Acta Crystallographica Section A
Foundations of Crystallography
ISSN 0108-7673

Received 20 November 2003
Accepted 18 December 2003

'Exact' formulation for π-polarization waves of dynamical X-ray diffraction

Wondong Cho,* XianRong Huang and Michael Dudley

Department of Materials Science and Engineering, State University of New York at Stony Brook, Stony Brook, NY 11794-2275, USA. Correspondence e-mail: wocho@ic.sunysb.edu

Self-consistent formulae for treating the π-polarization waves in two-beam X-ray diffraction from single crystals are refined within the Laue formalism. These formulae overcome the $\nabla \cdot \mathbf{D} \neq 0$ problem and avoid all the unnecessary approximations in previous dynamical theories. Therefore, they are as accurate as the formulae for σ-polarization waves.

1. Introduction

The extended dynamical theory of two-beam X-ray diffraction developed in the literature is almost rigorous for σ polarization except for some unavoidable approximations (*e.g.* De Caro *et al.*, 1997). For the π polarization, however, a well known approximation commonly adopted is that the polarization factor C, the cosine of the 'internal scattering angle', is assumed to be constant, equal to $\cos 2\theta_B$ (θ_B the Bragg angle), which is unjustified at the far tails of the diffraction maximum. Huang & Dudley (2003, hereafter referred to as paper I) recently improved this approximation by taking into account the variation of C with varying incidence direction. However, the mathematical treatments are still not 'perfect', as illustrated in the following.

Consider the coplanar Bragg case in Fig. 1, where the xy and xz planes are the crystal surface and the plane of incidence, respectively. Above the surface exist the incident wave $\mathbf{E}_I \exp(-i\mathbf{K}_0 \cdot \mathbf{r})$, the diffracted wave $\mathbf{E}_D \exp(-i\mathbf{K}_h \cdot \mathbf{r})$ and the specularly reflected wave $\mathbf{E}_R \exp(-i\mathbf{K}_R \cdot \mathbf{r})$ [all with a common time dependence $\exp(i\omega t)$]. The magnitudes of $\mathbf{K}_0$, $\mathbf{K}_h$ and $\mathbf{K}_R$ are all equal to $K = 2\pi/\lambda$ (λ is the wavelength). For each wavefield $\mathbf{D}_0 \exp(-i\mathbf{k}_0 \cdot \mathbf{r}) + \mathbf{D}_h \exp(-i\mathbf{k}_h \cdot \mathbf{r})$ inside the crystal, the two wavevectors are (see paper I)

$$\mathbf{k}_0 = \mathbf{K}_0 + K\delta\hat{\mathbf{z}} = k_{0x}\hat{\mathbf{x}} + k_{0z}\hat{\mathbf{z}}, \quad (1)$$

$$\mathbf{k}_h = \mathbf{K}_0 + \mathbf{h} + K\delta\hat{\mathbf{z}} = k_{hx}\hat{\mathbf{x}} + k_{hz}\hat{\mathbf{z}}, \quad (2)$$

where $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$ and $\hat{\mathbf{z}}$ are unit vectors along the x, y and z axes, respectively, and $\delta = \delta_r + i\delta_i$ (with δ_r and δ_i being real) is a small complex variable that can be solved from the dispersion equation. The complex δ makes k_{0z} and k_{hz} also complex (while k_{0x} and k_{hx} are real). Hence, $\mathbf{k}_0$ and $\mathbf{k}_h$ become *complex vectors* (Born & Wolf, 1999). For instance, $\mathbf{k}_0$ has the form $\mathbf{k}_0 = \mathbf{k}_{0r} + ik_{0i}\hat{\mathbf{z}}$, where $k_{0i} = K\delta_i$ and $\mathbf{k}_{0r} = \mathbf{K}_0 + K\delta_r\hat{\mathbf{z}}$. Generally, $\mathbf{k}_{0r}$ is not parallel to $\hat{\mathbf{z}}$.

Note that here a *complex vector* is defined to have the form $\mathbf{v} = v_1\hat{\mathbf{e}}_1 + v_2\hat{\mathbf{e}}_2$, where $\hat{\mathbf{e}}_1$ and $\hat{\mathbf{e}}_2$ are two different unit vectors while $v_1 = |v_1|\exp(i\phi_1)$ and $v_2 = |v_2|\exp(i\phi_2)$ are two complex quantities with different phases ϕ_1 and ϕ_2. Such a vector may also be written as $\mathbf{v} = \mathbf{v}_r + i\mathbf{v}_i$ with $\mathbf{v}_r = \mathrm{Re}(v_1)\hat{\mathbf{e}}_1 + \mathrm{Re}(v_2)\hat{\mathbf{e}}_2$ and $\mathbf{v}_i = \mathrm{Im}(v_1)\hat{\mathbf{e}}_1 + \mathrm{Im}(v_2)\hat{\mathbf{e}}_2$ being the real and imaginary components. In contrast, a *single vector* has the form $\mathbf{u} = u\hat{\mathbf{e}}$, where $\hat{\mathbf{e}}$ is a unit vector and u is a real or complex value, called the *scalar value* of $\mathbf{u}$. When $\mathbf{u}$ is resolved into two or more components, all the components always have the same phase.

In the literature, the amplitude $\mathbf{D}_0$ of any internal primary wave was assumed to be a single vector, *i.e.* it can be written as $D_0\hat{\mathbf{e}}_0$ with $\hat{\mathbf{e}}_0$ being a unit vector. For the σ polarization, since $\mathbf{k}_0$ lies within the xz plane while $\hat{\mathbf{e}}_0 = \hat{\mathbf{y}}$ is perpendicular to this plane, we always have $\nabla \cdot \mathbf{D}_0 = -i\mathbf{k}_0 \cdot \mathbf{D}_0 = 0$, which is the basic requirement used to derive Laue's wave equations (no free charge exists). For π polarization, however, $\hat{\mathbf{e}}_0$ also lies within the xz plane. Thus, we have $\mathbf{k}_0 \cdot \mathbf{D}_0 = D_0(\mathbf{k}_{0r} + ik_{0i}\hat{\mathbf{z}}) \cdot \hat{\mathbf{e}}_0 \neq 0$ unless $\mathbf{k}_{0r} \parallel \hat{\mathbf{z}}$ and $\hat{\mathbf{e}}_0 \perp \hat{\mathbf{z}}$ (normal incidence). Clearly, the internal diffracted waves have the same problem. Therefore, the previous dynamical theory for π polarization is not self-consistent (at least mathematically).

The other problem for π-polarization waves is that the 'internal scattering angle' between $\mathbf{k}_0$ and $\mathbf{k}_h$ is not strictly defined as each complex vector consists of two different vectors. In paper I, the cosine of this 'angle' was treated as $C = (\mathbf{k}_0 \cdot \mathbf{k}_h)/k_0k_h$ with k_0 and k_h being the 'scalar values' of $\mathbf{k}_0$ and $\mathbf{k}_h$, respectively. However, the 'scalar value' of a complex vector is not unambiguously defined, either (but $k_m^2 = \mathbf{k}_m \cdot \mathbf{k}_m$ is well defined). Although an intuitive treatment might be $k_m = \pm(\mathbf{k}_m \cdot \mathbf{k}_m)^{1/2}$ (for $m = 0, h$), the physical meaning of such a representation is not clear. Meanwhile, there is no criterion about how to choose the sign. Detailed discussions about complex vector algebra is out of the scope of this paper. Instead, we will demonstrate in the following that the π-polarization waves can be naturally and rigorously formulated without breaking the $\nabla \cdot \mathbf{D} = 0$ condition or using the 'internal scattering angle'.

2. Theory

Let us write the internal wave amplitudes as $\mathbf{D}_0 = D_{0x}\hat{\mathbf{x}} + D_{0y}\hat{\mathbf{y}} + D_{0z}\hat{\mathbf{z}}$ and $\mathbf{D}_h = D_{hx}\hat{\mathbf{x}} + D_{hy}\hat{\mathbf{y}} + D_{hz}\hat{\mathbf{z}}$. The first

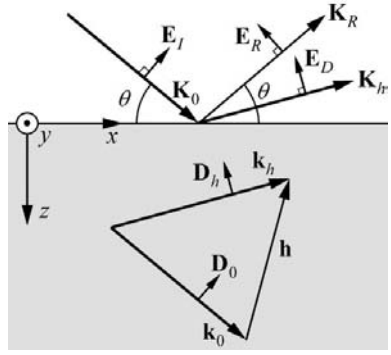

Figure 1
Coplanar Bragg-case X-ray diffraction geometry. The **D** and **E** vectors are for π polarization. $\mathbf{D}_0$, $\mathbf{D}_h$, $\mathbf{k}_0$ and $\mathbf{k}_h$ are complex vectors.

 DOI: 10.1107/S0108767303029362

restriction for $\mathbf{D}_0$ and $\mathbf{D}_h$ is that they must be transverse, *i.e.* $\mathbf{k}_0 \cdot \mathbf{D}_0 = 0$ and $\mathbf{k}_h \cdot \mathbf{D}_h = 0$. As a result, we have

$$D_{0z} = -k_{0x}D_{0x}/k_{0z}, \quad D_{hz} = -k_{hx}D_{hx}/k_{hz}. \tag{3}$$

Here, since k_{mz} ($m = 0$ or h) is complex, the phases of D_{mx} and D_{mz} are different. Thus, $\mathbf{D}_m$ is also a complex vector, which indicates that the wave $\mathbf{D}_m \exp(-i\mathbf{k}_m \cdot \mathbf{r})$ of π polarization is not strictly a plane wave. The physical meaning of the complex amplitude vector is that both the direction and magnitude of this vector change periodically with time t in real space, *i.e.* $\mathbf{D}_m(t) = |D_{mx}|\cos(\omega t)\hat{\mathbf{x}} + |D_{mz}|\cos(\omega t + \phi_m)\hat{\mathbf{z}}$, where ϕ_m is the phase difference between D_{mz} and D_{mx} and ω is the circular frequency of the wave. This is similar to an elliptical wave except that the polarization plane is parallel to the wavevector.

The other restriction for $\mathbf{D}_0$ and $\mathbf{D}_h$ is that they must satisfy Laue's wave equations

$$\varsigma_0 \mathbf{D}_0 = \chi_{\bar{h}} \mathbf{D}_{h[0]}, \tag{4}$$

$$\varsigma_h \mathbf{D}_h = \chi_h \mathbf{D}_{0[h]}, \tag{5}$$

where

$$\varsigma_0 = \chi_p - K^2/k_0^2, \quad \varsigma_h = \chi_p - K^2/k_h^2, \tag{6}$$

and $\chi_p = 1 - \chi_0$. Equations (4) and (5) are based on the fact that the susceptibility χ is very small so that $1/(1-\chi) \simeq 1 + \chi$ (which could be invalid for soft X-ray diffraction). Meanwhile, the possible contributions from other reflections are ignored (Holý & Fewster, 2003). Currently, we are unable to avoid these approximations.

The exact definitions of $\mathbf{D}_{h[0]}$ and $\mathbf{D}_{0[h]}$ are

$$\begin{aligned}\mathbf{D}_{h[0]} &= \mathbf{D}_h - (\mathbf{D}_h \cdot \mathbf{k}_0)\mathbf{k}_0/k_0^2 \\ &= D_{hy}\hat{\mathbf{y}} + (k_{0z}D_{hx} - k_{0x}D_{hz})(k_{0z}\hat{\mathbf{x}} - k_{0x}\hat{\mathbf{z}})/k_0^2, \end{aligned} \tag{7}$$

$$\begin{aligned}\mathbf{D}_{0[h]} &= \mathbf{D}_0 - (\mathbf{D}_0 \cdot \mathbf{k}_h)\mathbf{k}_h/k_h^2 \\ &= D_{0y}\hat{\mathbf{y}} + (k_{hz}D_{0x} - k_{hx}D_{0z})(k_{hz}\hat{\mathbf{x}} - k_{hx}\hat{\mathbf{z}})/k_h^2. \end{aligned} \tag{8}$$

Based on these two equations, the scalar forms of (4) and (5) along the y axis are

$$\varsigma_0 D_{0y} - \chi_{\bar{h}} D_{hy} = 0, \quad \chi_h D_{0y} - \varsigma_h D_{hy} = 0, \tag{9}$$

respectively. These two equations correspond to the σ-polarization configuration. Since this case has been accurately treated in paper I, we skip the related discussions here.

Based on (7), the scalar forms of (4) along the x and z axes are

$$\begin{aligned}\varsigma_0 D_{0x} - \chi_{\bar{h}} k_{0z}(k_{0z}D_{hx} - k_{0x}D_{hz})/k_0^2 = 0, \\ \varsigma_0 D_{0z} + \chi_{\bar{h}} k_{0x}(k_{0z}D_{hx} - k_{0x}D_{hz})/k_0^2 = 0, \end{aligned} \tag{10}$$

respectively. Eliminating D_{0z} and D_{hz} using (3), one can find that these two equations are identical and may be written as

$$\varsigma_0 D_{0x} - \chi_{\bar{h}} k_{0z}(\mathbf{k}_0 \cdot \mathbf{k}_h)D_{hx}/(k_0^2 k_{hz}) = 0. \tag{11}$$

Similarly, the scalar forms of (5) along x and z are also identical:

$$\chi_h k_{hz}(\mathbf{k}_0 \cdot \mathbf{k}_h)D_{0x}/(k_h^2 k_{0z}) - \varsigma_h D_{hx} = 0. \tag{12}$$

Equations (11) and (12) are the two wave equations for π polarization, from which the dispersion equation is then, also by (6),

$$(k_0^2\chi_p - K^2)(k_h^2\chi_p - K^2) = \chi_h\chi_{\bar{h}}(\mathbf{k}_0 \cdot \mathbf{k}_h)^2. \tag{13}$$

This equation is identical to the dispersion equation (3) in paper I under $C^2 = (\mathbf{k}_0 \cdot \mathbf{k}_h)^2/(k_0^2 k_h^2)$. Here we have explicitly proved it. As a consequence, we may use the same procedure as that used in paper I to convert (13) into a quartic equation in terms of δ:

$$\delta^4 + A_1^\pi\delta^3 + A_2^\pi\delta^2 + A_3^\pi\delta + A_4^\pi = 0. \tag{14}$$

The four coefficients in (14) have been defined in equations (11) of paper I,[1] where it has also been illustrated how the four roots δ_j ($j = 1, 2, 3, 4$) of (14) may be solved using simple complex arithmetic. After each δ_j is solved, the $\mathbf{k}_{0j}$ and $\mathbf{k}_{hj}$ and their components can be calculated from (1) and (2). Meanwhile, the corresponding k_{mj}^2 [$= k_{mjx}^2 + k_{mjz}^2$] and ς_{mj} can also be obtained ($m = 0, h$). From (12), the ratio between D_{hjx} and D_{0jx} is then

$$r_j = \frac{D_{hjx}}{D_{0jx}} = \frac{\chi_h k_{hjz}}{\varsigma_{hj} k_{hj}^2}\left(k_{hjz} + \frac{k_{0jx}k_{hjx}}{k_{0jz}}\right). \tag{15}$$

The strengths of the four wavefields are determined by the boundary conditions: the continuity of the tangential components of the **E** and **H** fields across the crystal surface (or interfaces). From the relation $\mathbf{E} \simeq (1 - \chi)\mathbf{D}$ (in c.g.s. units), one may convert the tangential components of the internal **D** waves into **E**-wave components using

$$\begin{aligned}E_{0jx} &= (\chi_p - r_j\chi_{\bar{h}})D_{0jx} = p_j D_{0jx}, \\ E_{hjx} &= (r_j\chi_p - \chi_h)D_{0jx} = q_j D_{0jx}, \end{aligned} \tag{16}$$

for $j = 1, 2$. In vacuum, $\mathbf{D} \equiv \mathbf{E}$. Therefore, we have $E_{Ix} = \gamma_0 E_I$, $E_{Rx} = -\gamma_0 E_R$ and $E_{Dx} = \gamma_h E_D$ with $\gamma_0 = K_{0z}/K = \sin\theta$ and $\gamma_h = K_{hz}/K = -[K^2 - (K_{0x} + h_x)^2]^{1/2}/K$. For a plane wave $\mathbf{D} = \mathbf{D}_0 \exp[i(\omega t - \mathbf{k} \cdot \mathbf{r})]$, Maxwell's equations $c\nabla \times \mathbf{H} = \partial\mathbf{D}/\partial t$ and $\nabla \cdot \mathbf{H} = 0$ lead to $\mathbf{H} = (K/k^2)(\mathbf{k} \times \mathbf{D})$. Based on this principle and equations (3) and (15), the tangential components of the internal **H** fields are

$$\begin{aligned}H_{0jy} &= D_{0jx}K/k_{0jz} = u_j D_{0jx}, \\ H_{hjy} &= r_j D_{0jx}K/k_{hjz} = v_j D_{0jx}. \end{aligned} \tag{17}$$

According to Fig. 1, the **H** fields of the external waves are all parallel to the y axis with $H_{Iy} = E_I$, $H_{Ry} = E_R$ and $H_{Dy} = E_D$.

In the above treatments, one can see that explicit use of the 'internal scattering angle' (as well as the polarization factor) is completely avoided, and it is no longer necessary to calculate the 'scalar value' of any complex vector involved. Meanwhile, all the **D** waves inside the crystal satisfy $\nabla \cdot \mathbf{D} = 0$.

3. Discussion and conclusions

Our numerical computations show slight differences between the rocking curves calculated with the current method and that in paper I for the π polarization. The similarity originates from the fact that the imaginary parts of the internal wavevectors are actually quite small. However, for the far tails of the rocking curve, the simulated intensity profile shows significant differences from that computed by assuming a constant polarization factor $C = \cos 2\theta_B$. As an example, let us consider π-polarization Bragg diffraction from a multilayer structure consisting of N layers on a substrate. Suppose that the multilayers are stacked above the xz plane in Fig. 1 along $-\mathbf{z}$. Then the diffraction pattern may be calculated with the 4×4 matrix equation

$$\tilde{\mathbf{D}}_v = \mathbf{S}_U^{(N)}(\mathbf{S}_L^{(N)})^{-1}\mathbf{S}_U^{(N-1)}(\mathbf{S}_L^{(N-1)})^{-1}\ldots\mathbf{S}_U^{(1)}(\mathbf{S}_L^{(1)})^{-1}\tilde{\mathbf{D}}_0, \tag{18}$$

where the superscripts (n) are the indices of the epilayers starting from 1 to N along $-\mathbf{z}$ (0 for the substrate). The matrices in (18) are defined as

[1] Note that the correct expression for A_3^π is $A_0A_3^\pi = 2\chi_p^2[\gamma_0(\alpha+1) + \varphi_h] - A_1^\pi(\chi_p + \chi_h\chi_{\bar{h}}\beta)$.

$$\mathbf{S}_{U,L}^{(n)} = \begin{pmatrix} p_1^{(n)} w_{1,n}^{U,L} & p_2^{(n)} w_{2,n}^{U,L} & p_3^{(n)} w_{3,n}^{U,L} & p_4^{(n)} w_{4,n}^{U,L} \\ q_1^{(n)} w_{1,n}^{U,L} & q_2^{(n)} w_{2,n}^{U,L} & q_3^{(n)} w_{3,n}^{U,L} & q_4^{(n)} w_{4,n}^{U,L} \\ u_1^{(n)} w_{1,n}^{U,L} & u_2^{(n)} w_{2,n}^{U,L} & u_3^{(n)} w_{3,n}^{U,L} & u_4^{(n)} w_{4,n}^{U,L} \\ v_1^{(n)} w_{1,n}^{U,L} & v_2^{(n)} w_{2,n}^{U,L} & v_3^{(n)} w_{3,n}^{U,L} & v_4^{(n)} w_{4,n}^{U,L} \end{pmatrix}, \quad (19)$$

where $w_{j,n}^{U,L} = \exp(iK\delta_j^{(n)} z_n^{U,L})$, $z_n^L = -\sum_{m=1}^{n-1} t_m$ and $z_n^U = z_n^L - t_n$ are the positions of the lower and upper interfaces of the nth layer, respectively ($z_1^L = 0$), and t_m is the thickness of the mth layer. $\tilde{\mathbf{D}}_v$ and $\tilde{\mathbf{D}}_0$ in (18) are two column vectors (for $E_I = 1$):

$$\tilde{\mathbf{D}}_v = \begin{pmatrix} \gamma_0(1 - E_R) \\ \gamma_h \Phi_D E_D \\ 1 + E_R \\ \Phi_D E_D \end{pmatrix}, \quad \tilde{\mathbf{D}}_0 = \begin{pmatrix} p_1^{(0)} D_{01x}^{(0)} + p_2^{(0)} D_{02x}^{(0)} \\ q_1^{(0)} D_{01x}^{(0)} + q_2^{(0)} D_{02x}^{(0)} \\ u_1^{(0)} D_{01x}^{(0)} + u_2^{(0)} D_{02x}^{(0)} \\ v_1^{(0)} D_{01x}^{(0)} + v_2^{(0)} D_{02x}^{(0)} \end{pmatrix}, \quad (20)$$

where $\Phi_D = \exp[iK(\gamma_0 - \gamma_h) z_N]$ and $D_{01x}^{(0)}$ and $D_{02x}^{(0)}$ are associated with the two valid wavefields in the substrate. E_R, E_D, $D_{01x}^{(0)}$ and $D_{02x}^{(0)}$ can be solved from the four linear equations in (18) for any incidence angle θ. Note that for grazing-incidence geometry the recursion-matrix method might be needed [see Stepanov *et al.* (1998) and references therein].

Fig. 2 shows the rocking curves simulated for a 20-period AlAs (154 Å)/GaAs (73 Å) superlattice on a (001) GaAs substrate (Stepanov *et al.*, 1998). The solid-line pattern was simulated using the above 4 × 4 transfer-matrix method (4F) while the dashed one was computed using the conventional 2 × 2 transfer-matrix method (2F) (upon which most commercial software is based). Apparently, only when the incidence angle falls within the range $|\Delta\theta| < 1000''$ ($\sim 0.3^\circ$) around the Bragg angle of the substrate do the two patterns coincide very well (Fig. 2*a*). Out of this range, the displacement of the dashed pattern tends to be significant with increasing $|\Delta\theta|$ (Fig. 2*b*). In wide angular ranges, the discrepancy caused by the 2F method is intolerable. Here the 4F method nicely solved this problem. It should be noted that in the 4F method the π-polarization superlattice peaks still exactly coincide with the σ-polarization peaks (not shown here) over the entire angular range. However, the former peaks are generally one order lower than the latter ones although the substrate peak heights for the two polarization states are very close to each other. This phenomenon does not mean that the π-polarization contribution could be ignored in simulations of superlattice (or multilayer) diffraction rocking curves for unpolarized laboratory X-ray sources. Instead, the (weighted) average of both contributions must be used to give accurate simulations.

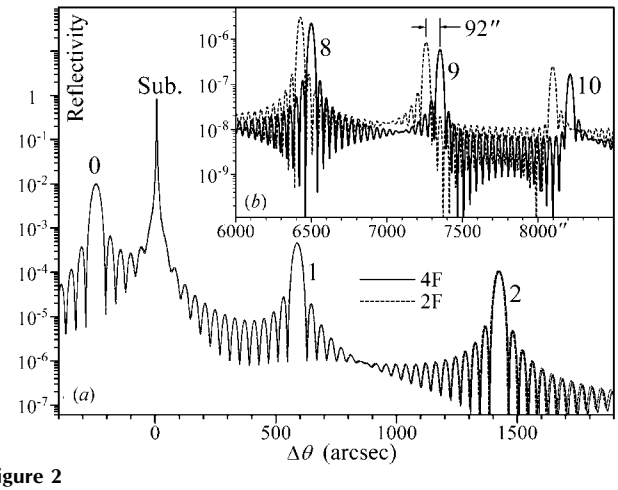

Figure 2
Simulated rocking curves for a pseudomorphic AlAs/GaAs superlattice, vertical lattice constant ratio assumed to be $a_{\rm AlAs}/a_{\rm GaAs} = 1.002775$. Symmetric 004 reflection, π polarization, Cu $K\alpha_1$ radiation.

In summary, we have derived simple but rigorous formulae for treating the π-polarization waves in the two-beam X-ray diffraction process. This model is mathematically self-consistent and is as accurate as the existing extended dynamical theory for σ-polarization waves. Similar extensions can also be developed for multiple-beam diffraction although the computations might be expensive.

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