of the high divergence of the radiation used many rocking curves are excited at each angular position. The function $\Delta\eta(\theta)$ is an odd function with respect to the centre of the rocking curves which means that both right-handed and left-handed elliptically polarized X-rays exist in the crystal simultanously. The interference of these waves gives a linearly polarized wave with the same polarization state as that of the incident wave. Absorption can be neglected. Hence, at any angular position the diffracted beam is linearly polarized where the electric-field vector is inclined by 45° with respect to the reflection plane of the phase shifter. The experimental results confirm the theoretical considerations accurately. Thus it is experimentally proved that a unique phase relation between the mutually perpendicularly polarized wave fields exists in the Bragg case of X-ray diffraction.

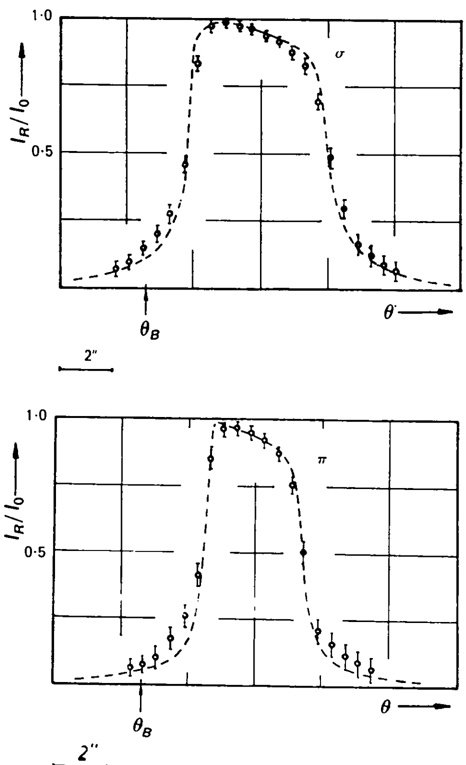

Fig. 7. 220 rocking curves of the σ- and π-polarized components.

The other experiment demonstrates the influence of polarization on the rocking curve at excitation of the reflection by unpolarized X-radiation. The steps on the flanks of the curve (see Fig. 6) are caused by the simultaneous occurrence of σ- and π-polarized components. In this case no phase relation between these components exists. If the analysing arrangement is used the rocking curves of the σ- and π-polarized wave fields can be recorded separately. The sum of these curves yields the rocking curve measured with the double-crystal diffractometer and unpolarized radiation. This effect must be taken into account in experiments with highly collimated X-radiation, in particular in contrast discussions of reflection topography using a double-crystal diffractometer to detect slow variations of lattice parameters and microdefects.

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