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Polarization factors in the symmetrical case of three-wave diffraction

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Particular results of an unconventional approach to the geometry of multiple diffraction are presented. The scalar relations between polarization components of three waves in the symmetrical case, *i.e.* when the triplet of diffraction vectors forms an isosceles triangle, are considered. The polarization factors are given in simple trigonometric form as functions of the Bragg angle of principal reflection and of a crystallographic parameter, which unambiguously describes such a three-wave configuration.

1. Introduction

The calculation of diffracted intensities in the case of simultaneous diffraction on more than one set of crystallographic planes is based on computational simulations. The algorithms of such simulations, derived from the algorithm of Colella (1974), are highly complex and formalized, and analytical investigations deal mostly with the interrelations between final and interim (such as dispersion surface) results of the calculations. The polarization factors, which describe geometrical correlations between different vector components of interacting electrical fields inside the crystal, are one sort of parameter integrated in a system of fundamental equations of multiple diffraction. Although the importance of an analytical representation of polarization factors has been known for a long time (Saccocio & Zajac, 1965*a*,*b*), there still exists a paradoxical situation where one can provide a wonderful representation of computational calculation results but with quite a shaky analysis of their dependence on the polarization state of the radiation and the geometry of the experiment. As follows from recent discussions on the possibility and the conditions for inversion of the asymmetry of three-wave interference profiles (Juretschke, 1986; Weckert & Hümmer, 1997; Stetsko & Chang, 1999), there is a lack of an analytical description of polarization factors. Although the corresponding vector equations are well known (Stetsko *et al.*, 2004), they are more likely to be a compact formulation of the problem than its solution.

During studies on an unconventional approach to the geometry of multiple diffraction, the author has obtained, as a side result, simple analytical expressions for polarization factors in the symmetrical case. In the general case, the choice of unit polarization vectors is questionable.

2. Unit polarization vectors and results

Consider an optional three-wave reflection $(hkl - h_ik_il_i/h_i^*k_i^*l_i^*)$ with diffraction vector $\mathbf{H} = \mathbf{H}(hkl)$ of principal two-wave reflection and diffraction vectors $\mathbf{H}_i = \mathbf{H}_i(h_ik_il_i)$ of additional and $\mathbf{H}_i^* = \mathbf{H}_i^*(h_i^*k_i^*l_i^*) = \mathbf{H}_i^*(h - h_i, k - k_i, l - l_i) = \mathbf{H} - \mathbf{H}_i$ of coupling reflections, such that $|\mathbf{H}_i| = |\mathbf{H}_i^*|$ as shown in Fig. 1(*a*). The principle of the choice of unit polarization vectors adopted in this communication is illustrated in Fig. 1(*b*), where an optional circle (*circle of configuration*) with its center Li circumscribes one of two possible triplets of reciprocal-lattice points $0, H, H_i$ or $0, H, H_i'$. These triplets form two triple configurations, C_3^i and $C_3^{i'}$, from a partial set of triple configurations C_3 with $|\mathbf{H}_i| = |\mathbf{H}_i^*|$.

Note at this point that, when the points H_i and H_i' belong to the same circle of configuration, there are at least two more points of the reciprocal lattice on the same circle and in this case six-wave intrinsic multiple diffraction takes place (Burbank, 1965; see also Chang, 1984). Another, at least four-wave, case of intrinsic multiple diffraction, being beyond the scope of this paper as the previous one, takes place when the vectors $\mathbf{H}_i$ and $\mathbf{H}_i^*$ are perpendicular.

Let us suppose that C_3^i (or $C_3^{i'}$) is realized in its borderline, the coplanar case (see Chang, 1984; Kshevetskii *et al.*, 1985). In such a case, all reciprocal-lattice points 0, H and H_i, diffraction vectors $\mathbf{H}$, $\mathbf{H}_i$ and $\mathbf{H}_i^*$, and wavevectors $\mathbf{K}_0$, $\mathbf{K}$ and $\mathbf{K}_i$ lie in a principal section of the Ewald sphere. The radius of the sphere is $R_{\rm copl} = \lambda_{3i,\max}^{-1}$, where $\lambda_{3i,\max}$ is the maximal wavelength of diffracted radiation inside the crystal, the center Lo of the sphere coincides with the center Li of the circle of configuration, and the angle $\Sigma_i^* = \Theta_{\max}$ is the maximal Bragg angle for principal reflection (hkl), when the exact geometrical conditions of three-wave diffraction can still be satisfied. In the case under consideration the conventional choice of unit polarization vectors for two-wave diffraction can be adopted: all $\boldsymbol{\sigma}$ vectors lie normal to and all $\boldsymbol{\pi}$ vectors lie in the common diffraction plane and each triplet of the $\mathbf{K}$, $\boldsymbol{\sigma}$ and $\boldsymbol{\pi}$ vectors forms a right-handed orthogonal axes system, as shown in Fig. 1(*b*). The transition to the general case of symmetric three-wave diffraction with $R > R_{\rm copl}$ means shifting of the Ewald sphere center Lo to the right or to the left, along the axis L; the value

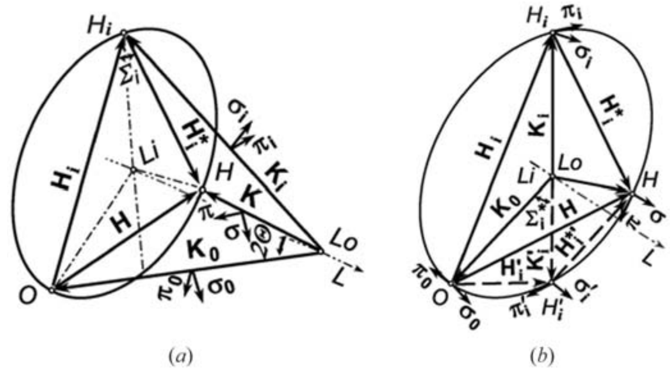

Figure 1
Choice of unit polarization vectors $\boldsymbol{\sigma}$ and $\boldsymbol{\pi}$ for symmetrical three-wave configuration in (*a*) the general and (*b*) the coplanar case

of the Bragg angle Θ decreases, the lengths of all three $\mathbf{K}$ vectors increase and they incline with respect to one another together with the corresponding unit polarization vectors $\boldsymbol{\sigma}$ and $\boldsymbol{\pi}$.

Let us dispose of the uncertainty in the directions of vectors $\boldsymbol{\sigma}$ and $\boldsymbol{\pi}$ by means of the following rule: all $\boldsymbol{\sigma}$ vectors lie normal to diffraction vector $\mathbf{H}$ and all $\boldsymbol{\pi}$ vectors lie parallel to the diffraction plane of principal reflection, formed by the wavevector $\mathbf{K}_0$ of the transmitted wave and the wavevector $\mathbf{K}$ of the diffracted one. In this way, we obtain a system of unit polarization vectors, clearly associated with the azimuthal scan (Renninger, 1937) and the reference-beam diffraction (Shen, 1998) techniques, which means the realization of large numbers of multiple reflections without any change of incident-radiation polarization state.

As mentioned earlier, the side result of an unconventional approach to the geometry of multiple diffraction is presented here. Let us consider the principle of this approach. The angular value Σ_i^* was introduced above as the maximal Bragg angle of the principal reflection (*hkl*) when the reciprocal-lattice points of the *i*th configuration C_3^i lie on the Ewald sphere. This value is also an invariant scalar characteristic of the configuration under consideration. It is really the acute angle between the crystallographic planes that corresponds to the diffraction vectors $\mathbf{H}_i$ and $\mathbf{H}_i^*$ and can be easily calculated as such if all the lattice constants and crystallographic indices $h_ik_il_i$ and $h_i^*k_i^*l_i^*$ are commonly known. Alternatively, the specified angle can be calculated from the angular positions of corresponding multiple diffraction peaks on the azimuthal scan plot.

Taking into account the possibility of distinguishing the cases C_3^i and $C_3^{i\prime}$ in Fig. 1(*b*), we shall use the angular value Σ_i, which is the angle between the outer surfaces of the planes, *i.e.* $\Sigma_i = \Sigma_i^*$ for the C_3^i case and $\Sigma_i = \pi - \Sigma_i^*$ for the $C_3^{i\prime}$ case.

Now, following Saccocio & Zajac (1965*b*) and Joko & Fukuhara (1967) (see also Chang, 1984), *i.e.* leaving out lengthy trivial trigonometric transformations, we can represent the final results as follows,

$$\gamma_1 = \boldsymbol{\pi}_0\boldsymbol{\pi} = \cos 2\Theta, \qquad (1)$$

$$\gamma_2 = \boldsymbol{\pi}_0\boldsymbol{\pi}_i = \boldsymbol{\pi}_i\boldsymbol{\pi} = -\operatorname{sign}(\cos\Sigma_i)\cos\Theta, \qquad (2)$$

$$\gamma_3 = \boldsymbol{\sigma}_0\boldsymbol{\sigma}_i = \boldsymbol{\sigma}_i\boldsymbol{\sigma} = \operatorname{sign}(\cos\Sigma_i)\frac{\cos\Sigma_i - \cos^2\Theta}{\cos\Theta(1-\cos\Sigma_i)}, \qquad (3)$$

$$\gamma_4 = \boldsymbol{\sigma}_i\boldsymbol{\pi}_0 = -\boldsymbol{\sigma}_i\boldsymbol{\pi} = \frac{\sin^2\Theta(\cos^2\Theta - \cos^2\Sigma_i)^{1/2}}{\cos\Theta(1-\cos\Sigma_i)}, \qquad (4)$$

$$\gamma_5 = \boldsymbol{\sigma}_0\boldsymbol{\pi} = \boldsymbol{\sigma}\boldsymbol{\pi}_0 = \boldsymbol{\sigma}_0\boldsymbol{\pi}_i = \boldsymbol{\sigma}\boldsymbol{\pi}_i = 0$$

and

$$\gamma_6 = \boldsymbol{\sigma}_0\boldsymbol{\sigma} = 1.$$

This is different from the similar results given by Saccocio & Zajac (1965*b*) for $\Sigma_i = \pi/3$, in addition to the change of sign in (2) (because of the insignificant difference in the choice of unit polarization vectors). Here we see in (3) and (4) only the constant crystallographic value Σ_i and only a directly measured Bragg angle Θ. In the above-mentioned work, γ_3 and γ_4 were represented as functions of Θ and a variable, which functionally depends upon Θ, crystallographic parameters and crystallographic indices and was simple to calculate in some special cases only. For example, taking into account $\cos(\pi/3) = 0.5$, one obtains immediately from (1)–(4) the simple relations between polarization factors, given in this work.

In the particular case of $\cos\Sigma_i = 0$, which corresponds to the exceptional case of four-wave intrinsic multiple diffraction, one obtains $\gamma_3 = \pm\cos\Theta$ and $\gamma_4 = \sin^2\Theta$, which coincides, taking into account the difference in terms $\boldsymbol{\sigma}$ and $\boldsymbol{\pi}$, with the result given by Joko & Fukuhara (1967).

Let us consider now the last verification example. As can be seen from $\gamma_3 = 0$ in (3), the total suppression of the $\boldsymbol{\sigma}$-polarized component of diffracted radiation with wavevector $\mathbf{K}_i$ takes place for $\boldsymbol{\sigma}$-polarized incident radiation under condition $\Sigma_i = \cos^2\Theta$ and, consequently, when γ_4 reaches its extreme value, for fixed angle Θ, $|\gamma_4| = \sin\Theta$. In this three-wave case, there should not arise any effect on the two-wave intensity for σ-polarized incident radiation. An example of such a case is the Ge$(220 - 115/11\bar{5})$ reflection with $\Sigma_i = 31.59^\circ$ and $\Theta = 22.65^\circ$ for Cu $K\alpha$ radiation, and the results of computations, represented by Stetsko & Chang (1999) for this reflection, are in perfect agreement with our conclusion.

Thus, one can use our results for solving more complex tasks. One of these tasks, which at the moment does not have an analytical solution in spite of intensive discussion, is the problem of the possibility and conditions of the appearance of inverse three-wave peaks asymmetry (see *Introduction*). Omitting the redundant consideration of its essentials, which was given by Juretschke (1986), Shen (1986), Weckert & Hümmer (1997), Stetsko & Chang (1999) and Stetsko *et al.* (2004), let us consider only the shortest final formulation of the problem: an analytical description must be obtained of conditions that correspond to the equality $\operatorname{sign}(P^{\pi}_{\rm dir}) = -\operatorname{sign}(P^{\pi}_{\rm um})$, where $P^{\pi}_{\rm dir} = \gamma_1$ and $P^{\pi}_{\rm um} = \gamma_2^2 - \gamma_4^2$.

Note that, on the poles of the Ewald sphere (points H_i and H_i' in Fig. 1*b*), $\Sigma_i = \Theta$, $\Sigma_i' = \pi - \Theta$ and $P^{\pi}_{\rm um} = \cos^2\Theta > 0$, but at the equatorial points, where $\cos\Sigma_i = \cos^2\Theta$ is fulfilled (see above), we obtain $P^{\pi}_{\rm um} = \cos 2\Theta$, *i.e.* the signs of $P^{\pi}_{\rm dir}$ and $P^{\pi}_{\rm um}$ can differ at least for $\Theta > \pi/4$.

Further, using (2) and (4) for $P^{\pi}_{\rm um} = 0$, we obtain an equation for the points, in which $P^{\pi}_{\rm um}$ changes its sign,

$$(\tan^4\Theta + 1)\cos^2\Sigma_i - 2\cos\Sigma_i - (\tan^4\Theta\cos^2\Theta - 1) = 0,$$

and easily find the solution of the problem as

$$\Sigma_i^{(\pm)} = \arccos\{[1 \pm \tan^3\Theta(-\cos 2\Theta)^{1/2}]/(1 + \tan^4\Theta)\}. \qquad (5)$$

Thus, in conclusion, for π-polarized incident radiation the azimuthal scan peaks, which correspond to a triplet configuration from a partial set of symmetrical triplet configurations C_3, have inverse asymmetry if and only if the following conditions are valid: $\Theta > \pi/4$ and either $\Sigma_i < \Sigma_i^{(+)}$ or $\Sigma_i > \Sigma_i^{(-)}$, where $\Sigma_i^{(\pm)}$ are the solutions of equation (5).

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