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A general expression of the polarization factor for multi-diffraction processes

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A general expression of the polarization factor of multi-diffracted beams is formulated. By assigning the diffracted beam direction of each diffraction process as the *y* axis of a Cartesian coordinate system, the polarization factor of multi-diffraction processes can be easily calculated for polarized and unpolarized beams without being limited by the number of diffraction processes. The method can be applied to processes with more than three scattering events such as multiple diffraction and extinction.

1. Introduction

Polarization factors have been formulated by several authors (Whittaker, 1953; Azároff, 1955; Vincent, 1982). In these studies the changes of the electric vector of X-rays in the diffraction processes were carefully checked geometrically and polarization factors were derived. Dwiggins (1983) formulated polarization factors by repeated matrix multiplication applicable to multiple coherent scattering of X-rays. However, the transformation matrix T_s of equation (1) of Dwiggins (1983) does not provide a generally applicable expression. The polarization factor of three-wave multiple diffraction processes remained a problem to be solved. It was investigated experimentally by Luh & Chang (1991) and was elaborately formulated by Sheludko (2004, 2005) taking into account the condition for three-wave diffraction to occur. In the present article the polarization factor for a general case is formulated. The *y* axis of the Cartesian coordinates is always taken as the propagating direction of the diffracted beam, as was done by Whittaker (1953). It is assumed that the diffraction for the reflection of which the polarization factor is calculated is clearly defined, say, in the laboratory coordinates. The method can be applied to any multi-diffraction process and can be easily coded into a computer program.

In our studies of multiple diffraction it became evident that the chance of two reciprocal points being simultaneously located on the Ewald sphere is considerable in PtP_2 (Tanaka *et al.*, 1994) and CeB_6 (Makita *et al.*, 2007, 2008) because of the finite size of the reciprocal points. Even in a synchrotron radiation experiment with parallel incident X-rays producing a very narrow full width at half-maximum (FWHM), most of the reflections measured at the bisecting position are seriously affected by multiple diffraction (Takenaka *et al.*, 2010). For crystals with heavy atoms like Pt and Ce, multiple diffraction cannot be neglected. Measurements avoiding it were done by rotating the crystal around the scattering vector using the method of Tanaka & Saito (1975) of indexing the secondary reflections and calculating the intensity perturbation. The perturbation of the intensity is calculated based on the method of Moon & Shull (1964). Since the number of secondary reciprocal points is usually a few tens and sometimes exceeds a hundred, a general expression for the polarization factor for X-rays diffracted more than twice has to be formulated without any restriction to the directions of the relevant beams. Therefore, a general expression not limited by the number of diffraction processes was formulated which allows the polarization factors to be computed in a consecutive manner following the diffraction processes.

2. Cartesian coordinates and transformation between them

Let a beam proceeding along $\mathbf{j}_0$ be diffracted consecutively at O_1, O_2, ..., O_{i-1}, O_i, ..., along the unit vectors $\mathbf{j}_1$, $\mathbf{j}_2$, ..., $\mathbf{j}_{i-1}$, $\mathbf{j}_i$, ..., respectively. Usually $\mathbf{j}_0$ is parallel to the beam from the X-ray source to a monochromator at O_1. Although a double-crystal monochromator is often used in the synchrotron experiments, in the present study the crystal is assumed to be located at O_2 for simplicity. Diffraction processes such as extinction and multiple diffraction take place in the crystal and occur consecutively at O_3, O_4 and so on. When an analyser crystal is used before the counter, it is the last

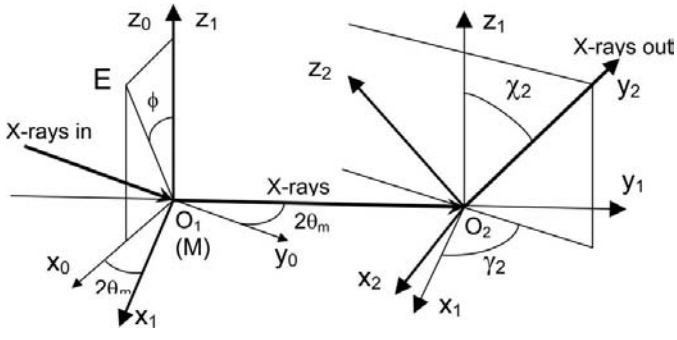

Figure 1
Definition of the Cartesian coordinates for the incident and diffracted beams of the first and second diffraction processes at O_1 and O_2. The monochromator is located at O_1. The incident polarized beam from an X-ray source with the electric vector **E** inclined φ to the $y_0 - z_0$ plane is diffracted at O_1 along y_1. It is diffracted again at O_2 along y_2. z_2 is in the plane of y_2 and z_1. For the process at O_2 and succeeding diffraction processes, the angles χ and γ are the polar coordinates of the unit vector along the diffracted beams defined in terms of the coordinates of the immediately preceding process, as illustrated here.

diffraction point. Cartesian coordinates are attached to each O_i and the unit vectors along the x_i, y_i and z_i axes are defined as $\mathbf{i}_i$, $\mathbf{j}_i$ and $\mathbf{k}_i$. Since relative directions of the diffracted beams determine the polarization factor, translation of these coordinates has no effect on the results. Fig. 1 shows x_1, y_1, z_1 axes at O_1, x_2, y_2, z_2 axes at O_2 and x_1, y_1, z_1 axes at O_2 translated from O_1 to O_2. The x_1, y_1 and z_1 axes at O_2 or at the monochromator are defined so that the beam from the monochromator to the crystal is parallel to the y_1 axis and the z_1 axis is perpendicular to the plane determined by the incident and the diffracted beams at the monochromator. A beam diffracted at O_1 propagates along $\mathbf{y}_1$ and is diffracted at O_2 along $\mathbf{y}_2$. The unit vector $\mathbf{k}_2$ is defined to be in the plane determined by y_2 and z_1 at O_2 in Fig. 1. The relationship between the two coordinates is similar to all the other coordinates at O_3, O_4, ..., O_{i-1}, O_i, and so on. As is stated in the following discussion, the direction of the z_{i-1} axis is determined from the successive diffraction processes at O_1 to O_{i-1} without any ambiguity.

When the polar coordinates of $\mathbf{j}_i(1, \chi_i, \gamma_i)$ are defined on the coordinate system $(x_{i-1}, y_{i-1}, z_{i-1})$ with the origin at O_i as is illustrated in Fig. 1, the orthogonal unit vectors $\mathbf{i}_i$, $\mathbf{j}_i$ and $\mathbf{k}_i$ are expressed in terms of $\mathbf{i}_{i-1}$, $\mathbf{j}_{i-1}$ and $\mathbf{k}_{i-1}$ as

$$\mathbf{j}_i = \sin\chi_i\cos\gamma_i\mathbf{i}_{i-1} + \sin\chi_i\sin\gamma_i\mathbf{j}_{i-1} + \cos\chi_i\mathbf{k}_{i-1}, \quad (1)$$

$$\mathbf{k}_i = -\cos\chi_i\cos\gamma_i\mathbf{i}_{i-1} - \cos\chi_i\sin\gamma_i\mathbf{j}_{i-1} + \sin\chi_i\mathbf{k}_{i-1}, \quad (2)$$

$$\mathbf{i}_i = \mathbf{j}_i \times \mathbf{k}_i = \sin\gamma_i\mathbf{i}_{i-1} - \cos\gamma_i\mathbf{j}_{i-1}. \quad (3)$$

In the following discussion when a vector $\mathbf{v}_m$ is expressed in the ith coordinates, it is denoted as $\mathbf{v}_{m,i}$. From equations (1), (2) and (3) any vector $\mathbf{v}_{m,i}$ is related to the same vector $\mathbf{v}_{m,i-1}$ expressed on the previous coordinates as

$$\mathbf{v}_{m,i} = \mathbf{v}_{m,i-1}T(\chi_i, \gamma_i), \quad (4)$$

$$T(\chi, \gamma) = \begin{pmatrix} \sin\gamma & \sin\chi\cos\gamma & -\cos\chi\cos\gamma \\ -\cos\gamma & \sin\chi\sin\gamma & -\cos\chi\sin\gamma \\ 0 & \cos\chi & \sin\chi \end{pmatrix}. \quad (5)$$

Since $\mathbf{j}_{i,i}$ is (0, 1, 0) by definition, $\mathbf{j}_{i,i-1}$ is expressed from equation (4) as

$$\mathbf{j}_{i,i-1} = \mathbf{j}_{i,i}{}^tT(\chi_i, \gamma_i) = (\sin\chi_i\cos\gamma_i, \sin\chi_i\sin\gamma_i, \cos\chi_i), \quad (6)$$

where ${}^tT$ is the transposed matrix of T. When $\mathbf{j}_{i,i-1} = (x_{i-1}, y_{i-1}, z_{i-1})$ is known, χ_i and γ_i are expressed from equation (6) as

$$\tan\chi_i = (x_{i-1}^2 + y_{i-1}^2)^{1/2}/z_{i-1}, \quad (7)$$

$$\tan\gamma_i = y_{i-1}/x_{i-1}. \quad (8)$$

In general diffraction experiments the diffraction condition of each reflection is always known clearly at least in terms of the laboratory coordinates. The axes at O_1 are translated to O_2 on the crystal as the laboratory coordinates in the present study. When $\mathbf{k}_0$ (= $\mathbf{k}_1$) is defined as $\mathbf{j}_0 \times \mathbf{j}_1$, the angles (χ_1, γ_1) of $\mathbf{j}_1$ at O_1 are $(\pi/2, \pi/2 + 2\theta_m)$, where θ_m is the Bragg angle of the monochromator, and $\mathbf{j}_{1,0}$ becomes

$$\begin{aligned}\mathbf{j}_{1,0} &= \mathbf{j}_{1,1}{}^tT(\chi_1, \gamma_1) = (0, 1, 0)^tT(\pi/2, \pi/2 + 2\theta_m) \\ &= (-\sin 2\theta_m, \cos 2\theta_m, 0). \end{aligned} \quad (9)$$

When the beam from O_1 along $\mathbf{j}_1$ is diffracted at O_2, the direction of the diffracted beam $\mathbf{j}_{2,1}$ becomes, using equations (4) or (6),

$$\begin{aligned}\mathbf{j}_{2,1} &= \mathbf{j}_{2,2}{}^tT(\chi_2, \gamma_2) = (0, 1, 0)^tT(\chi_2, \gamma_2) \\ &= (\sin\chi_2\cos\gamma_2, \sin\chi_2\sin\gamma_2, \cos\chi_2). \end{aligned} \quad (10)$$

Since $\mathbf{j}_{2,1}$ is usually known from the diffraction condition, χ_2 and γ_2 are evaluated using equations (7) and (8). When the diffracted beam along $\mathbf{j}_2$ proceeds to O_3 and is diffracted along $\mathbf{j}_3$, then using equation (4) consecutively

$$\mathbf{j}_{3,3} = \mathbf{j}_{3,2}T(\chi_3, \gamma_3) = \mathbf{j}_{3,1}T(\chi_2, \gamma_2)T(\chi_3, \gamma_3). \quad (11)$$

Since the laboratory coordinates of $\mathbf{j}_{3,1}$ are known and the angles (χ_2, γ_2) are already determined by equation (10), χ_3 and γ_3 are obtained from equations (7) and (8). In this way χ_i and γ_i are calculated consecutively according to the series of the diffraction processes and the transformation of a vector to any of the series of the coordinate systems is possible if the diffraction conditions of the relevant reflections are defined at least in one of the series of the coordinate systems, which is usually the laboratory coordinate system.

3. Polarization factor

Once the matrices T in equation (5) are known, the polarization factor is calculated systematically. In the first process at O_1 the unit vector $\mathbf{k}_0$ is defined to be perpendicular to the incident and diffracted beams, and the incident beam propagating along $\mathbf{j}_0$ is assumed to be polarized with the polarization plane inclined by φ to the plane defined by $\mathbf{j}_0$ and $\mathbf{k}_0$. The electric field vector $\mathbf{E}_0$ with the amplitude E_0 of the incident X-ray is divided into two components $\mathbf{E}_0 = (E_0\sin\varphi, 0, E_0\cos\varphi)$ perpendicular to the propagating direction of the X-rays along $\mathbf{j}_0$. Since the electromagnetic wave does not have a component along the direction y of the propagation, $\mathbf{E}'_{0,i}$ at the ith process on the ith coordinates is calculated from equation (4) as

$$\begin{aligned}\mathbf{E}'_{0,i} &= [E_{0,i}]_{x_i}, (E_{0,i})_{y_i}, (E_{0,i})_{z_i} = \mathbf{E}'_{0,i-1}T(\chi_i, \gamma_i) \\ &= [(E_{0,i-1})_{x_{i-1}}, 0, (E_{0,i-1})_{z_{i-1}}]T(\chi_i, \gamma_i). \end{aligned} \quad (12)$$

$\mathbf{E}_0$ decays after each diffraction process, because $(E_{0,i})_{y_i}$ in equation (12) is not zero. Only $(E_{0,i})_{x_i}$ and $(E_{0,i})_{z_i}$ are transmitted to the next process and are correlated to the polarization effect of the ith diffraction process. Therefore $\mathbf{E}'_{0,i}$ is replaced by $\mathbf{E}''_{0,i}$:

$$\mathbf{E}''_{0,i} = [(E_{0,i})_{x_i}, 0, (E_{0,i})_{z_i}]. \quad (13)$$

The null element of the y_{i-1} component of $\mathbf{E}_{0,i-1}$ of equation (12) and no contribution of $(E_{0,i})_{y_i}$ to the electric field after the ith process indicate the second row and column of T are not necessary for the calculation of the polarization factor. Therefore, all the electric vectors as well as $\mathbf{E}_0$ are truncated hereinafter to two-dimensional ones by deleting the y component as follows:

$$\mathbf{E}_{0,i} = [(E_{0,i})_{x_i}, (E_{0,i})_{z_i}]. \quad (14)$$

$\mathbf{E}_{0,i}$ is expressed on the ith coordinate system using the products of two-dimensional matrices $D(\chi_i, \gamma_i)$ made by removing the second row and column from each T and applying equation (12) i times as

$$\mathbf{E}_{0,i} = \mathbf{E}_{0,i-1}D(\chi_i, \gamma_i) = \mathbf{E}_0\prod_{k=1}^{i} D(\chi_k, \gamma_k) \equiv \mathbf{E}_0U_i. \quad (15)$$

Since the (2,1) element of D is always zero as is evident from equation (5), $(E_{0,i})_{x_i}$ and $(E_{0,i})_{z_i}$ are expressed as

$$(E_{0,i})_{x_i} = E_0\{U_i\}_{11}\sin\varphi \quad (16)$$

$$(E_{0,i})_{z_i} = E_0(\{U_i\}_{12}\sin\varphi + \{U_i\}_{22}\cos\varphi), \quad (17)$$

where $\{U_i\}_{mn}$ is an (m, n) element of the matrix U_i. When the incident beam is polarized and the laboratory system is the jth process $(j < i)$,

the polarization factor p_i of the beam diffracted i times is calculated as

$$\begin{aligned} p_i &= [(E_{0,i})_{x_i}^2 + (E_{0,i})_{z_i}^2]/[(E_{0,j})_{x_j}^2 + (E_{0,j})_{z_j}^2] \\ &= [(\{U_i\}_{11}\sin\varphi)^2 + (\{U_i\}_{12}\sin\varphi + \{U_i\}_{22}\cos\varphi)^2] \\ &\quad \times [(\{U_j\}_{11}\sin\varphi)^2 + (\{U_j\}_{12}\sin\varphi + \{U_j\}_{22}\cos\varphi)^2]^{-1}. \end{aligned} \tag{18}$$

When the incident beam is not polarized, each term in the above equation is averaged for φ as was done by Whittaker (1953) and the polarization factor becomes

$$p_i = (\{U_i\}_{11}^2 + \{U_i\}_{12}^2 + \{U_i\}_{22}^2)/\{U_j\}_{11}^2 + \{U_j\}_{12}^2 + \{U_j\}_{22}^2). \tag{19}$$

4. Examples of the polarization factor of X-rays diffracted more than twice

4.1. Synchrotron experiments with a double-crystal monochromator

In synchrotron experiments a double-crystal monochromator with two parallel planes is often used. When the Bragg angle of the monochromator is $2\theta_m$, the polarization factor of a reflection at a general position is calculated using the matrix U_3,

$$U_3 = D_1(\pi/2, \pi/2 - 2\theta_m)D_2(\pi/2, \pi/2 + 2\theta_m)D_3(\chi_3, \gamma_3). \tag{20}$$

Since $(E_{0,i})_{x_i}^2$ is multiplied by $\sin^2\varphi$ as is evident from equation (16) and since $\sin^2\varphi$ is less than 0.01 when φ is less than 5.74°, the x component of the synchrotron radiation can be neglected when the double-crystal monochromator is used, unless the experiment needs an accuracy better than 1%.

4.2. General experiment affected by extinction and multiple diffraction

When monochromated and unpolarized X-rays are incident on the crystal as in Fig. 1 and the intensity of the primary reflection $\mathbf{h}_2$ is measured, T matrices are

$$T_1 = T(\pi/2, \pi/2 + 2\theta_m), \tag{21}$$

$$T_2 = T(\chi_2, \gamma_2). \tag{22}$$

When $\mathbf{h}_2$ with Bragg angle θ is further diffracted into the incident beam direction, extinction occurs and from equation (4)

$$\mathbf{j}_{33} = \mathbf{j}_{32}T_3 = \mathbf{j}_{31}T_2T_3. \tag{23}$$

Since $\mathbf{j}_{33}$ and $\mathbf{j}_{31}$ are (0, 1, 0),

$$T_2T_3 = I, \tag{24}$$

where I is the unit matrix. However, D_2D_3 is not the unit matrix and the polarization factors of the third beam become, from equation (19) since $\chi_2 = \chi_3 = \pi/2$, $\gamma_2 = \pi/2 - 2\theta$ and $\gamma_3 = \pi/2 + 2\theta$,

$$\begin{aligned} p_3 &= (\{U_3\}_{11}^2 + \{U_3\}_{22}^2)/\{U_1\}_{11}^2 + \{U_1\}_{22}^2) \\ &= (1 + \cos^2 2\theta_m \cos^4 2\theta)/(1 + \cos^2 2\theta_m), \end{aligned} \tag{25}$$

when the incident and primary beams are on the equatorial plane. In an experiment for reflections at a general position, x and z components in equations (16) and (17) should be used for the extinction correction by Becker & Coppens (1974*a*,*b*, 1975).

When a secondary reflection $\mathbf{h}_3$ causes the multiple diffraction, $T_3 = T(\chi_3, \gamma_3)$ is necessary. The polarization factors for the two multiple diffraction processes (Moon & Shull, 1964) look similar but are different. One is the case where the diffracted beam along $\mathbf{j}_2$ is diffracted again into the direction along $\mathbf{j}_3$ as $\mathbf{h}_{23} = \mathbf{h}_3 - \mathbf{h}_2$ reflection (0–2–3 process), and the other is the case where diffracted beam along $\mathbf{j}_3$ is diffracted back to the $\mathbf{j}_2$ direction (0–3–2 process) as $\mathbf{h}_{32} = \mathbf{h}_2 - \mathbf{h}_3$ reflection. $T_1T_2T_3$ and $T_1T_3T_2$ are used for the first and the second cases, respectively. The polarization factors are derived from equations (18) and (19), respectively, for the investigations with laboratory X-rays by Funahashi *et al.* (2010), and for the synchrotron experiment by Takenaka *et al.* (2010).

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