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Multiple Scattering of X-Rays and Neutrons I A Recurrence Formula for the Taylor Series Expansion in the Calculation of Intensities*

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This is the first article of a series dealing with experimental and theoretical work on the multiple scattering of X rays and neutrons with emphasis on their application to the study of crystal defects which is a rather new use[1] of a known phenomenon [2]

The exact intensity solution for a crystal plate[1] is too complicated when more than 3 beams are simultaneously diffracted by the crystal Taylor series expansion limited to the first three orders[1,3] have been calculated but the errors due to the approximation were not assessed and the use of higher order terms involves exceedingly complicated expressions

In this paper a general recurrence formula for the successive terms of the Taylor series is presented Thus a rapid calculation of any number of terms is now possible

§1 Introduction

The change in power of an X ray or neutron beam traversing a mosaic crystal plate and experiencing absorption and simultaneous reflections i e, interchanging energy with the primary and other diffracted beams is given by[1]

$$\pm\frac{dP_i}{dx}=-\frac{P_i}{\gamma_i}\mu+\sum_{j=0}^{n}\bar{Q}_{ij}\left(\frac{P_j}{\gamma_j}-\frac{P_i}{\gamma_i}\right), \quad (1)$$

where P_i is the power of beam i γ_i and γ_j are direction cosines of beam i and j relative to the normal plate surface $\bar{Q}_{ij}$ is the effective reflectivity of plane $(i-j)$ μ is the absorption coefficient and x a coordinate taken along the normal to the plate Using eq (1) for the different beams direct and diffracted, involved in the process we obtain a set of simultaneous differential equations[1,3]

$$\pm\frac{dP_0}{dx}=-\frac{P_0}{\gamma_0}(\mu+\bar{Q}_{01}+\sum_j\bar{Q}_{0j})+\frac{P_1}{\gamma_1}\bar{Q}_{10}+\sum\frac{P_j}{\gamma_j}\bar{Q}_{j0}$$

$$\pm\frac{dP_1}{dx}=\frac{P_0}{\gamma_0}\bar{Q}_{01}-\frac{P_1}{\gamma_1}(\mu+\bar{Q}_{10}+\sum\bar{Q}_{1j})+\sum_j\frac{P_j}{\gamma_j}\bar{Q}_{j1}, \quad (2)$$

$$\pm\frac{dP_i}{dx}=\frac{P_0}{\gamma_0}\bar{Q}_{0i}+\frac{P_1}{\gamma_1}\bar{Q}_{1i}-\frac{P_i}{\gamma_i}(\mu+\bar{Q}_{i0}+\bar{Q}_{i1}+\sum_{\substack{j=2\\ j\neq i}}\bar{Q}_{ij})+\sum\frac{P_j}{\gamma_j}\bar{Q}_{ji}$$

The plus sign on the left of eqs (1) and (2) applies to transmitted beams and the minus sign to reflected beams The interaction coefficients $\bar{Q}_{ij}$ are given by

$$\bar{Q}_{ij}=Q_{ij}\ w(\Delta), \quad (3)$$

where Q_{ij} is the reflectivity per unit volume of a small (perfect) crystallite for neutrons and an arbitrary direction of rotation

$$Q_{ij}=\frac{\lambda^3}{v_c^2\sin 2\theta}\frac{|F_{ij}|^2}{A_{ij}}, \quad (4)$$

(in the X ray case two more factors are included $(e^2/mc^2)^2$ and the polarization factor), v_c is the volume of the unit cell, F_{ij} the structure factor of the plane $(i-j)$ that is the plane

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with Miller indices $h_j - f_j$, $k_t - k_j$, $l_t - l_j$, A_{tj} is a geometric factor first introduced by Zachariasen (1945)[4] and further discussed by Imakuma and Caticha Ellis (1974) * relating to the angle of rotation ε_t about an arbitrary axis and the corresponding angle $\Delta\theta$ around an axis normal to the plane of incidence

$$\Delta\theta = A_{tj}\varepsilon_t \tag{5}$$

$w(\Delta)$ is the mosaic angular distribution function generally assumed to be gaussian and isotropic

$$w(\Delta) = \frac{1}{\eta\sqrt{2\pi}} \exp(-\Delta^2/2\eta^2) \tag{6}$$

where Δ is an angular variable measuring the departure of a given crystalline direction $(h_0k_0l_0)$ in each mosaic block with respect to the mean of the direction $(h_0k_0l_0)$ taken for all the crystalline blocks in the crystal. The standard deviation η of the distribution is the mosaic spread of the crystal

It is to be noted that the mosaic distribution (6) is such as to assure that the different contributions that build up a given beam intensity do not have definite phase relationships so that eqs (1) and (2) are obtained by adding intensities and not amplitudes. It is then foreseeable that if the crystal treated turn out to be perfect or nearly so eqs (1) and (2) will cease to be valid and so will the conclusions obtained from their application

The simultaneous eqs (2) can be solved approximately or exactly provided one can establish adequate boundary conditions. This is not easy in general except for geometrically simple cases such as the case of a crystal plate

Zachariasen (1945)[4] found a solution for the two beam case *i.e.* incident and one diffracted beam when the latter is either reflected (Bragg case) or transmitted (Laue case). Bacon and Lowde (1948)[5] extended Zachariasen's result to the neutron two beam case, their results being subsequently used by several authors Wajima *et al.* 1960 [6] Jones 1963 [7] Moon and Shull 1964 [3]. An exact solution for the intensities when the system contains three or more beams becomes increasingly difficult

The problem was solved in general for any numbers of beams in the case of the crystal plate by Caticha Ellis (1969)[1] who also discussed the experimental conditions required to meet the assumptions made in the calculations. The n beam case gives rise to n simultaneous differential equations whose treatment leads to a set of n^2 simultaneous linear equations in the n^2 constants of integration. Then a practical solution for a number of 3 or more beams must involve the use of a computer. K. Imakuma (Thesis 1973)[8] has programmed the numerical calculation in the four beam case

Approximate solutions in the form of Taylor's series expansion introduced by Moon and Shull (1964) [3] are easier to handle provided a small number of terms gives sufficient approximation. This is actually not so except in cases of very small absorption and negligible secondary extinction. In fact the convergence of the series has not been studied in general

Caticha Ellis (1969)[1] has given the 2nd order expansion in the case of a reflected primary beam with n simultaneous secondary diffracted beams of both types as well as a 3rd order expansion for a reflected primary and transmitted secondary beam (see his eqs (16) (21) and (21')). Expansions using higher order terms providing more accurate results are obviously possible but the process of successive derivations is very lengthy and becomes increasingly difficult. A new approach to the calculation was then necessary. This is given in §3

§2 About the Convergence of the Series

The convergence of this series has not so far been studied in general neither we intend to perform such a study here. However Moon and Shull (1964)[3] state that the conditions

$$\bar{Q}_t/l_t \ll 1$$

and

$$\mu l_t \ll 1, \tag{7}$$

are enough to produce a rapid convergence of the series thus providing a reasonable approximation even when the expansion is limited to only the second or the third order terms. Conditions (7) imply that the crystal has a very low secondary extinction and a very low absorption

However conditions (7) are not generally

*K. Imakuma and S. Caticha Ellis to be published

fulfilled in the neutron case and not very often in the X ray case In fact Caticha Ellis (1969)[1] has shown that the total effective path length $\langle x \rangle$ of the incoming plus that of the outgoing beam in a crystal plate of thickness T is given by

$$\frac{\langle x \rangle}{\gamma} = \frac{1}{\mu} - \frac{T}{\gamma} \cdot \frac{\exp(-\mu T/\gamma)}{1-\exp(-\mu T/\gamma)} \qquad (8)$$

where

$$\frac{1}{\gamma} = \frac{1}{\gamma_0} + \frac{1}{\gamma_t}$$

γ_0 γ_t are directions cosines of incident and diffracted beams

$\langle x \rangle/\gamma_0 = l_0$ is the incident beam path length

and

$\langle x \rangle/\gamma_t = l_t$ is the diffracted beam path length

It is clear from eq (8) that for a highly absorbing or a thick crystal $\langle x \rangle/\gamma \simeq 1/\mu$ then for symmetric reflections

$$l_t \simeq \frac{1}{2\mu} \qquad (9)$$

That is for crystals where the path length is limited by absorption $\mu l_t \simeq 0.5$ and the convergence of the series should be slow

In practice values of l_t for most samples in neutron diffraction experiments are defined by the dimensions of the crystal For a plate one would have $l_t = T/\gamma_t$ except when the other dimensions of the plate have to be taken into account

In neutron diffraction values of $\bar{Q}_t l_t$ close to and even larger than unity are not uncommon and we have come across values of about 4 (Parente C B Thesis 1973)[9]

One can limit the values of l_t by using thin plates in the experiment to lower the secondary extinction (Bacon 1948) In the experiments described in part II this procedure was not feasible since the intensity was already quite low due to the high collimation used Thus the conditions (7) for rapid convergence were far from being fulfilled and a second or third order approximation in the Taylor expansion solution was entirely inadequate

Attempts to use higher order approximations were quite discouraging since the successive order of derivations increased the complexity of the expressions to impractical limits

A new approach was then attempted which resulted in the obtention of a general term of the Taylor expansion

§3 General Term of the Taylor Series Expansion

Let us rewrite eq (1) in the form

$$P_t^{(1)}(x) = S_t \sum_{j \neq t} \frac{P_j(x)}{\gamma_j} \bar{Q}_{jt} - S_t P_t(x) \frac{A_t}{\gamma_t} \qquad (1')$$

where

$$A_t = \mu + \sum_{j=0, j \neq t}^{n} \bar{Q}_{tj} \qquad (10)$$

$$S_t = \begin{cases} +1 \text{ for transmitted beams} \\ -1 \text{ for reflected beams} \end{cases}$$

and

$$P_t^{(1)}(x) = \frac{dP_t}{dx}$$

In what follows derivatives will be represented by

$$P_t^{(n)}(x) = \frac{d^n P_t(x)}{dx^n}$$

the symbol $P_t^{(n)}(0)$ representing the value of the derivative at $x=0$ Using this notation the McLaurin series can be written

$$P_t(x) = P_t(0) + x P_t^{(1)}(0) + \frac{x^2}{2!} P_t^{(2)}(0) + \cdots + \frac{x^n}{n!} P_t^{(n)}(0) + \cdots \qquad (11)$$

Let us define the coefficients

$$\left.\begin{aligned} Y_{jt} &= s_t \bar{Q}_{jt}/\gamma_j \quad (j \neq t) \\ Y_{tt} &= -s_t A_t/\gamma_t \quad (j = t) \end{aligned}\right\} \qquad (12)$$

Then eq (1) or (1') can be simply expressed as

$$P_t^{(1)}(x) = \sum_{j=0}^{n} Y_{jt} P_j(x) \qquad (13)$$

where the term $j=t$ is also included in the summation The second derivative is given by

$$P_t^{(2)}(x) = \sum_{j=0}^{n} Y_{jt} P_j^{(1)}(x) = \sum_{j=0} Y_{jt} \sum_{k} Y_{kj} P_k(x) = \sum_{j} \sum_{k} Y_{jt} Y_{kj} P_k(x) \qquad (14)$$

and successive derivatives are obviously given by

$$P_i^{(m)}(x)=\sum_j\sum_k \cdots \sum Y_j\, Y_{kj} \cdots Y\; P_i(x) \tag{15}$$

Apart from numerical factors the terms of the Taylor series expansion will be obtained by multiplying the expressions (13) (14) and (15) by the appropriate powers of x and taking the values of the derivatives at $x=0$

$$\left.\begin{aligned} xP_i^{(1)}(0)&=x\sum_j Y_j\, P_j(0)=\sum_j Y_j\, xP_j^{(0)}\\ x^2P_i^{(2)}(0)&=x^2\sum_j\sum_k Y_j\, Y_{kj}P_k(0)\\ &=\sum_j\sum_k (Y_j x)(Y_{kj}x)P_k(0)\\ x^mP_i^{(m)}(0)&=\sum_j\sum_k \cdots \sum (Y_j\, x)(Y_{kj}x)\\ &\qquad (Y\; x)P(0) \end{aligned}\right\} \tag{16}$$

Equation (16) can be further simplified by defining

$$\left.\begin{aligned} X_{ji}&=xY_{ji}=s_i\bar{Q}_j\, x/\gamma_j\\ X_{ii}&=xY_{ii}=-s\; A_i x/\gamma_i \end{aligned}\right\} \tag{17}$$

Then

$$\left.\begin{aligned} xP_i^{(1)}(0)&=\sum_j X_{ji}P_j(0)\\ x^2P_i^{(2)}(0)&=\sum_j\sum_k X_j\, X_{kj}P_k(0)\\ x^mP_i^{(m)}(0)&=\sum\sum \cdots \sum_{X_z\, P_z(0)} X_j X_{kj} \end{aligned}\right\} \tag{18}$$

Equations (18) show that the successive terms of the series can be expressed as linear combinations of the powers of the different beams at the origin the coefficients of the term m being obtained from the products of order m of the coefficients X_{ij} defined by eqs (17) Thus the coefficients for the m th term can be obtained from those of the $(m-1)$ th term recurrently In order to obtain such a recurrence formula let us define

$$^{(1)}X_{ij}=X_{ij} \tag{19 1}$$

so that

$$xP_i^{(1)}(0)=\sum {}^{(1)}X_k\, P_k(0) \tag{20 1}$$

and

$$x^2P_i^{(2)}(0)=\sum P_k(0)\sum X_{kj}\, {}^{(1)}X_j$$

Then defining

$$^{(2)}X_{ki}=\sum_j X_{kj}\, {}^{(1)}X_j \tag{19 2}$$

the second order term becomes simply

$$x^2P^{(2)}(0)=\sum_k P_k(0)\, {}^{(2)}X_k \tag{20 2}$$

which on turn allows the third order term to be written

$$\begin{aligned} x^3P^{(3)}(0)&=\sum_k P_k(0)\sum_j X_{kj}\, {}^{(2)}Y_j\\ &=\sum_k P_k(0)\, {}^{(3)}X_k \end{aligned} \tag{20 3}$$

where

$$^{(3)}X_{ki}=\sum_j X_{kj}\, {}^{(2)}X_j \tag{19 3}$$

Equations (19 1) (19 2) and (19 3) can obviously be generalized

$$^{(m)}X_{ki}=\sum_s X_{ks}\, {}^{(m-1)}X_{si} \tag{19}$$

and the m th order term takes on the form

$$x^mP_i^{(m)}(0)=\sum_k P_k(0)\, {}^{(m)}X_{ki} \tag{20}$$

Equation (19) is the recurrence formula sought after it will obviously permit a rapid calculation of the terms in succession starting with the coefficients X_{ji} and X_{ii} defined by eqs (17)

Equation (19) can also be applied to the case $m=1$

$$^{(1)}X_{ki}=\sum X_{kj}\, {}^{(0)}X_{ji}$$

provided the $^{(0)}X_{ji}$ are defined as the coefficients for the zero order term

$$P(0)=\sum_k P_k(0)\, {}^{(0)}X_k$$

where

$$^{(0)}X_{ki}=\begin{cases}0 & \text{for } k\neq i\\ 1 & \text{for } k=i\end{cases}$$

It is to be noted that in the absence of anomalous dispersion the reciprocity relations $Y_{ji}=Y_{ij}$ and $X_{ji}=X_{ij}$ hold since in such case $\bar{Q}_{ji}=\bar{Q}_{ij}$

§4 Conclusion

In conclusion an iterative method has been obtained to calculate the successive terms of the Taylor series expansion of the intensity of multiply diffracted X ray or neutron beam

The use of the recurrence formula (19) allows the calculation of the coefficients of the nth order term as soon as those of the $(n-1)$th

term are obtained The method is thus well suited for computer use There is now no need to limit oneself to the approximations of the 2nd or at the most 3rd order as were used in the past Approximations with 100 and 200 terms can be calculated in a matter of seconds of computer time

In a subsequent paper we analyse using this method the neutron intensities multiply scattered by an aluminum single crystal

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