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Accurate Determination of Structure Factor Ratios of α Quartz

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By the use of the X-ray Pendellösung fringes in α quartz including a single Dauphiné twin, the ratios of the structure factors having the forms of hk,l and $hk,\bar{l}$ were determined with an accuracy of about a few tenths of one per cent. In the most favourable case, namely, the case of $|F_{10.1}|/|F_{10.\bar{1}}|$, the agreement between independent measurements for the same specimen was better than 0·1 %. The values of the ratios for the extinction-corrected structure factors $|F_{corr}|$ and the structure factors $|F_{cal}|$ calculated from the proposed structure in recent investigations (Young & Post, *Acta Cryst.* (1962), **15**, 337; Zachariasen & Plettinger, *Acta Cryst.* (1965), **18**, 710) are in accordance with the present ones within the mean deviation of about a few per cent, whereas the ratios for the observed values $|F_{obs}|$ differ from ours more than ten percent. Some critical remarks on the Pendellösung method of determining structure factors, particularly by means of traverse-type topographs, are put forward.

1. Introduction

Recently, it has been shown that X-ray Pendellösung fringes, a diffraction-interference effect in crystals which was theoretically predicted by Ewald (1916*a*,*b*, 1917) and first observed by Kato & Lang (1959), can be used for determining structure factors accurately on the absolute scale (Hattori, Kuriyama, Katagawa & Kato, 1965). By the use of wedge-shaped crystals, the Pendellösung fringes are recorded in diffraction topographs of both section and traverse types. The structure factor $|F_g|$ is inversely proportional to the fringe spacing Λ_g in each type of topograph, *i.e.*

$$|F_g| = f_g(\lambda, \theta_B)\Phi_g/\Lambda_g \,. \tag{1}$$

In this equation, f_g is a well-defined function depending upon the type of topograph, λ and θ_B are the wavelength and the Bragg angle respectively, and Φ_g is a geometrical factor depending upon the shape and the experimental setting of the crystal (Kato, 1961; Hattori, Kuriyama, Katagawa & Kato, 1965; Hattori, Kuriyama & Kato, 1965; Hart & Lang, 1965). Although the applicability of the method is limited to crystals which are available in a very perfect state, it seems important to develop the present method further, because the structure factor is the most basic quantity in crystal structure analysis and because the present method is free from any ambiguity due to extinction effects.

In the previous work (Hattori, Kuriyama, Katagawa & Kato, 1965), the structure factors could be determined with an accuracy of about 1% for silicon single crystals. The difficulty of improving the accuracy was entirely due to the difficulty in determining the geometrical factor Φ_g. The fringe spacing itself could be determined with an accuracy of about 0·1% in favourable cases. Recently, therefore, Kato & Tanemura (1967) designed a new experiment for determining structure factors directly on the scale of the zero order structure factor, F_0, by eliminating the geometrical factor Φ_g. Obviously, the $|F_g|$ values obtained by this experiment are truly absolute because F_0 signifies the total number of electrons in a unit cell. The principle is to take the ratio of equation (1) and the similar equation in X-ray interferometry (Bonse & Hart, 1965),

$$F_o = g(\lambda, \theta_B)\Phi_0/\Lambda_0 \,, \tag{2}$$

where $g(\lambda, \theta_B)$ is a function similar to the function f in equation (1), Λ_0 is the spacing of the thickness fringes and Φ_0 is a geometrical factor corresponding to Λ_0. In fact, the condition $\Phi_g = \Phi_0$ could be realized easily in the separate experiments for observing the Pendellösung fringes and the thickness fringes.

The present paper describes another application of this principle. Here, we are concerned with the ratio of a particular pair of structure factors. In α quartz including a single Dauphiné twin the Pendellösung fringes due to a pair of different net planes, hk,l and $hk,\bar{l}$, can be recorded simultaneously on a single plate, with the same arrangements of X-ray beams by traversing the crystal without rotation, because the Bravais lattice is not altered by the twinning. Thus, we can realize the condition $\Phi_{hk,l} = \Phi_{hk,\bar{l}}$ by the use of well-prepared wedge crystals. The ratio of the structure factors, therefore, can be given simply by

$$P \equiv |F_{hk,l}|/|F_{hk,\bar{l}}| = \Lambda_{hk,\bar{l}}/\Lambda_{hk,l} \,, \tag{3}$$

where $\Lambda_{hk,l}$ and $\Lambda_{hk,\bar{l}}$ are the fringe spacings obtained from the respective parts of the crystal divided by the twin.

The main purpose of this experiment is to discuss critically the current state of accurate determination of structure factors. Since we believe that the accuracy in this experiment is better than that in conventional experiments for the determination of structure factors, their accuracy may be checked by comparing

the ratios of structure factors, obtained by conventional methods, with ours. Here, the values of structure factors obtained by Young & Post (1962) and Zachariasen & Plettinger (1965) are discussed.

In this experiment, traverse-type topographs are used specifically although the same thing can be done by using section-type topographs. A few critical remarks are made on the application of traverse patterns to the determination of structure factors.

2. Experimental

Specimens

We need crystals which include only a single Dauphiné twin but are free from dislocations and other plate-like defects. For this purpose several plates of natural quartz were examined by Lang's (diffraction) topograph method. From the selected crystals three wedge-shaped specimens [(*a*), (*b*) and (*c*)] were prepared by cutting with a diamond wheel and grinding with 4000-mesh alundum by the aid of a metal gauge. Surface damage could be eliminated by etching with 46% hydrofluoric acid for about three minutes. Since the etching time was short, neither rounding off of the wedge shape nor roughening of the crystal surfaces occurred appreciably.

X-ray diffraction topographs

The diffraction topographs were taken by an X-ray spectrometer having a traverse stage. Ag $K\alpha_1$ radiation was used throughout this experiment. The X-ray source was about 40 μ in diameter. The distance between the source and the specimen was about 35 cm. The net plane was always set vertically so that the Bragg reflexion took place in a horizontal plane. A long slit of 100 μ in width was put vertically in front of the specimen for the traverse patterns, whereas a 20 μ slit was used for the section patterns. The vertical divergence of X-rays effective to diffraction was less than 1°. The geometrical conditions described in the paper of Hattori, Kuriyama, Katagawa & Kato (1965) with respect to the specimen, the incident X-rays and the recording plate were nearly satisfied, although they were not required to be satisfied exactly in our experiment. Fig. 1 illustrates an example of the geometrical relations between the X-rays and the crystal including a twin. Nuclear emulsion plates were used for recording the topographs.

The measurement of the fringe spacing

Some examples of X-ray topographs used for the measurements are shown in Figs. 2, 3 and 4. The Pendellösung fringes appear in the wedge-shaped part of the crystal. They are parallel to one of the crystal edges. Other fine fringes are due to plate-like defects (Kato & Lang, 1959) which have been discussed theoretically (Authier & Sauvage, 1966; Kato, Usami & Katagawa, 1966).

In some cases, the Pendellösung fringes are slightly distorted in a band region of about 0·1 mm in width close to the twin boundary. The distortion is clearly due to a lattice distortion (Kato & Ando, 1966; Ando & Kato, 1966) .A few plate-like defects are always included in the crystals, but none of them exist in the vicinity of the twin boundary concerned in favourable cases [specimen (*a*)]. In one case, the fringes were systematically distorted along a straight line. This is presumably due of a simultaneous reflexion on another net plane (Hart & Lang, 1961). In view of these circumstances, the measurements were carried out along a set of two parallel lines, each running on one side of, and at about 1 mm apart from, the twin boundary dividing the crystal. One of the two parts is denoted by *A* and the other by *B*. The regions in which the fringes were deformed appreciably were carefully avoided as far as possible in the measurements.

Fringe spacings of both the maxima and the minima were measured by similar procedures to those described in the paper of Hattori, Kuriyama, Katagawa & Kato (1965). In this experiment, however, the original plates were used instead of the magnified plates. Since the size of the specimens was about 10 mm along the edge, the wedge angles along the lines of the measurement can be assumed safely to be equal to each other. This point was confirmed experimentally by comparing the wedge angles of the section patterns for the parts *A* and *B*.

3. Experimental results

From careful measurements it was evident that the fringe systems of the parts *A* and *B* of specimen (*a*) were exactly parallel to each other, whereas the fringe systems in specimens (*b*) and (*c*) were inclined by angles of 20 minutes and 40 minutes respectively. So far, we cannot explain properly the origin of such a difference. For this reason, only specimen (*a*) was used for the extensive studies on the various net planes. Since, however, the inclination was very small, specimens (*b*) and (*c*) were also used for comparison with the results obtained from specimen (*a*), as will be explained later.

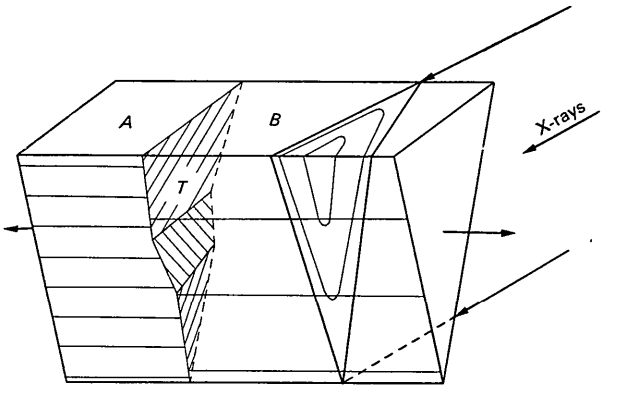

Fig. 1. The relations between the wedge-shaped twinning crystal and the X-ray intensity fields on the exit surface. *A* and *B* are the parts divided by the twin boundary *T*. The horizontal arrows indicate the traverse direction.

The net planes of the type $\{h0,l\}$ are specifically studied with specimen (*a*)*. As easily expected from the twin relation, the topographs due to $h0,l$ and $h0,\bar{l}$ reflexions can be recorded on a plate simultaneously corresponding to the parts *A* and *B*. The fringe spacings are listed in the column headed Λ of Table 1. Here the figures after the double sign imply the probable errors calculated on the assumption that the fringes should be equally spaced. In the column headed Λ_{corr}, the spacings corrected for the polarization effects are listed. The principles of the correction will be explained in § 4. The figures in the column headed *N* are the numbers of the fringe maxima and minima used for obtaining the spacing. The ratios, *P*, of the structure factors defined by equation (3), namely the ratios of the corrected values Λ_{corr}, are listed in Table 2. For comparison, the corresponding values calculated from the structure factors of Young & Post (1962), and those of Zachariasen & Plettinger (1965) are also included in Table 2.

Table 1. *The observed and corrected fringe spacings*

Net plane	Λ (μ)	Λ_{corr} (μ)	*N*
10,1	$73{\cdot}88_3 \pm 0{\cdot}02_6$	$73{\cdot}89_4 \pm 0{\cdot}02_6$	54
10,$\bar{1}$	$112{\cdot}36_4 \pm 0{\cdot}05_4$	$112{\cdot}37_5 \pm 0{\cdot}05_4$	36
10,2	$233{\cdot}3_2 \pm 0{\cdot}2_1$	$233{\cdot}4_6 \pm 0{\cdot}02_2$	20
10,$\bar{2}$	$473{\cdot}5_3 \pm 0{\cdot}8_7$	$473{\cdot}7_1 \pm 0{\cdot}8_7$	8
10,4	$233{\cdot}3_9 \pm 0{\cdot}2_6$	$232{\cdot}9_5 \pm 0{\cdot}1_9$	20
10,$\bar{4}$	$434{\cdot}5_0 \pm 0{\cdot}2_6$	$438{\cdot}1_0 \pm 0{\cdot}4_0$	6
10,5	$422{\cdot}3_7 \pm 2{\cdot}8_3$	$414{\cdot}9_4 \pm 1{\cdot}6_7$	8
10,$\bar{5}$	$252{\cdot}8_5 \pm 0{\cdot}6_8$	$253{\cdot}1_6 \pm 0{\cdot}2_8$	18
20,1	$110{\cdot}1_8 \pm 0{\cdot}1_5$	$110{\cdot}3_0 \pm 0{\cdot}1_5$	16
20,$\bar{1}$	$172{\cdot}7_8 \pm 0{\cdot}1_6$	$172{\cdot}9_2 \pm 0{\cdot}1_7$	12
20,2	$308{\cdot}5_8 \pm 0{\cdot}7_8$	$309{\cdot}3_2 \pm 0{\cdot}6_8$	12
20,$\bar{2}$	$146{\cdot}46_7 \pm 0{\cdot}12_8$	$146{\cdot}17_4 \pm 0{\cdot}09_8$	24
20,3	$115{\cdot}16_2 \pm 0{\cdot}04_6$	$115{\cdot}23_4 \pm 0{\cdot}04_2$	40
20,$\bar{3}$	$224{\cdot}7_4 \pm 0{\cdot}2_0$	$223{\cdot}1_7 \pm 0{\cdot}1_5$	20

In order to check our results, the following experiments were performed with respect to $\{10,1\}$ reflexions. Hereafter, 10,1 and 10,$\bar{1}$ are denoted by *R* and *r*, respectively.

(i) Using the same specimen, we took a pair of topographs in which part *A* gave the *R* reflexion or the *r* reflexion. Hereafter, these conditions will be denoted by *A*(*R*) and *A*(*r*), respectively. Obviously, conditions *A*(*R*) and *A*(*r*) are equivalent to conditions *B*(*r*) and *B*(*R*), respectively.

(ii) By the use of different specimens the ratios of the structure factors were compared with one another. The results are listed in Table 3. From these, we notice that the agreement between the independent measurements are very satisfactory. The discrepancy between *A*(*R*) and *A*(*r*) conditions in specimen (*a*) is less than 0·1%. Even for the most unfavourable specimen (*c*) in which plate-like defects could not be avoided, the agreement was within about 0·5%. In specimen (*b*), plate-like defects were actually included, as seen very faintly in Fig. 3. For this reason *A*(*R*) condition was not studied with this.

Table 3. *The ratio* $P=|F_{10,1}|/|F_{10,\bar{1}}|$ *for different geometrical conditions and different specimens*

Specimen	Geometry	P
(*a*)	*A*(*r*)	$0{\cdot}6575_6 \pm 0{\cdot}0003_7$
	A(*R*)	$0{\cdot}6571_3 \pm 0{\cdot}0004_1$
(*b*)	*A*(*r*)	$0{\cdot}6596_5 \pm 0{\cdot}0004_4$
(*c*)	*A*(*r*)	$0{\cdot}6566_1 \pm 0{\cdot}0006_6$
	A(*R*)	$0{\cdot}6603_3 \pm 0{\cdot}0006_4$

The geometrical conditions *A*(*R*) and *A*(*r*) are explained in the text.

4. The causes of errors and the corrections

The effects of X-ray polarization

The two components of polarization of the incident X-rays produce two mutually incoherent fringe systems having slightly different spacings in the crystal. Owing to the beating of these fringe systems the fading of the fringes appears in the topograph patterns even for perfect crystals (Hart & Lang, 1965; Hattori, Kuriyama & Kato, 1965). The example is indicated by arrows in one of the photographs. The observed fringe spacing is given by the inverse arithmetical mean of the spacings of the two fringe systems. In addition, the observed fringe positions are not equally spaced in the vicinity of the fading region. This phenomenon has previously been discussed in detail in the case of section topographs (Hattori, Kuriyama & Kato, 1965).

For traverse patterns, the displacements of the fringes from the equally spaced positions are given by

$$\Delta l_m = \tan^{-1}[\tan(\tan^2\theta_B \,.\, l_m) \,. \{(\cos 2\theta_B)^{3/2}-1\}/\{(\cos 2\theta_B)^{3/2}+1\}] , \quad (4)$$

* As mentioned in the *Introduction*, the present method can be applied to *hk*,*l* reflexions in general without limitation.

Table 2. *The ratios of the structure factors,* $P=|F_{h0,l}|/|F_{h0,\bar{l}}|$

*h*0,*l*	Present (P_0)	Zachariasen & Plettinger P_{obs}	Zachariasen & Plettinger P_{corr}	Zachariasen & Plettinger P_{cal}	Young & Post P_{obs}	Young & Post P_{cal}
10,1	$0{\cdot}6575_6 \pm 0{\cdot}0003_7$	0·7973	0·6602	0·6528	0·6589	0·6465
10,2	$2{\cdot}029_1 \pm 0{\cdot}004_3$	1·849	2·118	2·108	1·652	1·983
10,4	$1{\cdot}880_7 \pm 0{\cdot}002_3$	1·666	1·830	1·876	1·687	1·809
10,5	$0{\cdot}610_1 \pm 0{\cdot}002_5$	0·648	0·601	0·589	0·638	0·604
20,1	$1{\cdot}567_7 \pm 0{\cdot}002_6$	1·456	1·547	1·539	1·407	1·577
20,2	$0{\cdot}472_6 \pm 0{\cdot}001_1$	0·540	0·481	0·491	0·550	0·475
20,3	$1{\cdot}936_3 \pm 0{\cdot}001_5$	1·633	1·949	1·917	2·176	1·851

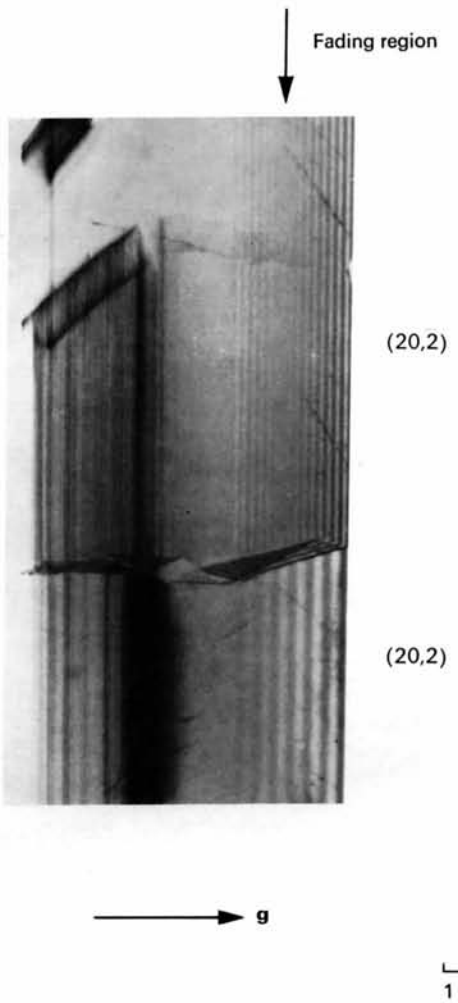

Fig. 2. The traverse topograph of specimen (*a*). A fading region is illustrated.

To face p, 234

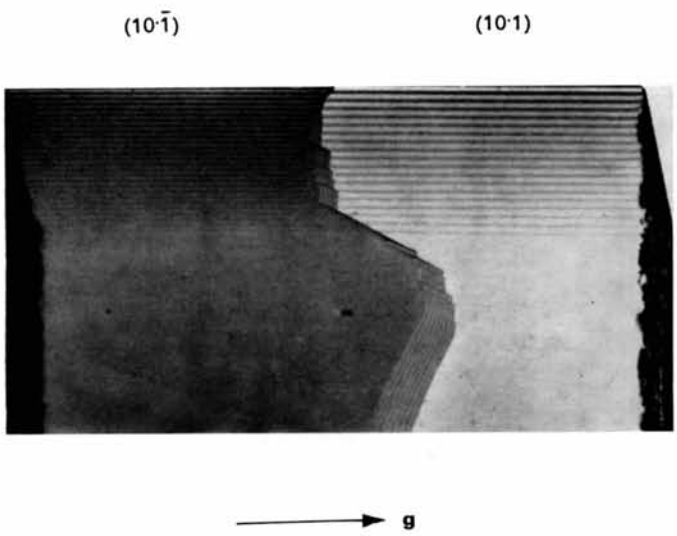

Fig. 3. Topograph of specimen (*b*).

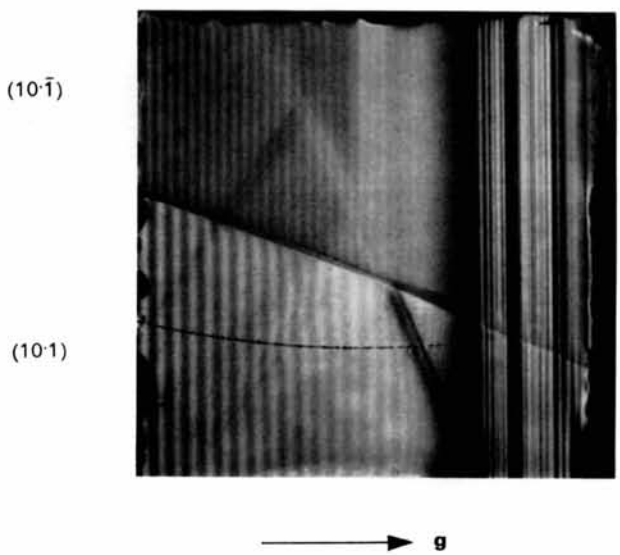

Fig. 4. Topograph of specimen (*c*).

where θ_B is the Bragg angle and l_m is the mth order extremum position on a normalized scale, defined practically by

$$l_m=(1+\cos 2\theta_B)A_m\,. \tag{5}$$

A_m is the value of Zachariasen's notation A (1945) at the position of the mth order fringe. A is defined by

$$A=(e^2/mc^2)|F_h|t/v(\gamma_0\gamma_h)^{\frac{1}{2}} \tag{6}$$

where e, m and c are usual physical constants, t the thickness of the crystal, v the volume of unit cell, and finally γ_0 and γ_h are the cosines of the angles between the normals of the incident and Bragg-reflected beams, respectively. Details of the arguments presented here will be given in a separate paper (Kato & Yamamoto, in preparation).

It should be noticed that the displacements of the maximum and minimum positions have the same sign in a particular group of the fringes between two fading regions, where $\{\tan^2\theta_B\,.\,l_m\}$ are $(m\pi\pm\pi/2)$ approximately. The sign changes before and after the fading region. The figures listed under Λ_{corr} of Table 2 are the fringe spacings corrected on the basis of this consideration. It is clear that the probable errors are reduced appreciably through this correction, particularly for higher order reflexions and the net planes with small structure factors.

Effects of absorption

Although the magnitude of $\mu_0 t$ (linear absorption coefficient times the crystal thickness) was less than about 0·5 in our specimens, the effects of absorption may not be neglected. From the intensity distribution for absorbing perfect crystals (Kato, 1967) it is possible to calculate the displacements of the maximum and minimum positions due to the absorption. Here, it should be noticed that the intensity profile of the fringes becomes asymmetric owing to the attenuation of X-ray intensity. The displacement of the extremum position is caused by this asymmetry, in such a way that the maxima displace towards the thinner part of the crystal, whereas the minima displace towards the thicker part. The displacements of the individual extrema amount to about a few tenths of one per cent of the distance l_m near the fading region for higher order reflexions.

As examples of the results, the values of Λ corrected for absorption and polarization were $253{\cdot}1_1\pm0{\cdot}35$, $309{\cdot}7_2\pm1{\cdot}2_0$ and $222{\cdot}7_6\pm0{\cdot}3_8$ for $10,\bar{5}$, $20,2$ and $20,\bar{3}$ reflexions, respectively. On the other hand, the polarization-corrected values Λ_{corr} were $253{\cdot}1_6\pm0{\cdot}2_8$, $309{\cdot}3_2\pm0{\cdot}6_8$ and $223{\cdot}1_7\pm0{\cdot}1_5$ in the respective cases, as listed in Table 1. Thus, it turns out that the correction for absorption is comparable to the probable error in the measurements of the fringe spacing. Here, however, it is to be noticed that the probable errors are slightly increased if the correction for absorption is taken into account. This implies that our consideration of the absorption effect may not be adequate for obtaining the true values of fringe spacings.

At the present moment, we interpret this fact as follows. Since we determined the fringe positions by a visual method, the observed fringe position is not exactly the extremum position of the fringe profile as assumed in the theoretical treatments of the correction. Since, as mentioned above, the displacements of the extrema are simply due to the asymmetry of the fringe profile, a visually determined position is more likely to be such a position where the oscillating part of the intensity on the smooth background curve, not the intensity itself, becomes extremum. In fact, the background curve is large in the traverse pattern, and attenuates with the increasing thickness of crystal. If this consideration is admitted, in practice we need not correct Λ values for the absorption effect. For this reason, and since the probable error is not actually decreased by the absorption-correction as mentioned above, the correction for absorption has been disregarded in this paper.

The distortion of the lattice

The lattice distortion is the most unfavourable factor to the present method of determining structure factors. Both theory (Kato, 1964*a*,*b*; Kato & Ando, 1966) and experiment (Hart, 1965, 1966; Ando & Kato, 1966) show that the fringe spacing decreases by a long-range distortion of lattice. The contraction of the spacing becomes predominant for net planes with small structure factors and in a thicker part of the crystal. Associated with this contraction of the fringe spacing, the background of the intensity distribution is changed, and the margin effects (Kato, 1960) in the section patterns become faint to some extent. In our present experiment, however, no evidence of such a long-range distortion of lattice is detected. In particular, the fringes were equally spaced except in the fading regions within the experimental error. Nevertheless, the fact mentioned in § 3 for specimen (*c*) indicates that this cause of error should not be neglected in some cases.

5. Discussion

Since we measured only the ratio of the structure factors, and the number and type of the net planes concerned are limited, it is not to be expected that direct information as to the charge distribution in the crystal will now be available. Nevertheless, because the accuracy of the measurement is higher than that in other experiments based on the conventional method, and because fewer ambiguities are involved in applying the theoretical formulae to determine $P=|F_{h0,l}|/|F_{h0,\bar{l}}|$, our results can be used for checking the conventional methods of crystal analysis. For recent work on the structure determination of α quartz, we take the experiments of Young & Post (1962) and of Zachariasen & Plettinger (1965)*. In their work the intensity data

* Hereafter we shall denote these two papers 'Y & P' and 'Z & P', respectively.

were collected with small spherical crystals. Here, it is to be emphasized that our intention is not to compare the errors in the data of the individual authors but simply to make clear the current state of the experiments based on the conventional method.

Now, we consider two sets of the values of corresponding structure factor ratios, say P_1 and P_2, of independent sources, and define a quantity D as a numerical measure of the mean discrepancy between the two sets of data, as

$$D=\left[\frac{1}{N}\Sigma\{(P_1/P_2)-1\}^2\right]^{\frac{1}{2}}, \qquad (7)$$

where $N=7$ is the number of the pairs of the net planes concerned and, the summation should be taken over seven pairs.

Table 4 gives the D values by putting $P_1=P$ and $P_2=P_0$ in equation (7), where P represents one set of P values and P_0 represents our values of the ratios listed in Table 2. The abbreviated subscripts of P and D in Table 4 and in the following correspond to those of the various $|F|$ values in the papers cited here. Their meanings will be explained respectively in due course.

Table 4. *The deviations D of the ratios P of other authors from the present result P_0*

Zachariasen & Plettinger			Young & Post	
D_{obs}	D_{corr}	D_{cal}	D_{obs}	D_{cal}
13·6%	2·2%	2·7%	11·9%	2·5%

$D=[\frac{1}{7}\Sigma\{(P/P_0)^2-1\}]^{\frac{1}{2}}$; P and P_0 are given in Table 2.

First of all, it is clear from D_{obs} that the observed $|F|$ values, $|F_{obs}|$, of Z & P and Y & P include errors of more than 10% for the low-order reflexions concerned. This result is not particularly surprising because extinction effects must be predominant and errors due to the mixing of twinned planes have not been eliminated in their $|F_{obs}|$ values.

In the paper of Z & P the corrected values $|F_{corr}|$ with respect to the extinction and the twinning are listed. The D_{corr} value corresponding to P_{corr} of Table 2 is much reduced after these corrections, down to about 2%. Thus, the correction formulae adopted by Z & P are significantly correct (see also Zachariasen, 1963). Incidentally, the D_{corr} value obtained from $|F_{corr}|$ values of Brill, Hermann & Peters (1942) amounts to 4·5%*. Nevertheless, recalling the fact that the values of P_{corr} should agree with our P_0 values in principle, we may say that the D_{corr} indicates the extent of the error in $|F_{corr}|$ values of Z & P.

Next, we shall consider D_{cal} values corresponding to the calculated values of the structure factors, $|F_{cal}|$. As is generally admitted, in the values $|F_{cal}|$ based on a proposed crystal structure, the effects on thermal vibrations are taken into account and the effects on a charge redistribution due to the bonding of atoms are partly considered through the temperature coefficient, β_{ij}. The difference between P_0 and P_{cal}, therefore, must be mainly due to the effects of the charge distortion which is not taken into account by β_{ij}-coefficients, provided that the proposed structure is satisfactorily correct.

In order to estimate this bonding effect, obviously the difference between $|F_{corr}|$ and $|F_{cal}|$ must be significantly larger than the error E involved in $|F_{corr}|-|F_{cal}|$, particularly in low-order reflexions. If it is so, we have $D_{cal}\gg E$ because, as mentioned above, it is anticipated that the amount of D_{cal}, namely the difference between P_{cal} and P_0, is due to the bonding effects. Although, as shown in Table 4, D_{cal} is actually larger than $D_{corr}\simeq$(the error in $|F_{corr}|)\simeq E$, such a small difference may not be significant. Since $|F_{corr}|$ is not listed in the paper of Y & P, we shall not discuss their data.

On the other hand, if we estimate the error in $|F_{cal}|$ by the mean deviation D defined by equation (7), using P_{cal} values of Z & P and of Y & P in place of P_1 and P_2 respectively, it turns out that the error in $|F_{cal}|$ amounts to 3·4%. With this estimation we are led to a pessimistic conclusion on a possibility of finding the bonding effects on the basis of their data.

As conclusions, the present analysis of various data implies that, at least for quartz, it is very necessary to determine individual structure factors $|F_{cal}|$ for low order reflexion with an accuracy better than 1% in order to obtain meaningful information on the bonding effect. The amounts of extinction effects and the errors due to twinning are estimated to be more than 10%. The correction formulae used by Zachariasen & Plettinger can reduce this value to 2%.

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* $|F|$ values in their paper can be interpreted to be $|F_{corr}|$ values in the present notation. They used milky single crystals for the intensity measurements.

References

ANDO, Y. & KATO, N. (1966). *Acta Cryst.* **21**, 284.

AUTHIER, A. & SAUVAGE, M. (1966). Colloque International sur les Dislocations, Toulouse.

BONSE, U. & HART, M. (1965). *Appl. Phys. Letters*, **6**, 155; **7**, 99.

BRILL, R., HERMANN, C. & PETERS, C. (1942). *Ann. Phys. Lpz.* **41**, 233.

EWALD, P. P. (1916*a*). *Ann. Phys. Lpz.* **49**, 1.

EWALD, P. P. (1916*b*). *Ann. Phys. Lpz.* **49**, 117.

EWALD, P. P. (1917). *Ann. Phys. Lpz.* **54**, 37. (See also *Handbuch der Physik*, **23** 2, p.285. Berlin: Springer, 1933.)

HART, M. (1965). *Appl. Phys. Letters*, **7**, 96.

HART, M. (1966). *Z. Physik*, **189**, 269.

HART, M. & LANG, A. R. (1961). *Phys. Rev. Letters*, **7**, 120.

HART, M. & LANG, A. R. (1965). *Acta Cryst.* **19**, 73.
HATTORI, H., KURIYAMA, H., KATAGAWA, Y. & KATO, N. (1965). *J. Phys. Soc. Japan*, **20**, 988.
HATTORI, H., KURIYAMA, H. & KATO, N. (1965). *J. Phys. Soc. Japan*, **20**, 1047.
KATO, N. (1960). *Acta Cryst.* **13**, 349.
KATO, N. (1961). *Acta Cryst.* **14**, 627.
KATO, N. (1964*a*). *J. Phys. Soc. Japan*, **19**, 67.
KATO, N. (1964*b*). *J. Phys. Soc. Japan*, **19**, 971.
KATO, N. (1967). *J. Appl. Phys.* In the press.
KATO, N. & ANDO, Y. (1966). *J. Phys. Soc. Japan*, **21**, 964.
KATO, N. & LANG, A. R. (1959). *Acta Cryst.* **12**, 787.
KATO, N., USAMI, K. & KATAGAWA, T. (1966). Read at Denver Conference. *Advanc. X-ray Analysis*, **10**, 46.
KATO, N. & TANEMURA, S. (1967). *Phys. Rev. Letters*, **19**, 22.
YOUNG, R. A. & POST, B. (1962). *Acta Cryst.* **15**, 337.
ZACHARIASEN, W. H. (1945). *Theory of X-ray Diffraction in Crystals*, Section III, p.123. New York: John Wiley.
ZACHARIASEN, W. H. (1963). *Acta Cryst.* **16**, 1139.
ZACHARIASEN, W. H. & PLETTINGER, H. A. (1965). *Acta Cryst.* **18**, 710.

Acta Cryst. (1968). A**24**, 237

Anomale inkohärente Streuung thermischer Neutronen bei Bildung stehender Neutronenwellen in nahezu idealen Kristallen von Kaliumdihydrogenphosphat (KDP)

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In nearly perfect crystals of KDP standing waves of thermal neutrons are produced, causing an anomalous weak or strong incoherent scattering by hydrogen nuclei in the 004 reflexion. At the double crystal diffractometer in Rossendorf for the observation of anomalous incoherent scattering rocking curves in the Laue-case were measured in the reflexion- and transmission directions. The observed effect is interpreted and computed by the dynamic interference theory. The results of the experiments show that the incoherent scattering gives a contribution to the imaginary part of the scattering amplitude; its value is a function of the chemical bonding of the H atoms.

Vorbemerkung

Bei Neutronenbeugung an nahezu idealen Einkristallen ohne Mosaikstruktur und mit nur geringer Versetzungsdichte kommt es durch Bildung stehender Neutronenwellen zu speziellen Interferenzerscheinungen wie z.B. der anomalen Absorption und dem Pendellösungseffekt, die auf der Grundlage der von Ewald (1917) vorgelegten dynamischen Interferenztheorie erklärt und berechnet werden können. Die anomale Absorption, die von Borrmann (1941) mit Röntgenstrahlen entdeckt wurde, und der Pendellösungseffekt (Ewald, 1917; Kato & Lang, 1959) wurden von Sippel, Kleinstück & Schulze (1962, 1964, 1965) bereits in früheren Arbeiten auch mit Neutronen untersucht. Während diese Interferenzerscheinungen leichter mit Röntgenstrahlen zu beobachten sind, ist der in vorliegender Arbeit gefundene Effekt vor allem mit Neutronen gut zu finden, da sie an Atomkernen mit Spin für parallele und antiparallele Streuung erheblich unterschiedliche Streuamplituden und dadurch eine starke inkohärente Streuung aufweisen können. Bei Wasserstoffkernen unterscheiden sich die beiden Streuamplituden für parallele und antiparallele Streuung auch in ihrem Vorzeichen. Deshalb ist die Streuung der H-Kerne im Kristall bis zu 97% inkohärent.

Die anomale inkohärente Streuung lässt sich analog zum Effekt der anomalen Absorption interpretieren. Die stehenden Neutronenwellenfelder werden dabei nicht durch die Absorption der Atome, sondern durch die inkohärente Streuung im Kristall anomal geschwächt. Die anomale inkohärente Streuung liefert einen erneuten Nachweis der stehenden Neutronenwellenfelder in Kristallen unabhängig von der Absorption und zeigt den Einfluss der stehenden Neutronenwellen auch auf die inkohärente Streuung.

Experimentelles

Bei Versuchen mit einem 14,7 mm dicken KDP-Kristall als zweiten Kristall auf dem in Fig. 1 schematisch gezeigten Neutronen-Doppelkristall-Diffraktometer wurden in symmetrischer Laue-Stellung bei 004-Reflexion die Rockingkurven in Fig. 2 erhalten. In der