$$\left[\frac{\partial}{\partial q_z}\det m_H(\mathbf{q})\right]_{q_t=p_{0,t};\ q_z=\alpha_i} = 4(-1)^i\alpha_i R \qquad (A.15)$$

$$[\Omega(\mathbf{q})]_{q_t=p_{0,t};\ q_z=\alpha_i} = [\Omega(\mathbf{q}-\mathbf{H})]_{q_t=p_{0,t};\ \alpha_i+H_z} = \frac{1}{\tau}\,[\varepsilon+(-1)^i R] \qquad (A.16)$$

$$[\Omega(\mathbf{q}+\mathbf{H})]_{q_t=p_{0,t};\ q_z=\alpha_i} = -[\varepsilon-(-1)^i R]\,. \qquad (A.17)$$

Substituting these results in (A.6) to (A.8) we can obtain equations (3·13) to (3·16) for dynamical field functions.

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Some Properties of the Single-Crystal Rocking Curve in the Bragg Case

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In the present article (i) a new formula for the single-crystal rocking curve for both the centrosymmetric and polar Bragg case is given; (ii) a geometrical construction is derived, whence many properties of the rocking curve are easily visualized; (iii) the different analytical expressions introduced by several authors are shown to be a consequence of different definitions of the variable 'y'.

1. Introduction

The rocking curve (RC) for perfect crystals in the Bragg case has been discussed by many authors. A list of references may be found in well-known books (*e.g.* Zachariasen, 1945; Laue, 1960; James, 1950) or in the articles of Hirsch & Ramachandran (1950), Batterman & Cole (1964) and Bucksch, Otto & Renninger (1967). Although many of these authors follow the treatment of Zachariasen, the expressions in the 'normalized variable y'* differ slightly one from the other.

The first difference was pointed out by Hirsch & Ramachandran (1950). Their expression is more general than that of Zachariasen, and the discrepancy is shown to be a consequence of some approximations. Cole & Stemple (1962) derived a general expression for both the centrosymmetric and polar case and Bucksch, Otto & Renninger (1967) made a physical analysis of it. In their paper (p. 508) one reads (in a loose translation) – 'For small g and κ† the tails of the RC approach the Darwin curve asymptotically from below. But for greater g and κ they rise above this curve.'

On the other hand, Battermann & Cole (1964, p. 707) state 'The Darwin–Prins curve matches the Darwin curve only at a point close to the low angle slope of the peak. It is less than the Darwin curve at all other points'. Since they also use some approximations (κ) the last statement seems to be their consequence. These two statements are rather contradictory. Therefore in the first part of the present article we shall follow Batterman & Cole's (1964) (hereafter BC) derivation, showing that their statement is quite general, and we shall derive a modified formula for the RC.

In the second part we shall give a simple geometrical construction by means of which one can visualize many interesting properties of the RC.

Finally we shall show, that by redefining the 'normalized variable y' by means of a linear transformation, one gets the formulae of Hirsch & Ramachandran and Cole & Stemple, and thus the statement of Bucksch, Otto & Renninger will be brought into agreement with that of BC.

2. Derivation of the general formula

By definition the RC is the ratio of the diffracted and incident power plotted as a function of deviation from the Bragg angle $\theta-\theta_B$, *i.e.*

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* By 'normalized y' we mean a linear transform of the deviation from the Bragg angle such that the range of total reflexion lies between $y=-1$ and $y=1$

† Defined as

$$\kappa = \left|\frac{F_H''}{F_{H'}}\right|,\quad g = -\tfrac{1}{2}\,\frac{1-b}{|b|^{1/2}|P|}\cdot\frac{F_0''}{|F_{H'}|}\,.$$

$$C_H^p(\theta-\theta_B)=\frac{P_H(\theta-\theta_B)}{P_0}.$$

If we denote by S_0 and S_H the cross sections of the incident and diffracted beams respectively, we have

$$C_H^p=(S_H\, I_H)/(S_0\, I_0)=(S_H/S_0)\,.\,|(E_H/E_0)|^2,$$

where E_H and E_0 are the respective field amplitudes. Introducing the usual parameter $b=\gamma_0/\gamma_H$, where γ_i are the direction cosines of the respective beams, one may show that $S_H/S_0=|b|^{-1}$. Following BC, p. 706, equation 103, we get

$$C_H^p=|F_H/F_{\bar H}|\;|\eta\pm(\eta^2-1)^{1/2}|^2,$$

(with the sign chosen in such a way that $C_H^p\to 0$ for $|\eta|\to\infty$), where F_H is the structure factor (generally complex, including the Hönl corrections), and

$$\eta=\frac{-b(\theta-\theta_B)\,\sin 2\theta_B-\frac{1}{2}\Gamma F_0(1-b)}{\Gamma|P|\;|b|^{1/2}(F_H\,F_{\bar H})^{1/2}} \tag{1}$$

with the same notation as in *BC*. (The only difference is the change of sign in η, making η and θ increase in the same sense). Here P is the polarization factor (1 or $\cos 2\theta_B$), $\Gamma=r_e\;\lambda^2/(\pi\,V)$ where $r_e=e^2/(4\pi\varepsilon_0\,mc^2)$ and V the unit cell volume.

As η is invariant under the transformation $H\to\bar H$, we may write

$$C_H^p=|F_H/F_{\bar H}|C_H \tag{2}$$

with

$$C_H=|\eta\pm(\eta^2-1)^{1/2}|^2=C_{\bar H} \tag{3}$$

Thus the shape of both C_H^p and $C_{\bar H}^p$ is equal and their ratio is

$$C_H^p/C_{\bar H}^p=|F_H/F_{\bar H}|^2.$$

Therefore in the following we shall deal with C_H only. Using $\xi=(\eta^2-1)^{1/2}$, which implies $\eta^2-\xi^2=1$, we have (with * denoting the complex conjugate)

$$C_H=(\eta\pm\xi)\,(\eta^*\pm\xi^*)=|\eta|^2+|\xi|^2\pm(\xi\eta^*+\eta\xi^*).$$

But

$$(\eta^2-\xi^2)\,(\eta^{*2}-\xi^{*2})=1$$

wherefrom

$$(\xi\eta^*+\eta\xi^*)^2=(|\eta|^2+|\xi|^2)^2-1$$

If we define $L=|\eta|^2+|\xi|^2$, we get the important formula

$$C_H=L-(L^2-1)^{1/2}\,\dagger \tag{4}$$

first obtained by Miller (1935).

Further

$$|\xi|^2=[(\eta^2-1)\,(\eta^{*2}-1)]^{1/2}$$

gives

$$|\xi|^2=[(|\eta|^2-1)^2+4\eta''^2]^{1/2},$$

where

$$\eta''=\mathrm{Im}(\eta).$$

This gives the result

$$L=|\eta|^2+[(|\eta|^2-1)^2+4\eta^{2''}]^{1/2}. \tag{5}$$

† Here again, if we use $\xi\eta^*+\eta\xi^*=+(L^2-1)^{1/2}$, we must take the minus sign for the same reason as above.

Now we return to the definition of η [equation (1)]. The denominator is generally complex, and we may therefore write (with $\varphi=\theta-\theta_B$) $\eta=\alpha\varphi+\beta$

with

$$\alpha=\frac{-b\,\sin 2\theta_B}{\Gamma|P|\;|b|^{1/2}\,(F_H\,F_{\bar H})^{1/2}}\;,\;\beta=-\tfrac{1}{2}\,\frac{F_0(1-b)}{|P|\;|b|^{1/2}(F_HF_{\bar H})^{1/2}} \tag{6}$$

φ is a real variable, $\alpha=\alpha'-i\alpha''$ and $\beta=\beta'-i\beta''$, therefore

$$\left.\begin{aligned}\eta'&=\alpha'\,\varphi+\beta'\\ \eta''&=\alpha''\varphi+\beta''\end{aligned}\right\} \tag{7}$$

By eliminating φ we get

$$\eta''=(\alpha''/\alpha')\eta'-[(\alpha''/\alpha')\beta'-\beta'']$$

or by defining the important parameters

$$K=\alpha''/\alpha'\text{ and }G=K\beta'-\beta'', \tag{8}$$

we get

$$\eta''=K\eta'-G. \tag{9}$$

If we realize that

$$|\eta|^2=\eta'^2+\eta''^2$$

from equations (5) and (9) we can write

$$L=\eta'^2+(K\eta'-G)^2+\{[\eta'^2+(K\eta'-G)^2-1]^2+4(K\eta'-G)^2\}^{1/2}. \tag{10}$$

We may choose η' for our 'normalized y'. For the case of no absorption ($\alpha''=\beta''=0$) we get the Darwin curve ($K=G=0$), and the region of total reflexion lies in the interval $-1\le\eta'\le 1$, as it should be. If absorption is present, however, one point always remains, namely $\eta'_D=G/K$, where the Darwin curve is reached, otherwise the curve is always lower. Thus the statement of BC is proved to be right even in those cases, where the condition $-'F''_H/F'_H$ is small enough compared with unity' (BC, p. 707) – is not fulfilled.*

3. Some geometrical considerations

The RC C_H^p may be constructed in three steps, namely:

(i) According to (3), C_H is a real function of the complex variable η, and thus one can construct a surface $z=C_H(\eta)$ above the (η',η'') plane. This surface is general and independent of the actual physical situation.

(ii) The physics is introduced by relation (9), with two real constants K and G. The actual C_H is a cut of this plane with the general surface, followed by a projection, paralled to the $(0\eta'')$ axis, of this cut on the (η',z) plane.

(iii) Conversion of the variable η' to the angle variable $\varphi=\theta-\theta_B$ according to (7) and change of scale of z by multiplying with $|F_H/F_{\bar H}|$ according to (2).

* To be more exact this is true for the curve C_H, which equals the RC only in the non-polar case.

An insight into the properties of the general surface may be obtained in the following way [see equations (4) and (5)]:

$$C_H \equiv z = L - (L^2-1)^{1/2} \leq 1$$
$$L = \eta'^2 + \eta''^2 + [(\eta'^2+\eta''^2-1)^2 + 4\eta''^2]^{1/2} \geq 1$$

This equation may be transformed into

$$\frac{2\eta'^2}{L+1} + \frac{2\eta''^2}{L-1} = 1$$

and from the preceding one we have $L=(z^2+1)/(2z)$; thus

$$\tfrac{1}{2}(L+1) = (z+1)^2/(4z)$$
$$\tfrac{1}{2}(L-1) = (z-1)^2/(4z)$$

If we take a fixed $z \in (0,1)$ we get an ellipse

$$\frac{\eta'^2}{a_1^2(z)} + \frac{\eta''^2}{a_2^2(z)} = 1 \qquad (11)$$

with axes

$$a_1(z) = \tfrac{1}{2}(1+z)/\sqrt{z} \text{ and } a_2(z) = \tfrac{1}{2}(1-z)/\sqrt{z}$$

and foci in

$$\eta'_F(z) = \pm(a_1^2 - a_2^2)^{1/2} = \pm 1 ,$$

common to all ellipses (11), independent of z. Thus the general part of our construction is solved.

The straight line (9) cuts these ellipses in points $(\eta', \eta''=K\eta'-G, z)$. (Because of $|G/K| \geq 1$ (see Appendix I), this straight line never passes between the two foci.) C_H on the η' scale is then the projection of these points in the (η', z) plane (see Fig. 1). One sees immediately some important properties of C_H:

(i) If $K=G=0$, the Darwin curve is obtained as a cut of the general surface with the (η', z) plane.
(ii) If K and $G \neq 0$, the curve is asymmetric and in the point $\eta'_D = G/K$ matches the Darwin curve; in all other points it is lower. For $G=K$ this point coincides with one of the foci, and thus 'total reflexion' is reached.
(iii) If $K=0$, $G \neq 0$, the curve is symmetrical but always lower than the Darwin curve.

The constants K and G are the same for both the H and $\bar{H}$ reflexion; thus the shapes of both RC's are equal. The only difference is in the ratio $|F_H/F_{\bar{H}}|$ which appears in (2). From this construction some important points are easily obtained. The sole 'Darwin' point η'_D was just mentioned. The maximum of the RC may be found by finding an ellipse to which the straight line (9) is tangent. This leads to

$$\left.\begin{aligned} &\eta'_M = (K/G)\,(G^2+1)\,(1+K^2)^{-1} \\ \text{and} \quad &z_M = (1+K^2)^{-1}[(G^2+1)^{1/2} - (G^2-K^2)^{1/2}]^2 . \end{aligned}\right\} \qquad (12)$$

$C_H^p(\max) = |F_H/F_{\bar{H}}| \,.\, z_M$; thus one must show that $C_H^p \leq 1$. The proof is given in Appendix II.

At the midpoint of the region of total reflexion $(\eta'_0=0), z_0 = [(G^2+1)^{1/2} - |G|]^2$; z_0 is independent of K and thus common to all C_H's with the same G.

4. Alternative definitions of 'y'

Any linear transformation $y = C\eta' + D$, with $C(K,G)$ and $D(K,G)$ such that for $K=G=0$ $C(0,0)=1$, $D(0,0)=0$ may be used as an alternative 'normalized y'. Let us try

$$y = (1+K^2)\eta' - KG \qquad (13)$$

which gives

$$\eta' = (1+K^2)^{-1}\,(y+KG).$$

Substituting this in (10), we get

$$L = (1+K^2)^{-1}\{y^2 + G^2 + [(y^2 - G^2 - 1 + K^2)^2 + 4(Gy-K)^2]^{1/2}\} \qquad (14)$$

which is identical with the formula from Hirsch & Ramachandran (1950) for centrosymmetrical crystals. In our derivation, however, it is valid for the general case. We therefore must discuss the definitions of G and K, which are obviously generalizations of the g and κ for the centrosymmetrical case. We note that

$$F_H \,.\, F_{\bar{H}} = (F'_H + iF''_H)\,(F'^*_H + iF''^*_H)$$
$$= |F'_H|^2\,[1 - |F''_H/F'_H|^2 + i(F''_H/F'_H + F''^*_H/F'^*_H)]$$

which in Cole & Stemple's (1962) notation reads

$$F_H \,.\, F_{\bar{H}} = |F'_H|^2\,(1 - \kappa^2 + i2p) .$$

For our purposes we set

$$1 - \kappa^2 + i2p = (a' + ia'')^2, \quad |a|^2 = a'^2 + a''^2 \qquad (15)$$

and write (6)

$$\alpha = A/a = A/|a|^2 \,.\, (a' - ia'')$$
$$\beta = B/a \,.\, (F'_0 + iF''_0) = B/|a|^2 \,.\, [(a'F'_0 + a''F''_0) - i(a''F'_0 - a'F''_0)]$$

with

$$A = \frac{-b \sin 2\theta_B}{\Gamma |P|\,|b|^{1/2}\,|F'_H|} \text{ and } B = -\tfrac{1}{2}\,\frac{1-b}{|P|\,|b|^{1/2}\,|F'_H|} . \qquad (16)$$

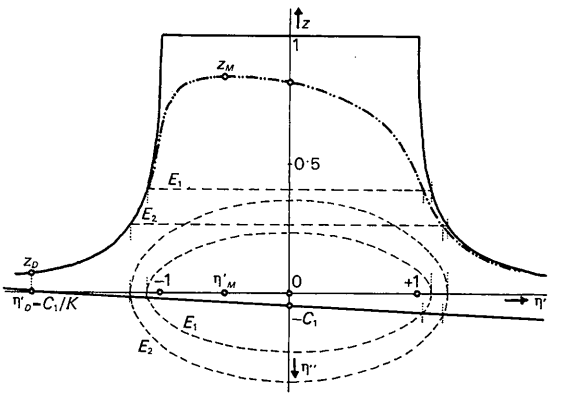

Fig. 1 The construction of C_H ($G=-0{,}1$, $K=0{,}05$). – Darwin curve, — · · — Darwin–Prins curve. The straight line [equation (9)] and the construction of two points of C_H are shown, using both projections of the ellipses E_1 and E_2. Some important points are indicated.

Then (8)

$$K=\alpha''/\alpha'=a''/a', \quad G=K\beta'-\beta''=B/a' \,.\, F_0'' . \qquad (17)$$

For the centrosymmetric case $\kappa=p=F_H''/F_H'$, which gives $a'=1$, $a''=\kappa$, thus $K=\kappa$, $G=g$, as defined by Hirsch & Ramachandran. Therefore, we may write

$$G=g/a' . \qquad (18)$$

From (15) we get

$$1-\kappa^2=a'^2(1-K^2),\ a'a''=p$$
$$a'^2(1+K^2)=|a|^2=[(1-\kappa^2)^2+4p^2]^{1/2} .$$

Substituting this result into (14)

$$L=[(1-\kappa^2)^2+4p^2]^{-1/2}$$
$$\times\{(ya')^2+g^2+[((ya')^2-g^2-1+\kappa^2)^2+4\{g(ya')-p\}^2]^{1/2}\} .$$

and defining

$$y'=ya' , \qquad (19)$$

we get the formula (13) of Cole & Stemple (1962) with all consequences stated in Bucksch, Otto & Renninger (1967). For the case of no absorption, all constants g, k, p and of course K and G are zero, and $\eta'=y=y'$ [from (13), (19)]. From the definition of y (13) and using (9) one gets

$$y=\eta'+K\eta''$$

Looking at the Fig. 2, one sees that one may obtain C_H on the y scale by projecting the cut on the (η',z) plane in the direction normal to this cut. Therefore one can conclude, that on the y scale C_H will be broadened, and one can find two points where C_H cuts the Darwin curve. For the non-polar case those points are found by comparing the expression (14) with that for the Darwin curve $L=2y^2-1$, ($|y|>1$) and one gets *e.g.* for $K=-G$ $y_1=-1$, $y_2=1+(1+K^2)^{-1/2}$.

Conclusions

We have shown that by the suitable definition of the 'normalized variable y' (and the constants G and K) one may obtain a general formula for the RC for the Bragg case. This means that one needs only one computing program. All the physics is involved in the constants K and G, and in the final transform of the RC to the desired angle variable. By dividing the problem in the 'general' and 'physical' part, one could, at least theoretically, draw a map of the general surface once and for all, and from the computed K and G draw the straight line (9) and read directly from the graph the coordinates for some points of the RC.

Further we have shown that different formulae, introduced by several authors are consequence of different definitions of the 'normalized' variable and we have found the transformations between these variables.

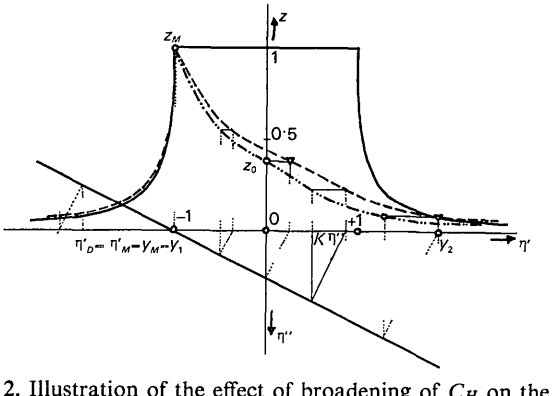

Fig. 2. Illustration of the effect of broadening of C_H on the y scale ($-G=K=0{,}5$). C_H on the η' scale is always lower except in the point η_D'. The points of intersection of the RC on the y scale with the Darwin curve are shown (y_1, y_2).

APPENDIX I

According to equation (17)

$$G/K=BF_0''/a'' .$$

From (15) we get

$$a''^2=\tfrac{1}{2}\{[(1-\kappa^2)^2+4p^2]^{1/2}-(1-\kappa^2)\} .$$

As $p=\mathrm{Re}(F_H''/F_H')$, $\kappa=|F_H''/F_H'|$ and $s=\mathrm{Im}\,(F_H''/F_H')$ we may put κ^2-s^2 instead of p^2 and get

$$a''^2=\tfrac{1}{2}\{[(1+\kappa^2)^2-4s^2]^{1/2}-(1-\kappa^2)\}\leq\kappa^2$$

the equality being reached only for $s=0$.
Thus using (16)

$$|G/K|\geq\frac{|B|\,|F_0''|}{\kappa}=\tfrac{1}{2}\frac{|1-b|}{|P|\,|b|^{1/2}}\left|\frac{F_0''}{F_H''}\right|\geq 1 \; q.e.d.$$

APPENDIX II

$C_H^p(\max)=|F_H/F_{\bar{H}}|z_M$. Now (see *e.g.* Cole & Stemple, 1962)

$$|F_H/F_{\bar{H}}|=\frac{1+\kappa^2+2s}{[(1-\kappa^2)^2+4p^2]^{1/2}}$$

Equation (12) gives

$$z_M=(1+K^2)^{-1}\,[(G^2+1)^{1/2}-(G^2-K^2)^{1/2}]^2$$
$$=(1+K^2)\,[(G^2+1)^{1/2}+(G^2-K^2)^{1/2}]^{-2} .$$

From (15)

$$1+K^2=[(1-\kappa^2)^2+4p^2]^{1/2}/a'^2$$

and using (18) we get

$$C_H^p(\max)=\frac{1+\kappa^2+2s}{[(g^2+a'^2)^{1/2}+(g^2-a''^2)^{1/2}]^2}=$$
$$\frac{1+\kappa^2+2s}{2g^2+(a'^2-a''^2)+2[g^4+g^2(a'^2-a''^2)-a'^2a''^2]^{1/2}} .$$

Now $a'^2-a''^2=1-\kappa^2$, $a'a''=p$

But $g^2\geq|F_0''/F_H'|\geq|F_H''/F_H'|=\kappa^2$.

Thus

$$C_H^p(\max)\leq\frac{1+\kappa^2+2s}{1+\kappa^2+2(\kappa^2-p^2)^{1/2}}=\frac{1+\kappa^2+2s}{1+\kappa^2+2|s|}\leq 1 \; q.e.d.$$

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A Neutron Fourier Chopper for Single Crystal Reflectivity Measurements: Some General Design Considerations*

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Neutron single-crystal analysis of complex protein structures requires the collection of intensity data on a large number of reflections. The use of time-of-flight analysis (in particular Fourier time-of-flight analysis employing phase detection) of diffraction data, offers a significant increase in the efficiency of the use of reactor neutrons. Such a method would facilitate the use of neutron diffraction in protein structure work. In this paper, we investigate the use of the Fourier chopper in single-crystal work. We examine the effect of various system parameters (frequency stability, flight path fluctuations, collimation, sample mosaic spread, inaccuracy in phase detection, and statistical counting errors) and determine general specifications for such an apparatus. None of these specifications appears to be beyond present day technology.

1. Introduction

The use of neutron diffraction in structure studies of complex organic molecules is attracting the interest of crystallographers (Moore, Willis & Hodgkin, 1967; Schoenborn, 1969). Neutrons offer the following advantages: (1) hydrogen atoms become visible, (2) nitrogen atoms may be distinguished from carbon or oxygen atoms, (3) damage to specimens during irradiation is absent and (4) anomalous dispersion offers the possibility of easily phasing diffraction data. The chief disadvantage of neutrons is that compared with X-ray sources, the neutron flux (monochromated and collimated) available from even the highest flux reactors is down by a factor of 10^5 [as compared with a copper target rotating anode X-ray source (Arndt, 1969)].

Neutrons and neutron sources differ from X-rays and their sources in a number of additional ways. Neutron velocity is low (thermal neutron velocity is of the order of 2200 meter/second). The neutron source emits a Maxwellian distribution of velocities unlike the characteristic radiation of X-ray sources. In spite of these fundamental differences, present day neutron diffraction apparatus is basically identical to its X-ray counterpart. Typically, a narrow 'slice' of the reactor spectrum is selected by a crystal, providing a beam of monochromatic radiation which is then fed into a spectrometer equipped with a computer-controlled four-circle goniometer (Hamilton, 1968). Because this scheme of collecting data requires an essentially monochromatic beam, most of the source neutrons are wasted.

Other inefficiencies are built into this scheme. Data is taken sequentially, rocking the crystal through one reflection, then another, and so on. At any given time, typically less than one part in 10^6 of the neutrons striking the monochromator are detected at the counter.

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