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The Influence of In-Plane Collimation on the Precision and Accuracy of Lattice-Constant Determination by the Bond Method. II. Verification of the Mathematical Model. Systematic Errors

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Abstract

The mathematical model described in part I [Urbanowicz (1981). *Acta Cryst.* A**37**, 364–368] has been verified by measurements performed on the Bond diffractometer constructed in the author's institute. A computer program has been written allowing the synthesis of the diffraction profile $h(\omega)$ for an arbitrary original function $f(\omega)$, distribution of tube-focus emissivity $i_s(x)$, collimator length L_2, and for given widths d_1, d_2 of collimator slits. The calculations have been performed to estimate systematic errors related to the collimation conditions, caused by insufficient accuracy (shift Δs) of the collimator setting in relation to the focus centre and asymmetry function φ of the distribution of tube-focus emissivity. A simplified formula is given for fast estimation of the errors caused by overlapping of the $K\alpha_1$–$K\alpha_2$ doublet and by the Lp factor.

1. Introduction

The accuracy $\Delta d/d$ of the determination of the lattice spacing d (and thus the lattice constants) depends on the systematic error of the Bragg angle determination which is expressed by the formula

$$\Delta d/d = -\cot\,\theta\Delta\theta. \tag{1}$$

In the Bond (1960) method the angle θ is based on the measurement of positions of peaks ω_0' and ω_0'' in two diffraction profiles, so

$$\Delta\theta = (\Delta\omega_0' + \Delta\omega_0'')/2 = \Delta\omega. \tag{1a}$$

The total error $\Delta\omega$ consists of a number of errors caused by various physical (*e.g.* Lp, refraction) and experimental (*e.g.* specimen tilt) factors. The present paper deals with systematic errors connected with collimation conditions. Since, as a rule, the values of the errors increase with widening of the collimator slits (detailed calculations of this dependence are given in the paper), it would be advantageous to use slits as narrow as possible. When, however, intensity of radiation is too small, wider slits should be used, as is discussed in part I (Urbanowicz, 1981). In this case it is necessary to take into account the dependence of systematic errors on the slit widths. The determination of the influence of collimation parameters on the shape of the measured profile is carried out on a mathematical model and verified for experimental data.

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2. Experimental data

An example will be presented of the application of the mathematical model and its verification with measurements on the Bond diffractometer constructed in this institute (Łukaszewicz, Kucharczyk, Malinowski & Pietraszko, 1978). In the present work the results of test measurements by Łukaszewicz, Pietraszko, Kucharczyk, Malinowski, Stępień-Damm & Urbanowicz (1976) are used. In these measurements the collimator parameters (Fig. 1) were $L_1 = 54$ mm, $L_2 = 297$ mm, and all 25 combinations of d_1 and d_2 with the nominal values d_1, d_2 = 0·05, 0·10, 0·25, 0·5 and 1 mm were used. Cu $K\alpha_1$ radiation was used from a tube with a focal spot size of 1 × 1 mm. The projection of the distribution of tube-focus emissivity was obtained by measurement and is shown in Fig. 2.

The measurement data consist of two groups.

I. The distribution of the primary beam intensity $i(x)$ along the X axis in the form of $i(\beta)$ dependence [$i(\beta)$ is the angle of rotation of the counter] measured by scanning of the primary beam (without a crystal) with the counter with a narrow slit, $d_c = 0\cdot05$ mm. The radius of the counter arm was $R = 75$ mm, the distance L_z (Fig. 1) was 175 mm.

II. The profiles of the 444 diffraction peak for a given silicon single crystal were measured with a counting time $t = 4$ s.

3. The synthesis of $h(\omega)$ profiles and its verification. The $i_s(x)$ functions

The shape of $f(\omega)$ was found by deconvolution of experimental profiles $h(\omega)$ and theoretically derived functions $g(\omega)$ for four combinations of the narrowest

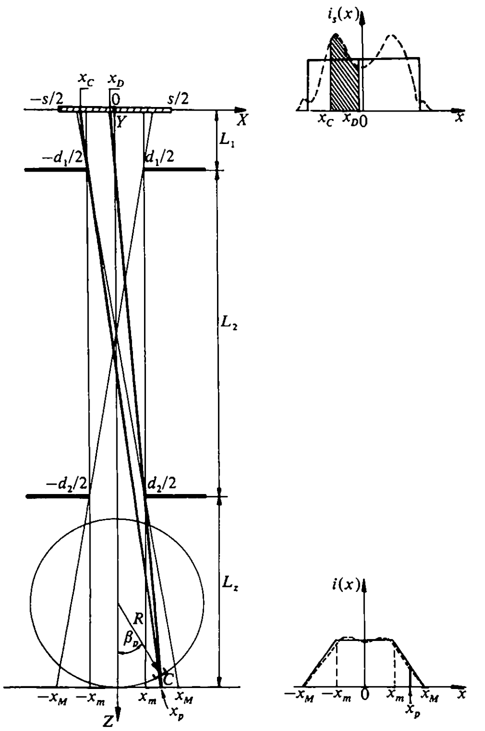

Fig. 1. Theoretical determination of the intensity distribution $i(x)$ of the primary beam.

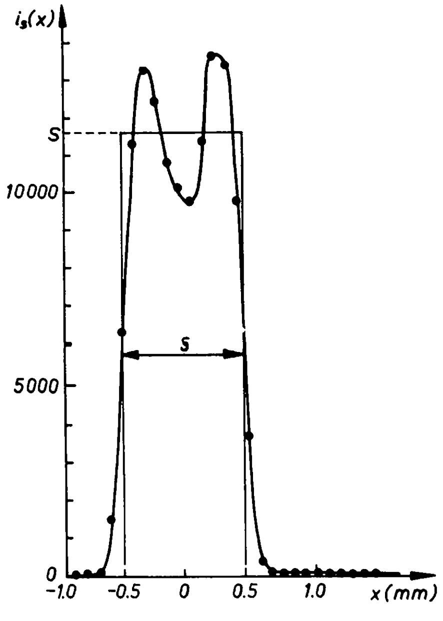

Fig. 2. The tube-focus emissivity, $i_s(x)$, obtained experimentally, with one collimator slit, $d_2 = 0{\cdot}05$ mm.

d_1, d_2. Stokes's (1948) method of deconvolution was applied. Next, the synthesis of $h(\omega)$ profiles for all 25 combinations of d_1 and d_2 was carried out by the Algol computer program $E40$.

Comparison of theoretical $h(\omega)_t$ with experimental profiles $h(\omega)_e$ showed that the former have slightly smaller half-widths; also the theoretical height of the peaks for big slits was greater (by up to 35%) than the experimental one. To explain these differences it was necessary to take into account the intensity of a primary beam in the cross section perpendicular to the collimator axis. The analysis of experimental distributions of primary beam intensity and distributions calculated for given parameters of collimation allowed suitable corrections taking into account

(1) scattering of radiation by the collimator edges,

(2) shift of the focus centre in relation to the collimator axis.

Calculations of the distribution of the primary beam intensity were carried out with an Algol computer program $E34$. The theoretical form of $i(x)$ was determined in a similar way to that of Alexander (1950), *i.e.* assuming the primary beam intensity at a given point $(x_p, L_1 + L_2 + L_z)$ to be directly proportional to the number of quanta emitted from a part of the focus surface (here the sector x_D–x_C) seen through the collimator from this point (Fig. 1):

$$i(x_p) = k_2 \int_{x_C(x_p)}^{x_D(x_p)} i_s(x')\,dx', \qquad (2)$$

where k_2 is a coefficient of proportionality depending on a distance $z = L_1 + L_2 + L_z$ from the focus, $k_2 \simeq 1/z^2$,

$$x_C = \max\{-[1 + L_1/(L_2 + L_z)]\,d_1/2 - xL_1/(L_2 + L_z), -[1 + (L_1 + L_2)/L_z]\,d_2/2 - x(L_1 + L_2)/L_z, -s/2\} \qquad (3a)$$

$$x_D = \min\{[1 + L_1/(L_2 + L_z)]\,d_1/2 - xL_1/(L_2 + L_z), [1 + (L_1 + L_2)/L_z]\,d_2/2 - x(L_1 + L_2)/L_z, s/2\}, \qquad (3b)$$

$$x_D - x_C > 0. \qquad (3c)$$

While comparing theoretical and experimental values $i(x)$, two basic differences were observed:

I. The maximal range r_T of the primary beam determined theoretically was smaller than r_M indicated by measurements, and their ratio $m = r_M/r_T$ was to a good approximation independent of d_1 and d_2 and was estimated as $1{\cdot}25 \pm 0{\cdot}05$. This effect originated from dispersion effects at the collimator edges (a guard aperture was not used) and absorption by air.

II. For higher values of d_1 and d_2 the experimental profiles $i(x)_M$ were asymmetrical which, as was found theoretically, resulted both from asymmetry of the $i_s(x)$ function (Fig. 2), and from a shift Δs of the centre of focus projection in relation to the collimator axis (Fig. 3). The value of the shift, $\Delta s = 0{\cdot}25 \pm 0{\cdot}05$ mm, was estimated, for the experimental data taken into account, by a trial-and-error method.

These two corrections, m and Δs, were taken into account at repeated calculations of $g(\alpha)$ and $f(\omega)$ for the synthesis of $h(\omega)$ profiles. The profiles calculated for the second time showed good agreement with experiment. Figs. 4 and 5 present examples of $i(\beta)$ and $h(\omega)$ for selected widths of collimator slits after the corrections have been introduced.

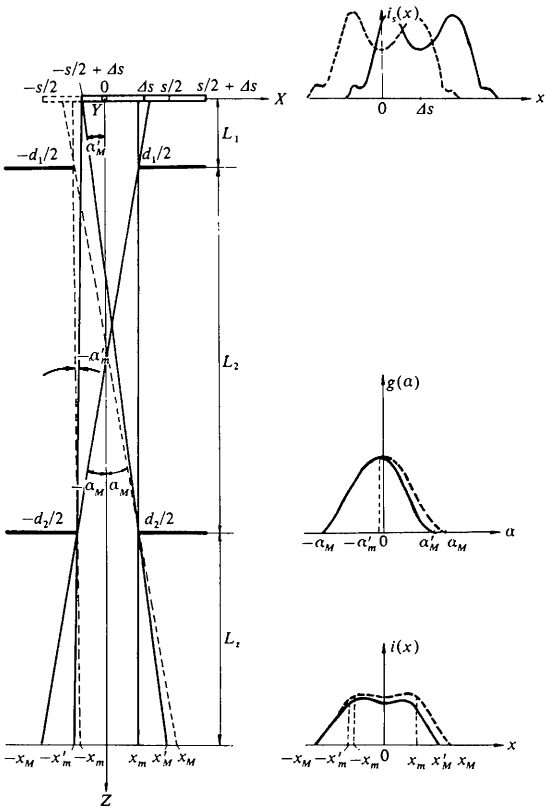

Fig. 3. The influence of the focal spot shift, Δs, on the course of the primary beam.

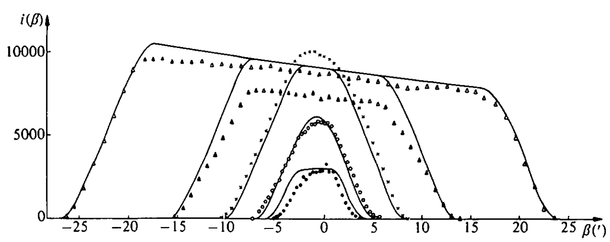

Fig. 4. The experimental functions $i(\beta)$ (points) compared with theory (continuous line), calculated with a focal spot shift $\Delta s = 0{\cdot}2$ mm and with the factor of ω scale $m = 1{\cdot}25$. $d_1 = 0{\cdot}5$ mm = constant; ● $d_2 = 0{\cdot}05$ mm; ○ $d_2 = 0{\cdot}10$ mm; × $d_2 = 0{\cdot}25$ mm; ▲ $d_2 = 0{\cdot}50$ mm; △ $d_2 = 1{\cdot}00$ mm.

4. Systematic errors due to shift of focus and asymmetry of the distribution of tube-focus emissivity

If we assume that the distribution of the tube-focus emissivity is symmetrical about the collimator axis, then the distribution of $g(\alpha)$ will also be symmetrical about this axis, and the position of the peak of the $h(\omega)$ function will not change in relation to the position of the peak in the original function $f(\omega)$. An example of a symmetrical distribution of tube-focus emissivity is given by Čermak (1960). In practice, however, the function $i_s(x)$ is most likely to be asymmetrical (*e.g.* Wilson, 1963). Hubbard & Mauer (1976) consider this asymmetry by estimating the apparatus function [$g(\alpha)$ according to symbols used in this paper] by means of a gate function.

In the present work the calculations were carried out taking into account the real shape of $i_s(x)$ determined experimentally (Fig. 2). This distribution, $i_s(x)$, can be presented in good approximation as

$$i_s(x) = (1 + \varphi x)\, i_s(x)_{\text{sym}}, \tag{4}$$

where $i_s(x)_{\text{sym}}$ is an idealized symmetrical function and φ is an asymmetry coefficient. For the experimental data used in this paper φ was estimated to be $0{\cdot}02$.

The synthesis of $h(\omega)$ profiles for all the combinations of d_1, d_2 slit widths was carried out with $\varphi = 0{\cdot}02$ and $0{\cdot}05$. With the procedure of extrapolated peak position a peak was each time determined. The results

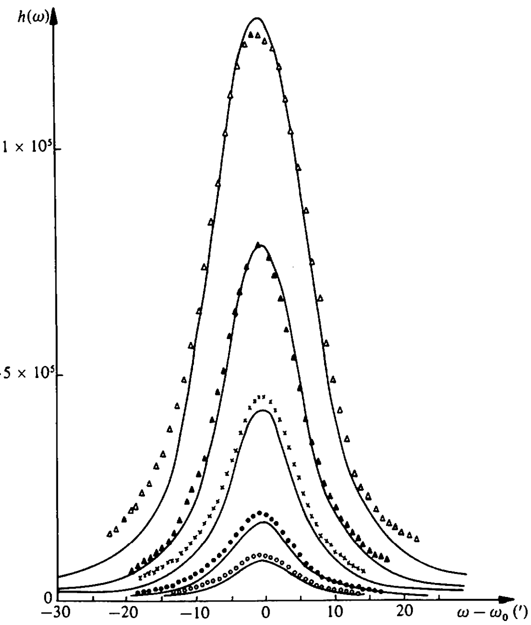

Fig. 5. The experimental profiles $h(\omega)$ (points) compared with theory (continuous line), calculated with a focal spot shift $\Delta s = 0{\cdot}2$ mm and with the factor of ω scale $m = 1{\cdot}25$. $d_1 = 0{\cdot}5$ mm = constant; ○ $d_2 = 0{\cdot}05$ mm; ● $d_2 = 0{\cdot}10$ mm; × $d_2 = 0{\cdot}25$ mm; ▲ $d_2 = 0{\cdot}50$ mm; △ $d_2 = 1{\cdot}00$ mm.

of the test for $\varphi = 0{\cdot}02$ are given in Table 1. The ideal position of the peak is treated as the zero point. It can be seen that the value of the error depends on the width of the collimator slits and that the slit d_1 (nearer to the focus) has a greater influence on the error. Analogous calculations were carried out assuming $i_s(x)_{\rm sym}$ to have the form of a gate function. It was found that the errors obtained from previous calculations were about twice as big as those estimated on this assumption (Fig. 6). The real shape of the $i_s(x)$ function (for $\varphi \neq 0$) can thus be a source of additional systematic error. Additional systematic error may also result from the shift of collimator axis in relation to the focus centre. The consequences of this shift are mentioned in § 3.

Syntheses of $h(\omega)$ profiles for $\Delta s = 0{\cdot}1$ and $0{\cdot}2$ mm at the focus size 1 mm were carried out with the program $E40$. The plots in Fig. 7 show that the shift $\Delta\omega$ depends strongly on the slit sizes.

Table 1. *The shift $\Delta\omega_0('')$ of the peak position caused by asymmetry of the $i_s(x)$ distribution*

$\varphi = 0{\cdot}02$

d_2 (mm)	d_1 (mm) 0·05	0·10	0·25	0·5	1·00
0·05	0·00	−0·02	−0·18	−0·87	−2·78
0·10	−0·01	−0·04	−0·20	−1·04	−2·81
0·25	−0·04	−0·06	−0·22	−1·04	−2·86
0·5	−0·11	−0·16	−0·33	−1·14	−2·64
1·00	−0·77	−0·75	−0·95	−1·89	−2·40

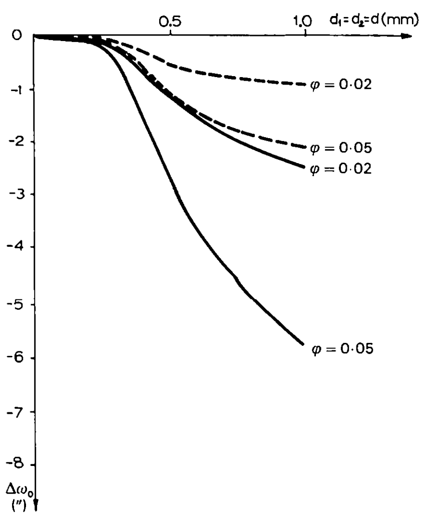

Fig. 6. The influence of the asymmetry coefficient φ on the shift $\Delta\omega_0$ of the peak position ω_0. The broken line relates to the case when $i_s(x)$ is approximated by a gate function.

5. Lorentz–polarization error

Bond (1960) gives the correction for the Lp factor assuming that the profile has the form of a Cauchy function. When $h(\omega)$ is considered to be the convolution of $f(\omega)$ in the form of the Cauchy function and $g(\omega)$ in the form of a trapezium (part I, equations 10, 14, 16), the correction $(\Delta\omega_0)_{\rm Lp}$ to be added to ω_0 is given by

$$(\Delta\omega_0)_{\rm Lp} = \left(\frac{\omega_f}{2}\right)^2 W(x_1,x_2) \times 2\cot 2\theta \frac{2+\sin^2 2\theta}{2-\sin^2 2\theta}, \quad (5)$$

where

$$W(x_1,x_2) = [x_1 \tan^{-1} x_1 - \tfrac{1}{2}\ln(1+x_1^2) - x_2\tan^{-1} x_2 + \tfrac{1}{2}\ln(1+x_2^2)] \times [(x_1^2-x_2^2)/(1+x_1^2)(1+x_2^2)]^{-1} \quad (5a)$$

$$x_1 = (d_1+d_2)/(2L_2\,\omega_f) \quad (5b)$$

$$x_2 = (d_1-d_2)/(2L_2\,\omega_f). \quad (5c)$$

For d_1 and d_2 tending to 0 the factor $W(x_1,x_2)$ tends to $\frac{1}{2}$, *i.e.* a value similar to that given by Bond (1960, *erratum*).

When the optimal (from the point of view of statistical errors) values of collimator parameters, discussed in part I, are used, the factor $W(1{\cdot}13, 0) = 1{\cdot}487$, so the value $(\Delta\omega_0)_{\rm Lp}$ is about $1{\cdot}5$ times larger than that given by Bond for $h(\omega)$ in the Cauchy form.

6. Effect of the $K\alpha$ doublet overlapping

For the large θ angles preferable in the Bond method there is no trouble in resolving the components of the

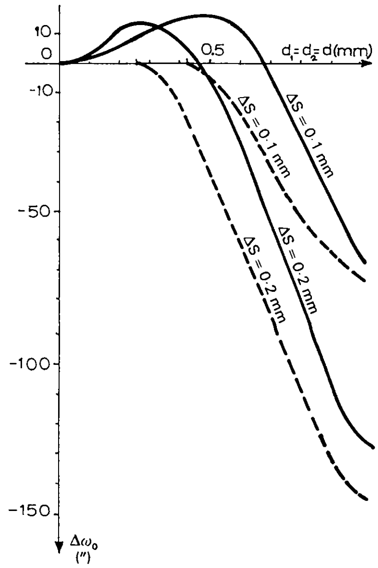

Fig. 7. The influence of the shift Δs on the shift $\Delta\omega_0$ of the peak position ω_0. The broken line relates to the case when $i_s(x)$ is approximated by a gate function.

$K\alpha$ doublet. However, for accurate determination of the peak position, it is necessary to take into consideration the effect of overlapping of the two profiles $h_1(x)$ and $h_2(x)$ connected with the $K\alpha_1$ and $K\alpha_2$ radiation. This effect depends on collimation parameters.

Let us assume (Delhez & Mittemeijer, 1975*a*,*b*) that

(1) both profiles have the same shape,

(2) their intensity ratio, $M = h_2(x)_{\max}/h_1(x)_{\max}$, is known,

(3) the doublet separation, δ, is also known.

Since, in the present work, the substitution $x = 2(\omega - \omega_0)/\omega_f$ is used, δ is determined as

$$\delta = 2\Delta/\omega_f, \tag{6}$$

where Δ is the distance between the two peak positions on the ω scale.

The measured profile $h(x)$ is the sum

$$h(x) = h_1(x) + Mh_1(x - \delta). \tag{7}$$

The peak position can be determined from

$$h_1'(x) = -Mh_1'(x - \delta). \tag{8}$$

When the $h_1(x)$ has the form presented in part I (Urbanowicz, 1981, equation 18),

$$h_1(x) = Ax_4[\tan^{-1}(x + x_3) - \tan^{-1}(x - x_3)],$$

the shift $\Delta\omega_0$ of the peak position of $h(\omega)$ in relation to the peak position of $h_1(\omega)$, with the assumptions $\delta^2 \gg 1$, $\delta^2 \gg x_3^2$, $x^2 \ll 1$, can be written as

$$\Delta\omega_0 = M\frac{\omega_f}{2}\frac{(1 + x_3^2)^2}{\delta^3}. \tag{9}$$

Similarly, when $h_1(x)$ has the form given in equation (16) of part I, the correction is

$$\Delta\omega_0 = M\frac{\omega_f}{2}\frac{(1 + x_1^2)(1 + x_2^2)}{\delta^3}. \tag{10}$$

For the 444 reflection of a silicon crystal $\Delta = 2640''$. For the example presented earlier, when $x_1 = 1\cdot13$, $x_2 = 0$, $\omega_f = 456''$, assuming that $M = 0\cdot5$, we have

$$\Delta\omega_0 = 0\cdot5 \times \frac{456}{2} \times \frac{1 + 1\cdot13^2}{12\cdot39^3} = 0\cdot14'' \text{ (from 10)}$$

or

$$\Delta\omega_0 = 0\cdot5 \times \frac{456}{2} \times \frac{(1 + 1\cdot13^2/2)^2}{12\cdot39^3} = 0\cdot16''. \text{ (from 9)}$$

All the computer programs mentioned in this paper were written by the author and may be obtained on request.

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