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The expressions derived for the calculation of multiple diffraction patterns for a perfect crystal in the framework of the kinematical theory are compared with the relevant result of the kinematical power-transfer equation deduced for a mosaic crystal. It is shown that the results of the two concepts differ appreciably.

1. Introduction

As pointed out in Ro-00*c*,[1] multiple diffraction in the framework of the kinematical theory was first considered by Renninger (1937). In §7 of his famous paper, two kinematical approaches are discussed, the 'simplest approach' (*einfachster Ansatz*), which is used by the author, and a 'more sophisticated approach' (*verfeinerter Ansatz*), which is similar to the power-transfer equation presented by Moon & Shull (1964) for neutron diffraction. Whereas chapters 5.2 and 5.4 of Chang's book (Chang, 2004) are mainly based on the paper by Moon & Shull (1964), chapter 5.5 refers to Ro-00*c* and Ro-03.

Some of the statements as well as the conclusions drawn by Chang (2004) at the end of his chapter 5, namely '*This* (the result of the power-transfer equation, expression 5.36 in his book) *is the same as that derived from Renninger's simplest approach* (the author's result)' and '*the appreciable interference, especially the profile asymmetry, can be observed when a perfect crystal or strong reflection are involved. This effect is therefore dynamical rather than kinematical. The addition of the interference term to the expression for integrated intensity is reasonable from the physics point of view. However, it is not mathematically rigorous, because such a term cannot be obtained directly from the power-transfer equation governing the multiple-wave diffraction process. In fact, to reveal the phase effect, the EM wavefields rather than the reflection powers need to be considered in describing the multiple-wave diffraction behaviour*' need some rectification, comments and completion.

To be able to do this, in the first step the relevant expressions derived for perfect crystals in the framework of the kinematical theory by the author, as well as the relevant result of the power-transfer equation introduced by Moon & Shull (1964) and used in chapter 5 of Chang's book, are collated in the next two sections.

[1] Most of the expressions and figures discussed in this paper were derived or presented in previous papers of the author. These expressions, figures, Appendices *etc.* will be referenced in the following by the abbreviation Ro-*xxy*-(*z*), where *xx* represents the two last digits of the year of publication, *y* stands for *a*, *b*, *c etc.* if more than one paper in the respective year is referenced and *z* represents the number of the expression, figure, Appendix *etc.* under consideration.

2. Summary of expressions derived in the framework of the kinematical approach for an ideally perfect spherical crystal bathed in the incident X-ray beam

In Ro-04-(Fig. 1), the conditions of a three-beam case are shown. $\mathbf{s}_0$ is a unit vector parallel to the beam incident on the crystal and $\mathbf{s}_1$ and $\mathbf{s}_2$ are unit vectors parallel to the simultaneously diffracted primary and operative reflection, respectively. According to Zachariasen (1945), the electric vectors $\mathbf{E}_1$ and $\mathbf{E}_2$ corresponding to the diffracted beams depend on the direction of diffraction and the electric vector of the incident beam $\hat{\mathbf{E}}_0$ according to

$$\mathbf{E}_1 = -\mathbf{s}_1 \times (\mathbf{s}_1 \times \hat{\mathbf{E}}_0) \tag{1}$$

$$\mathbf{E}_2 = -\mathbf{s}_2 \times (\mathbf{s}_2 \times \hat{\mathbf{E}}_0). \tag{2}$$

In the case of multiple diffraction [see Ro-04-(§2)], the beam diffracted in the $\mathbf{s}_2$ direction with the electric unit vector $\hat{\mathbf{E}}_2$ is re-diffracted in the $\mathbf{s}_1$ direction (Renninger, 1937) generating the *Umweg* wave with the electric unit vector $\hat{\mathbf{E}}_{21} = \mathbf{E}_{21}/|\mathbf{E}_{21}|$. Assuming that the two beams in the $\mathbf{s}_1$ direction are excited independently of each other, $\mathbf{E}_{21}$ is given by

$$\mathbf{E}_{21} = -\mathbf{s}_1 \times (\mathbf{s}_1 \times \hat{\mathbf{E}}_2) = \mathbf{s}_1 \times (\mathbf{s}_1 \times (\mathbf{s}_2 \times (\mathbf{s}_2 \times \hat{\mathbf{E}}_0)))/|\mathbf{E}_2|. \tag{3}$$

This *Umweg* wave interferes with the wave represented by $\hat{\mathbf{E}}_1$. In general, the two electric unit vectors $\hat{\mathbf{E}}_1$ and $\hat{\mathbf{E}}_{21}$ will not be parallel. Therefore, according to Hecht (1989, §9.1), the intensity of the superimposition of the two waves is given by

$$I = I_1 + I_{21} + 2(I_1 I_{21})^{1/2} \cos(\angle \hat{\mathbf{E}}_1, \hat{\mathbf{E}}_{21}) \cos\varphi, \tag{4}$$

where I_1 and I_{21} are the intensities of the two waves, respectively, $\cos(\angle \hat{\mathbf{E}}_1, \hat{\mathbf{E}}_{21})$ is the cosine of the angle between the two unit vectors $\hat{\mathbf{E}}_1$ and $\hat{\mathbf{E}}_{21}$ and φ is the phase difference between the two waves. The evaluation of the angles $\angle \hat{\mathbf{E}}_1, \hat{\mathbf{E}}_{21}$ and φ is fully discussed in Ro-04. The derivation of the intensities I_1 and I_{21} is based on Renninger's 'simplest approach'.

Starting with Ro-00*c*-(1) given by Renninger (1937), it was shown in Ro-00*c*-(§2) and Ro-00*c*-(§3.1) that the power received in the counter during the rotation about the ψ axis is given by [Ro-00*c*-(20)]

$$I_{s_1}^{(\omega)}(\psi) = \{I_{\rm prim}^{(\omega)} - I_{\rm Aufh}^{(\omega,\psi)} f(\psi)\} + I_{\rm Umweg}^{(\omega,\psi)} f(\psi) + I_{\rm interfer}^{(\omega)}(\psi) \quad (5)$$

with

$$\begin{aligned} I_{\rm prim}^{(\omega)} &= (I_0 q_{s_0})(\lambda L_\omega)\frac{\bar{t}_{s_0}}{\Lambda_{\rm prim}^2} \\ I_{\rm Aufh}^{(\omega,\psi)} &= (I_0 q_{s_0})(\lambda L_\omega)(\lambda L_\psi)\frac{\bar{t}_{s_0}}{\Lambda_{\rm prim}^2}\left(\frac{\bar{t}_{s_0}}{\Lambda_{\rm op}^2} + \frac{\bar{t}_{s_1}}{\Lambda_{\rm coop}^2}\right) \\ I_{\rm Umweg}^{(\omega,\psi)} &= (I_0 q_{s_0})(\lambda L_\omega)(\lambda L_\psi)\left(\frac{\bar{t}_{s_0}}{\Lambda_{\rm op}^2}\frac{\bar{t}_{s_2}}{\Lambda_{\rm coop}^2}\right). \end{aligned} \quad (6)$$

The approximations necessary to obtain the expression (5) are fully discussed in Ro-00*c*. The term in the curly brackets in (5) corresponds to I_1, the second term to I_{21}. For simplicity, in a first step in Ro-00*c*, the polarization effects were not considered. The corresponding interference term (Klein & Furtak, 1988, chapter 5) results in Ro-00*c*-(21). Taking into account the polarization of the beams I_1 and I_{21}, the derivation of the interference term given by Hecht (1989), *i.e.* the third term of expression (4) has to be used:

$$\begin{aligned} I_{\rm interfer}^{(\omega)}(\psi) = 2[\{I_{\rm prim}^{(\omega)} - I_{\rm Aufh}^{(\omega,\psi)} f(\psi)\} I_{\rm Umweg}^{(\omega,\psi)} f(\psi)]^{1/2} \\ \times \cos\varphi(\psi)\cos(\angle\hat{\mathbf{E}}_1, \hat{\mathbf{E}}_{21}). \end{aligned} \quad (7)$$

I_0 in (6) is the incident intensity (energy s^{-1} cm^{-2}) and q_{s_0} is the cross section of the crystal volume normal to the incident beam, *i.e.* $(I_0 q_{s_0}) = J_0$, the incident power, and Λ_h is defined by

$$1/\Lambda_h = r_0\lambda|F_h|TK/V_{\rm cell}, \quad (8)$$

where r_0 is the classical electron radius, λ is the wavelength of the radiation used for diffraction, $|F_h|$ is the modulus of the complex structure factor, T is the temperature factor [Ro-03-(5)] and $V_{\rm cell}$ is the volume of the unit cell. K is the polarization coefficient, defined by Ro-04-(16). The Lorentz factors L_ω and L_ψ, corresponding to the rotation about the ω and ψ axis, respectively, were developed in Ro-92. L_ω is defined by [Ro-92-(9*h*), -(10*c*), -(16), Ro-00*a*-(15)]

$$L_\omega = \Delta\omega/(\ell_{{\rm Umweg},\omega}\lambda) \approx 1/\sin 2\theta_{\rm prim} \quad (9a)$$

and L_ψ [Ro-92-(19*a*),-(19*c*)]

$$L_\psi = \Delta\psi/(\ell_{{\rm op},\psi}\lambda) \approx 1/[\lambda h_{\rm op}^n \cos\theta_{\rm prim}\sin\beta], \quad (9b)$$

where ℓ_i is the 'effective thickness' of the Ewald sphere in the direction of the reflected beam i, $\theta_{\rm prim}$ is the Bragg angle of the primary reflection and $h_{\rm op}^n$ and β are defined in Ro-03-(Fig. 1). The validity of the new generalized Lorentz factor L_ψ in cases where the approximation given in (9*b*) results in an infinite value is demonstrated in Ro-00*d* and Ro-01. L_ω is essential for comparing the intensities of multiple diffraction patterns of different primary reflections (Ro-95) or for the estimation of the intensity of multiple diffraction events on an absolute scale.

The extinction-corrected mean thickness of the ideally perfect crystal sample in the direction of the incident beam, $\bar{t}_{s_i}$, is defined by [Ro-00*c*-(6)]

$$\bar{t}_{s_i} = (V_{\rm cry}/q_{s_i})y_p, \quad (10)$$

where $V_{\rm cry}$ is the crystal volume and q_{s_i} is the cross section of the crystal volume normal to the beam incident in the $\mathbf{s}_i$ direction and y_p is the primary extinction correction discussed in Ro-00*b*.

It was found by comparisons of theoretical and experimental multiple diffraction patterns that $f(\psi)$ can be well approximated by an asymmetric normalized split-pseudo-Voigt function consisting of two halves, $\mathrm{PV}(\psi)_{\rm left}$ and $\mathrm{PV}(\psi)_{\rm right}$, with different widths and different mixing parameters for the left and right sides, respectively, but with common maximum value, where $\mathrm{PV}(\psi)$ is the normalized pseudo-Voigt function [Ro-02*b*-(Appendix *A*) and Ro-02*b*-(Fig. 14)]

$$\begin{aligned} \mathrm{PV}(\psi) = \frac{1}{\Delta\psi_{\rm integral}^{\rm total}}\left\{(1-\eta)\exp\left[-\pi\left(\frac{\psi-\psi_0}{\Delta\psi_{\rm integral}^{\rm total}}\right)^2\right]\right. \\ \left. + \eta\frac{1}{1+\left(\frac{\pi(\psi-\psi_0)}{\Delta\psi_{\rm integral}^{\rm total}}\right)^2}\right\}. \end{aligned} \quad (11)$$

ψ_0 corresponds to the value of the variable in the peak maximum. The mixing parameter η [see Ro-03-(8) for an approximate estimation of this parameter] is a measure of the Lorentzian contribution to the pseudo-Voigt distribution. $\Delta\psi_{\rm integral}^{\rm total}$, the total integral width of the reflection with respect to the rotation about the ψ axis, mainly depends on the size of the perfect crystallites and the divergence and wavelength spread of the incident beam. In the case of a mosaic crystal, additionally the mosaic spread will broaden the distribution.

$$\begin{aligned} \Delta\psi_{\rm integral}^{\rm total} = (\Delta\psi_{\rm crystal\ size} + \Delta\psi_{\rm divergence} + \Delta\psi_{\rm wavelength\ spread} \\ + \Delta\psi_{\rm mosaic\ spread}). \end{aligned} \quad (12)$$

The concept of the evaluation of $\Delta\psi_{\rm integral}^{\rm total}$ by means of purely geometrical considerations is discussed in detail in Ro-92. The validity of this approach, questioned by Mathieson (1994), is proved in Ro-02*a* and Ro-02*b*. The presentation and discussion of the extensive expressions developed for use in the program *UMWEG* (Ro-03) for the calculation of the peak widths for all possible experimental conditions, although not published in detail until now, is beyond the scope of this paper.

The phase difference $\varphi(\psi)$ between the primary and the *Umweg* wave [see Ro-03-(9)] is discussed in detail in Ro-04-(§2) and Ro-04-(Appendix *A*).

According to Ro-00*c*, Ro-03 and Ro-04, the total power $I_{s_1}(\psi)_{\rm total}$, received in the counter during the rotation about the ψ axis, is approximated by [Ro-03-(1)]

$$I_{s_1}(\psi)_{\rm total} = g(\alpha_{1,2})\left\{I_{\rm prim}^{(\omega)} + \sum_{\rm event}[I_{s_1}^{(\omega)}(\psi)_{\rm event}]\right\}, \quad (13)$$

where, in the case of X-ray tubes, $g(\alpha_{1,2})$ depends on the intensity ratio of the $K\alpha_1$ and $K\alpha_2$ radiations, and in other cases it is equal to 1 and the subscript 'event' refers to a particular three-beam case.

3. Power-transfer equations. Ideally mosaic crystal plate

For a plane parallel plate with thickness T, consisting of many plate-like ideal crystal blocks, *i.e.* for an ideal mosaic crystal, the power-transfer equation can be expressed as (Moon & Shull, 1964; Chang, 2004, expression 5.4)

$$\pm\frac{\mathrm{d}P_i}{\mathrm{d}x}=\frac{\mu_0 P_i}{\gamma_i}+\sum_j\left(\frac{Q_{ji}P_j}{\gamma_j}-\frac{Q_{ij}P_i}{\gamma_i}\right), \tag{14}$$

where P_i is the power of the i wave, μ_0 is the linear absorption coefficient and γ_i is the absolute value of the direction cosine of the wavevector of the ith wave with respect to the surface normal of the crystal plate, and Q_{ij} is the linear reflection coefficient of the reflection from the incident i wave to the reflected j wave.

An approximate solution to the power-transfer equation for a mosaic crystal plate is given by (Chang, 2004, expression 5.36)

$$\Delta P_G=\tfrac{1}{2}\sum_{\text{events}}\left(Q_{\text{prim}}l_{s_0}Q_{\text{op}}l_{s_0}-Q_{\text{prim}}l_{s_0}Q_{\text{coop}}l_{s_1}+Q_{\text{op}}l_{s_0}Q_{\text{coop}}l_{s_2}\right), \tag{15}$$

where the subscripts have the same meaning as in the foregoing section and the path lengths of the beam i in the mosaic crystal, l_i, are defined by

$$l_i=T/\gamma_i. \tag{16}$$

In the case of neutron diffraction [$K=1$ in expression (8)] for the rotation about the ψ axis, the products $Q_{ij}l_i$ can be expressed as

$$Q_{ij}l_i=W(\Delta\psi)_{ij}\frac{\lambda}{\sin\gamma_{ij}\cos\chi_{ij}\cos\xi_{ij}}\frac{l_i}{\Lambda_{ij}^2}, \tag{17}$$

where the angles γ, χ and ξ are defined in Ro-92-(Fig. 2) and $W(\Delta\psi)_{ij}$ is the normalized Gaussian describing the mosaic structure of the sample:

$$W(\Delta\psi)_{ij}=\left(\frac{\sin\gamma_{ij}\cos\chi_{ij}\cos\xi_{ij}}{\sin 2\theta_{ij}}\right)\frac{1}{(2\pi)^{1/2}\eta}\times\exp\left(-\frac{\left(\frac{\sin\gamma_{ij}\cos\chi_{ij}\cos\xi_{ij}}{\sin 2\theta_{ij}}\Delta\psi\right)^2}{2\eta^2}\right). \tag{18}$$

The θ_{ij} are the corresponding Bragg angles and η refers to the mosaic spread.

4. Comparison of the two concepts

According to Chang (2004, p. 79), expression (15) is the same as that derived from Renninger's simplest approach in Ro-00*c*. But this is only partly correct, *i.e.* although expression (15) shows some similarity with (13) – even in the case of neutron diffraction [$K=1$ in expression (8)] – the two expressions are not identical. For simplicity, this will be shown for the case of a forbidden primary reflection. From expression (15), we obtain with Ro-92-(8a) and Ro-92-(19*c*)

$$\Delta P_G=\tfrac{1}{2}\frac{\lambda}{(\sin\gamma_{\text{coop}}\cos\chi_{\text{coop}}\cos\xi_{\text{coop}})}(\lambda L_\psi)\times\left\{\frac{l_{s_0}}{\Lambda_{\text{op}}^2}\frac{l_{s_2}}{\Lambda_{\text{coop}}^2}\right\}W(\Delta\psi)_{\text{op}}W(\Delta\psi)_{\text{coop}}, \tag{19}$$

whereas the corresponding dimensionless quantity $I_{\text{Umweg}}^{(\omega,\psi)}f(\psi)/(I_0q_{s_0})$ is given by

$$I_{\text{Umweg}}^{(\omega,\psi)}f(\psi)/(I_0q_{s_0})=(\lambda L_\omega)(\lambda L_\psi)\left\{\frac{\bar{t}_{s_0}}{\Lambda_{\text{op}}^2}\frac{\bar{t}_{s_2}}{\Lambda_{\text{coop}}^2}\right\}f(\psi), \tag{20}$$

i.e. apart from the factor $\frac{1}{2}$ the two expressions (19) and (20) differ in the path lengths, the distribution functions and Lorentz factors. The $\bar{t}_{s_i}$ are the extinction-corrected mean thicknesses of the ideally perfect crystallites (10), whereas the path lengths of the beam i in the mosaic crystal, l_i, are defined by (16). The distribution function $f(\psi)$ defined in (11) depends on the mosaic spread – which of course is zero in the case of a perfect crystal – and crystallite size of the sample as well as on the divergence and wavelength spread of the incident beam, whereas the distribution function $W(\Delta\psi)=W(\Delta\psi)_{\text{op}}W(\Delta\psi)_{\text{coop}}$, being the product of two Gaussians with different heights and widths, depends on the mosaic spread only. Even in this case, the mosaic spread would be the dominating part in the width (12), $f(\psi)$ would differ appreciably from $W(\Delta\psi)$. Last but not least, the Lorentz factor L_ω defined in (9*a*) is not identical with $1/(\sin\gamma_{\text{coop}}\cos\chi_{\text{coop}}\cos\xi_{\text{coop}})$, see Ro-92-(§$B(a)$2) and Ro-92-(§$B(b)$2). All three factors may have a large influence on the calculated intensities of a ψ scan. The multiple diffraction patterns corresponding to expressions (19) and (20) will differ appreciably. This difference will be even more obvious in the case of a non-zero primary reflection, even if the interference term in expression (13) is neglected.

5. Discussion

Chang's statements cited in the *Introduction* that *the appreciable interference ... is therefore dynamical rather than kinematical ... such a term cannot be obtained directly from the power-transfer equation ... the EM wavefields rather than the reflection powers need to be considered* is refuted by the papers Ro-00*c*, Ro-03 and Ro-04. It is correct that *the interference term cannot be obtained from the power-transfer equation* but, as was pointed out in Ro-00*c*, Ro-03 and more explicitly shown in Ro-04, the interference term can be deduced from the kinematical wavefields according to the expressions (1)–(4), *i.e.* '*the addition of the interference term to the expression for integrated intensity*' is '*mathematically rigorous*'. The necessary approximations used for the evaluation of multiple diffraction intensity patterns according to §2 are without any doubt very crude, but so are the approximations used by Moon & Shull (1964) as well as those used in the framework of the dynamic theory for the solution of the multiwave processes.

The applicability of the kinematical concept represented in the second section of this paper was proved in various papers.

In these papers, it was demonstrated by many examples that satisfactory qualitative and quantitative agreement between experimental and theoretical multiple diffraction patterns are obtained with the kinematical approximation presented in §2.

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