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Some Theoretical Properties of the Double-Crystal Spectrometer Used in Neutron Diffraction

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A treatment is given of the angular dependence of certain intensity functions which are important in spectrometer design. It provides a basis for the choice of various parameters, including, in particular, the Bragg angle of the crystal monochromator.

1. Introduction

The structures of many single crystals have been examined by neutron diffraction: the instrument normally used is the double-crystal spectrometer, whose main features are sketched in Fig. 1. Slits S_1 and S_2, representing the collimator, transmit a neutron beam with a divergence in the horizontal plane of about $\pm\frac{1}{2}°$. The beam is reflected by the crystal monochromator A to the crystal B under investigation, and thence is reflected again into the counter C. The 'parallel' arrangement, with the incident and twice-reflected beams on opposite sides of the once-reflected beam, is used because of its well-known focusing property (Compton & Allison, 1935).

In §§ 2–4 the following features of the spectrometer are examined theoretically:

(*a*) The intensity of the beam reflected in a given direction by the monochromator A as a function of the angular setting of A. ('Rocking curve' of monochromator).

(*b*) The intensity of the beam reflected by the crystal B as a function of the angular setting of B. ('Double-reflexion curve'.)

(*c*) The divergence of the beam reflected by B.

The solution of (*b*) leads to the best choice of the parameters governing the properties of the collimator and monochromator. The treatment of (*a*) suggests a convenient method of measuring the mosaic spread of the monochromator, and (*c*) determines the minimum aperture which the counter must have to receive the whole of the beam reflected by the crystal.

Related problems have been examined theoretically by Sailor *et al.* (1956) and by Caglioti *et al.* (1958).

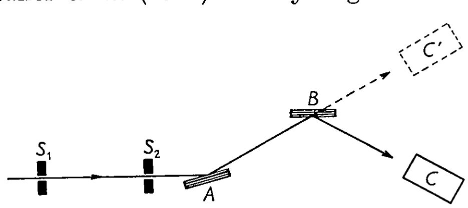

Fig. 1. Sketch of projection of spectrometer in horizontal plane.

Sailor studied the angular dependence of the intensity of the neutron beam reflected by the monochromator, with Soller slits restricting the horizontal divergence of incident and reflected beams to a few minutes of arc. Caglioti calculated the intensity of the beam, reflected by the monochromator and then by a powder sample, as a function of the angular position of the counter; again the incident, once- and twice-reflected beams were limited by Soller slits.

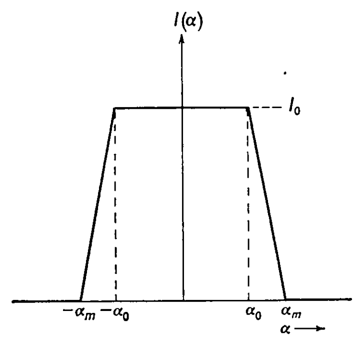

Fig. 2. Angular dependence of intensity of neutron beam striking monochromator.

In the absence of Soller slits the angular distribution of intensity of the incident neutron beam is approximately of the form shown in Fig. 2. The number of neutrons, $I(\alpha)d\alpha$, with a horizontal divergence between α and $\alpha+d\alpha$ is constant for $|\alpha|$ less than α_0, and decreases uniformly to zero as $|\alpha|$ increases from α_0 to α_m. The formulae derived below refer to the special case $\alpha_0=\alpha_m$, corresponding to a rectangular angular distribution of intensity. The general formulae for $\alpha_0 \neq \alpha_m$ are quoted elsewhere (Willis, 1959): they are cumbersome and introduce no modifications to the general conclusions given in § 5.

If crystal B is set at the Bragg angle θ_{hkl}, the change in glancing angle arising from a vertical divergence φ is $\frac{1}{2}\varphi^2 \tan \theta_{hkl}$. Observations in neutron diffraction rarely extend beyond $\theta_{hkl}=60°$, and so this change can be neglected compared with that arising from the horizontal divergence. For this reason it is only necessary to consider the projection of the neutron beam in the horizontal plane.

It is assumed that each crystal consists of mosaic blocks with a Gaussian distribution of orientation. Thus the fraction of mosaic blocks in A having their normals in the angular range Δ, $\Delta+d\Delta$ (as measured from the mean orientation) is $W(\Delta)d\Delta$, where

$$W(\Delta)=\frac{1}{\eta\sqrt{\pi}}\exp\left(-\Delta^2/\eta^2\right). \qquad (1)$$

$\eta/\sqrt{2}$ is the standard deviation of the distribution and is a measure of the mosaic spread of A. A similar equation can be written for crystal B:

$$W'(\Delta')=\frac{1}{\eta'\sqrt{\pi}}\exp\left(-\Delta'^2/\eta'^2\right), \qquad (1a)$$

where the primed quantities have the same meanings as the unprimed quantities but now refer to B. (Corresponding quantities for A and B will always be denoted by the same symbol with a prime to distinguish the symbol for B.) As η, $\eta' \ll 1$, (1) and (1a) also give the angular distributions of the projections of the normals in the horizontal plane.

2. Rocking curve of monochromator

The rocking curve of the monochromator is defined as the curve giving the intensity, $I(\beta)$, reflected by A in a fixed direction as a function of the angular setting, β, of A. A fixed direction is stipulated, as the incident beam has a white spectrum and the monochromator can reflect any wavelength in this spectrum, depending on the glancing angle of the neutrons incident on it. To measure the rocking curve experimentally crystal B must be removed and the counter moved round to the 'straight-through' position C' (Fig. 1); it is also necessary to define the fixed direction for reflexion by slits, restricting the horizontal divergence of the reflected beam to a value which is small compared with the mosaic spread, η.

Let $2\theta_B$ be the angle between the axis of the collimator and the fixed direction for reflexion. Neutrons with an initial divergence of α are reflected in this direction by the mosaic blocks whose orientation is Δ, where

$$\Delta=-(\beta+\tfrac{1}{2}\alpha). \qquad (2)$$

The signs of α, β, Δ are such that the glancing angle on A increases with these angles. $\beta=0$ is defined as the angular position of the monochromator, when neutrons passing along the axis of the collimator are reflected at the angle θ_B by the mosaic blocks of mean orientation.

Ignoring the crystal reflectivity, which varies relatively slowly with angle, the number of neutrons, of initial divergence α, reflected in the fixed direction is $I(\alpha)d\alpha W(\Delta)$, where

$$\left.\begin{aligned} I(\alpha) &= I_0, \quad |\alpha|<\alpha_0 \\ &= 0, \quad |\alpha|>\alpha_0 . \end{aligned}\right\} \qquad (3)$$

The total number reflected for all values of α is

$$I(\beta)=\int_{-\alpha_m}^{\alpha_m} I(\alpha)W(\Delta)d\alpha ,$$

which from (1), (2) and (3) becomes

$$I(\beta)=\frac{I_0}{\eta\sqrt{\pi}}\int_{-\alpha_m}^{\alpha_m}\exp\left[-(\beta+\tfrac{1}{2}\alpha)^2/\eta^2\right]d\alpha$$

$$=I_0\left[\operatorname{erf}\left(\frac{\beta+\frac{1}{2}\alpha_m}{\eta}\right)-\operatorname{erf}\left(\frac{\beta-\frac{1}{2}\alpha_m}{\eta}\right)\right]. \qquad (4)$$

Here erf x is the tabulated error function, defined by

$$\operatorname{erf} x=\frac{2}{\sqrt{\pi}}\int_0^x \exp\left(-\xi^2\right)d\xi .$$

$I(\beta)$ has been calculated as a function of β from (4), and the results for $\alpha_m=0$, η, 2η are presented in Fig. 3. Provided $\alpha_m \not> 2\eta$, the half-width at half-height, H_1, of the rocking curve is within 10% of $0{\cdot}95\eta$. Extinction will tend to make H_1/η larger, but not unduly so. Thus an approximate value of η can be found by measuring H_1 and using $H_1/\eta \approx 1$, unless the value obtained is less than $\frac{1}{2}\alpha_m$; in the latter case η is found by fitting the rocking curve to the equation (4).

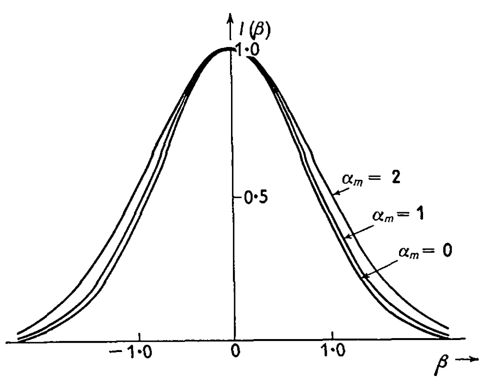

Fig. 3. Rocking curves of monochromator for different values of collimation angle, α_m, β and α_m are expressed in units of η, the mosaic spread of the monochromator.

3. Double-reflexion curve

This curve gives the intensity, $I(\beta')$, of the hkl Bragg reflexion of crystal B as a function of the angular position, β', of B. The area under this curve is proportional to F^2_{hkl} cosec $2\theta_{hkl}$. The dependence of the shape of this curve on the mosaic spreads and Bragg angles of the two crystals and on the collimation angle is investigated below.

A beam of neutrons of initial divergence α is reflected at a fixed wavelength by the mosaic blocks in A with a given orientation Δ; this wavelength can then be reflected again only by the mosaic blocks in B, whose orientation, Δ', satisfies Bragg's law. This value of Δ' is readily shown to be

$$\Delta'=(k-1)\alpha+(k-2)\Delta-\beta', \qquad (5)$$

where $k = \tan\theta_{hkl}/\tan\theta_B$. The sign of β' is chosen so that the glancing angle on B increases with β', and the angular position $\beta'=0$ is defined as that for which a neutron of no initial divergence is reflected by the mosaic blocks in A and B of mean orientation.

The number of neutrons reflected into the counter for fixed α, Δ is proportional to $I(\alpha)W(\Delta)W'(\Delta')$. $I(\beta')$ is obtained by integrating over α and Δ:

$$I(\beta') = \int_{-\alpha_m}^{\alpha_m}\int_{-\infty}^{\infty} I(\alpha)W(\Delta)W'(\Delta')d\alpha d\Delta\ ,$$

which from (1), (1a) and (3) becomes

$$I(\beta') = \frac{I_0}{\pi\eta\eta'}\int_{-\alpha_m}^{\alpha_m}\int_{-\infty}^{\infty} \exp(-\Delta^2/\eta^2) \times \exp(-\Delta'^2/\eta'^2)d\alpha d\Delta\ . \quad (6)$$

Substituting for Δ' from (5) and using the identity

$$\int_{-\infty}^{\infty}\exp(-A\Delta^2-2B\Delta-C)d\Delta = \left(\frac{\pi}{A}\right)^{\frac{1}{2}}\exp\left(\frac{B^2}{A}-C\right)\ ,$$

the second integral in (6) can be written

$$\frac{\eta\eta'}{\varepsilon}\sqrt{\pi}\exp\left[-(k\alpha-\alpha-\beta')^2/\varepsilon^2\right]\ ,$$

where

$$\varepsilon^2 = \eta'^2 + (k-2)\eta^2.$$

Thus

$$I(\beta') = \frac{I_0}{\sqrt{(\pi)}\,\varepsilon}\int_{-\alpha_m}^{\alpha_m}\exp\left[-(k\alpha-\alpha-\beta')^2/\varepsilon^2\right]d\alpha = \frac{I_0}{k-1}\left[\operatorname{erf}\left(\frac{\beta'+k\alpha_m-\alpha_m}{\varepsilon}\right)-\operatorname{erf}\left(\frac{\beta'-k\alpha_m+\alpha_m}{\varepsilon}\right)\right]. \quad (7)$$

For the special case $k=1$, when the incident and twice-reflected beams are parallel, (7) simplifies to

$$I(\beta') = \frac{2}{\sqrt{\pi}}\frac{I_0\alpha_m}{\sqrt{(\eta^2+\eta'^2)}}\exp\left(-\frac{\beta'^2}{\eta^2+\eta'^2}\right).$$

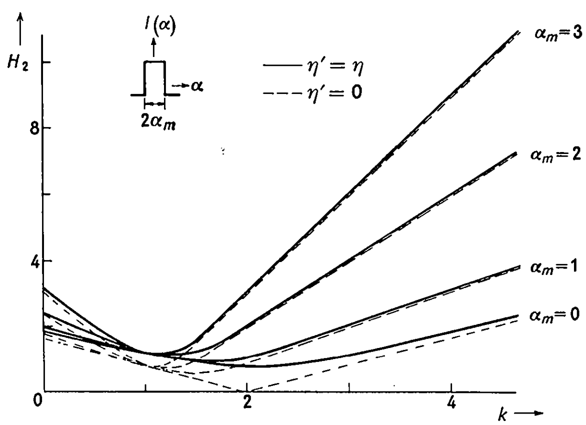

Fig. 4. Half-width of double-reflexion curve as a function of k ($=\tan\theta_{hkl}/\tan\theta_B$). H_2 and α_m are expressed in units of η, the mosaic spread of the monochromator.

The half-width at half-height, H_2, of the double-reflexion curve is then

$$H_2 = \sqrt{(\ln 2(\eta^2+\eta'^2))}\ ,$$

and is independent of the collimation angle, α_m.

For all other values of k, H_2 depends on α_m and on η and η'. The nature of this dependence is illustrated by Fig. 4, where H_2, calculated from equation (7), is plotted as a function of k. The different pairs of curves refer to $\alpha_m = 0, \eta, 2\eta, 3\eta$, the unbroken curve in each pair corresponding to $\eta'=\eta$ and the broken curve to $\eta'=0$. H_2 passes through a minimum between $k=1$ and $k=2$ and is approximately proportional to α_m for $k>3$.

It is clearly desirable that the Bragg angle, θ_B, of the monochromator should be chosen to make H_2 approximately the same at either end of the range of θ_{hkl} under investigation. This implies that, for a range of 0–60° in θ_{hkl}, θ_B will be about 40°.

4. Angular divergence of twice-reflected beam

We shall consider only the case when crystal B is set exactly at the Bragg angle θ_{hkl}. The condition that neutrons are reflected by the mosaic blocks in A of orientation Δ and then by the blocks in B of orientation Δ' is given by equation (5) with $\beta'=0$:

$$\Delta' = (k-1)\alpha + (k-2)\Delta\ . \quad (8)$$

Let φ' denote the angular position of these twice-reflected neutrons, where the sign of φ' is chosen to make the glancing angle on B increase with φ', and the position $\varphi'=0$ is defined as that corresponding to neutrons of no initial divergence reflected by the mosaic blocks in A and B of mean orientation. Then φ' is given by

$$\varphi' = \alpha + 2\Delta + 2\Delta', \quad (9)$$

and (8) and (9) together define the orientations Δ, Δ' of the mosaic blocks in A, B, which change the angular position of the neutrons from the incident value of α to the doubly-reflected value of φ'. Solving (8) and (9), these orientations are

$$\left.\begin{aligned}\Delta &= \frac{1}{2(k-1)}(\varphi'-2k\alpha+\alpha)\\ \Delta' &= \frac{1}{2(k-1)}(k\varphi'-2\varphi'+k\alpha)\ .\end{aligned}\right\} \quad (10)$$

The total intensity of neutrons reflected for all values of α into the direction φ' is $I(\varphi')$, where

$$I(\varphi') = \int_{-\alpha_m}^{\alpha_m} I(\alpha)W(\Delta)W'(\Delta')d\alpha$$

and Δ, Δ' are given by (10). Using equations (1), (1a) and (3), $I(\varphi')$ can be finally manipulated into the form

$$I(\varphi') = c \exp\left[-\frac{(k-1)^2\varphi'^2}{k^2\eta^2+(2k-1)^2\eta'^2}\right] \times \left(\operatorname{erf}\left[\frac{\sqrt{(a)}(\alpha_m+b/a)}{2(k-1)}\right]+\operatorname{erf}\left[\frac{\sqrt{(a)}(\alpha_m-b/a)}{2(k-1)}\right]\right),$$

where

$$a = \frac{(2k-1)^2}{\eta^2}+\frac{k^2}{\eta'^2}$$

$$b = \varphi'\left(\frac{1-2k}{\eta^2}+\frac{k^2-2k}{\eta'^2}\right)$$

and c is a constant.

The curves in Fig. 5 have been plotted from this expression for the special case $\eta'=\eta$: they give the variation with k of H_3, the half-width at half-height of the $I(\varphi')$ versus φ' distribution. H_3 is a measure of the divergence of the twice-reflected beam; it varies appreciably with k and attains a maximum value between $k=1$ and $k=3$, according to the value of α_m. For $\eta'<\eta$, H_3 is reduced approximately in the ratio $\eta':\eta$.

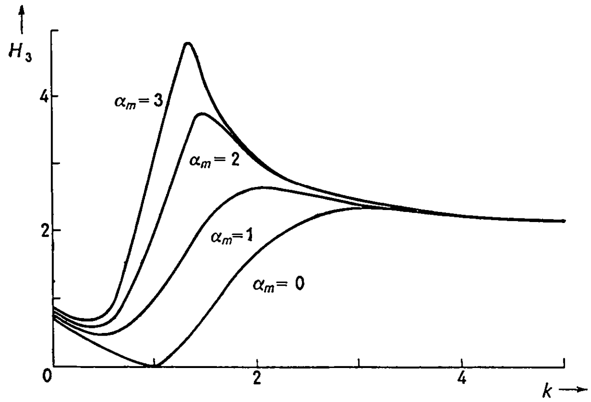

Fig. 5. Curves showing the dependence of the divergence of the twice-reflected beam on k ($=\tan\theta_{hkl}/\tan\theta_B$). H_3 and α_m are expressed in units of η, the mosaic spread of the monochromator.

As a typical case, $\alpha_m=30'$, $\eta=20'$ and $\eta'=10'$, giving a maximum value of H_3 of 30'. More than 99% of the beam lies within the angular range $|\varphi'|<2{\cdot}5H_3$, and so the aperture of the counter must be large enough to receive a beam diverging from the crystal at an angle of $\pm 1{\cdot}25°$.

5. Conclusions

The analysis above provides a basis for the choice of various parameters appearing in spectrometer design. Using § 2 the mosaic spread of the monochromator can be determined from the experimental rocking curve; the best choice of collimation angle and Bragg angle, θ_B, of the monochromator then follows from § 3. Finally, the minimum counter aperture can be determined from § 4.

The choice of θ_B is particularly important, because of its influence on the widths of the Bragg reflexions of the second crystal. The curves in Fig. 4 show that, as θ_B increases, the reflexions become narrower over the whole range of θ_{hkl} of the crystal. This effect is to be ascribed primarily to the reduction in width of the wavelength band reflected by the monochromator. At the focusing position the whole of this band is reflected simultaneously by the mosaic blocks set at the Bragg angle θ_{hkl}, and the width of the reflexion is determined only by the mosaic spread. However, away from the focusing position simultaneous reflexion does not occur. As the crystal turns, each mosaic block reflects a sharp wavelength, which gradually sweeps across the wavelength band, and under these conditions the width of the reflexion is determined mainly by the width of the incident wavelength band.

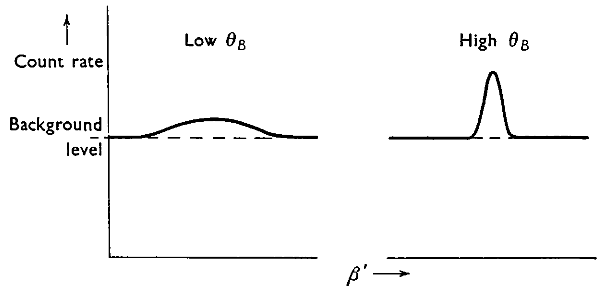

Fig. 6. Diagram illustrating advantage of using a relatively high value of θ_B to observe weak high-order reflexions.

By using relatively high values of θ_B(~45°) the hkl reflexions are made particularly narrow at high angles, where the integrated reflexions are reduced by the Lorentz (cosec 2θ) and Debye–Waller

$$(\exp[-2B\sin^2\theta/\lambda^2])$$

factors. Frequently the peak intensities of the high-angle reflexions are only fractionally higher than the background intensity (produced, for example, by incoherent scattering from hydrogen atoms in the sample), and there is clearly much advantage in increasing θ_B (see Fig. 6). The fall-off with θ_B of the reflectivity of the monochromator can be partly off-set by relaxing the degree of collimation, but, in any case, measurements with several copper crystal monochromators have indicated that the reflectivity falls by a factor of only 2 or 3, as θ_B is increased from 10° to 45°.

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