# SHORT COMMUNICATIONS

*Acta Cryst.* (1998). A**54**, 1034–1036

## Darwin's approach to X-ray dynamical diffraction in distorted crystals

I. R. Prudnikov *at Physics Department, M. V. Lomonosov Moscow State University, Moscow 119899, Russia.*
*E-mail: prudnik@xray.phys.msu.su*

(*Received 24 March 1998; accepted 29 May 1998*)

**Abstract**

The well known Darwin approach is generalized to study the problem of X-ray dynamical diffraction in distorted crystals with a three-dimensional deformation field of arbitrary form. The recursion equations describing X-ray diffraction in the above crystals have been derived. The analytical solution of these equations has been obtained within the kinematical approximation.

At present, there exist at least two different methods that are widely used in theoretical studies of X-ray dynamical diffraction from distorted crystals. One method is based on the Takagi–Taupin differential equations (Taupin, 1964; Takagi, 1969) and the other on the recursion equations for the reflection and transmission coefficients of layered crystals. The recursion equations were first obtained by Darwin (1914) for solving the problem of Bragg-case dynamical diffraction by perfect crystals. Then Borie (1967) demonstrated that Darwin's approach could be extended to Laue-case diffraction. In fact, the method based on recursion formulae is a convenient approach to the problem of X-ray scattering from layered crystals and multilayered materials such as superlattices, heterostructures *etc.* [see *e.g.* Belyaev & Kolpakov, 1983; Vardanyan *et al.*, 1985; Bartels *et al.*, 1986; Caticha, 1994; Andreev & Prudnikov, 1998]. Up to now, the recursion formulae have generally been used for the description of X-ray diffraction by layered crystals with one-dimensional variation of the deformation field $u(z)$ and/or the polarizability $\chi(z)$ along the $z$ axis normal to the crystal surface. The aim of the present communication is to extend Darwin's approach to the case of X-ray dynamical diffraction from distorted crystals which are characterized by a three-dimensional deformation field $\mathbf{u}(\mathbf{r})$ of arbitrary form.

In the spirit of Darwin's approach, we consider first the problem of X-ray scattering by a single distorted atomic plane (Fig. 1). Let the atomic plane be irradiated by a plane monochromatic wave

$$\mathbf{E}(\mathbf{r}'_{xy}) = \mathbf{E}^0 \exp[i(\mathbf{k}_0 \cdot \mathbf{r}'_{xy} - \omega t)],$$

where $k_0 = 2\pi/\lambda$ is the modulus of the wave vector in vacuum. The wave field scattered by a small area d$x$d$y$ in the atomic plane is given by (see *e.g.* Penning, 1966)

$$\mathrm{d}E^s = -(1/r_{xy}) r_e P F(2\vartheta_0) N_{xy} E^0 \times \exp[i(\mathbf{k}_0 \cdot \mathbf{r}'_{xy} + k_0 r_{xy})]\,\mathrm{d}x\mathrm{d}y, \quad (1)$$

where $r_e$ is the classical electron radius, $F(2\vartheta_0)$ is the structure factor, $N_{xy}$ is the number of scattering centers per unit area in the $XY$ plane, $\vartheta_0$ is the incidence angle, $P$ is the polarization factor ($P = 1$ for $\sigma$ polarization and $P = \cos(2\vartheta_0)$ for $\pi$ polarization), and $r'_{xy}$, $r_{xy}$ are the distances depending on the position of d$x$d$y$. In the case of the distorted atomic plane (see Fig. 1), $\mathbf{r}'_{xy} = \boldsymbol{\rho} + \mathbf{u}(\boldsymbol{\rho})$, $\mathbf{r}_{xy} = \mathbf{r}_0 - [\boldsymbol{\rho} + \mathbf{u}(\boldsymbol{\rho})]$, where $\mathbf{r}_0$ is the position vector of the observation point $A$, $\boldsymbol{\rho} = [x, y, 0]$ is the two-dimensional position vector in the $XY$ plane, and $\mathbf{u}(\boldsymbol{\rho})$ is the displacement field. The vector $\mathbf{r}_0$ lies in the $XZ$ plane and makes the angle $\vartheta_0$ with the $X$ axis. Note that in (1) we omit the time factor $\exp(-i\omega t)$. If the conditions $u(\boldsymbol{\rho})/r_0 \ll 1$ and $\rho/r_0 \ll 1$ are simultaneously fulfilled, then the phase $\varphi = (\mathbf{k}_0 \cdot \mathbf{r}'_{xy} + k_0 r_{xy})$ in (1) can be written in the following approximate form:

$$\varphi \approx k_0 r_0 + k_0(x^2 \sin^2\vartheta_0 + y^2)/(2r_0) - \mathbf{Q} \cdot \mathbf{u}(\boldsymbol{\rho}). \quad (2)$$

Here, $\mathbf{Q} = \mathbf{k}_s - \mathbf{k}_0$ and $\mathbf{k}_s = k_0(\mathbf{r}_0/r_0)$ is the wave vector of the scattered wave. For a distorted atomic plane, the phase $\varphi$ differs from that in the case of an ideal (undistorted) atomic plane by the term $\mathbf{Q} \cdot \mathbf{u}(\boldsymbol{\rho})$. In the following, we will consider two-beam Bragg-case diffraction with the reciprocal-lattice vector $\mathbf{g}$. Then we can take $\mathbf{Q} = \mathbf{g}$. Let us represent the phase term $\exp[-i\mathbf{g} \cdot \mathbf{u}(\boldsymbol{\rho})]$ in (1) as the two-dimensional Fourier integral

$$\exp[-i\mathbf{g} \cdot \mathbf{u}(\boldsymbol{\rho})] = \int \widehat{G}(\mathbf{q}) \exp(i\mathbf{q} \cdot \boldsymbol{\rho})\,\mathrm{d}\mathbf{q}, \quad (3)$$

where $\mathbf{q} = [q_x, q_y, 0]$. Taking into account (2) and (3) and performing the integration of (1) with respect to $x$ and $y$ from $-\infty$ to $+\infty$ (the integral is reduced to the Fresnel integral), we obtain the following expression for the scattered wave field $E^s(\mathbf{r}_0)$:

$$E^s(\mathbf{r}_0) = \int \widehat{E}^s(\mathbf{q}) \exp[i\mathbf{k}(\mathbf{q}) \cdot \mathbf{r}_0]\,\mathrm{d}\mathbf{q}, \quad (4)$$

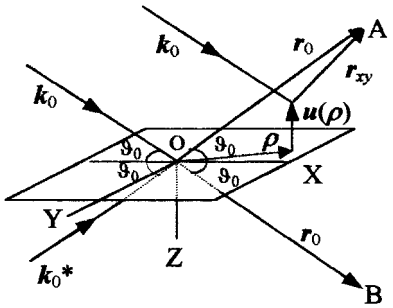


Fig. 1. Sketch of diffraction from a deformed reflecting plane. $A$ and $B$ are observation points. $\mathbf{u}(\boldsymbol{\rho})$ is the displacement of the scattering center at $\boldsymbol{\rho}$. $\mathbf{k}_0^*$ represents the wave vector of the wave incident onto the reverse side of the plane.

where $\widehat{E}^s(\mathbf{q}) = \tau^s \widehat{G}(\mathbf{q})E^0$, $\tau^s = -ir_e F(2\vartheta_0)N_{xy}\lambda P/\sin\vartheta_0$ is the reflection coefficient of an ideal atomic plane (Darwin, 1914) and $\mathbf{k}(\mathbf{q})$ is the wave vector with components $k_x = k_0\cos\vartheta_0 + q_x$, $k_y = q_y$ and $k_z = -(k_0^2 - k_x^2 - k_y^2)^{1/2}$. If the condition $|q_{x,y}|/(k_0\sin\vartheta_0) \ll 1$ holds, then

$$k_z \approx -[k_0\sin\vartheta_0 - (q_x^2/\sin^2\vartheta_0 + q_y^2)/(2k_0\sin\vartheta_0) - q_x\cot\vartheta_0]$$

and the scalar product $\mathbf{k}(\mathbf{q})\cdot\mathbf{r}_0$ in (4) is

$$\mathbf{k}(\mathbf{q})\cdot\mathbf{r}_0 = k_0 r_0[1 - q_x^2/(2k_0^2\sin^2\vartheta_0) - q_y^2/2k_0^2].$$

It follows from (4) that the scattered wave field is the superposition of plane waves. Proceeding in (4) to coordinates $k_x$ and $k_y$, we can write the following expression for the spectral amplitude (angular spectrum) of the wave field:

$$\widehat{E}^s(\mathbf{k}_\parallel) = \tau^s \widehat{G}(\mathbf{k}_\parallel - \mathbf{k}_\parallel^0)E^0, \tag{5}$$

where $\mathbf{k}_\parallel = [k_x, k_y, 0]$ and $\mathbf{k}_\parallel^0$ is the projection of $\mathbf{k}_0$ parallel to the $XY$ plane. Thus, in the case of X-ray scattering by a deformed reflecting plane, in addition to the Bragg wave with the wave vector $\mathbf{k}_s = \mathbf{k}(0)$, there also appear scattered waves with the wave vectors $\mathbf{k}(\mathbf{q})$, where $\mathbf{q} \neq 0$.

Let us generalize (5) to the case where the incident wave is a superposition of plane waves:

$$E(\boldsymbol{\rho}, z) = \int \widehat{E}^0(\boldsymbol{\nu}_\parallel)\exp[i\boldsymbol{\nu}_\parallel\cdot\boldsymbol{\rho} + iK_z(\boldsymbol{\nu}_\parallel)z]\,\mathrm{d}\boldsymbol{\nu}_\parallel. \tag{6}$$

Here $\boldsymbol{\nu}_\parallel = [\nu_x, \nu_y, 0]$ and $K_z(\boldsymbol{\nu}_\parallel) = (k_0^2 - \boldsymbol{\nu}_\parallel^2)^{1/2}$. Note that the wave field (6) is a scalar one. The correctness of this approximation was discussed by Kato (1961). In accordance with the superposition principle, the expression for the spectral amplitude $\widehat{E}^s(\mathbf{k}_\parallel)$ has the form

$$\widehat{E}^s(\mathbf{k}_\parallel) = \tau^s \int \widehat{G}(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel)\widehat{E}^0(\boldsymbol{\nu}_\parallel)\,\mathrm{d}\boldsymbol{\nu}_\parallel. \tag{7}$$

In the vicinity of the Bragg peak, the coefficient $\tau^s$ in (7) can be considered constant, $\tau^s \approx \tau^s|_{\vartheta_0=\vartheta_B}$, where $\vartheta_B$ is the Bragg angle.

Now we calculate the forward-scattered wave field at $B$ (see Fig. 1). Taking $\mathbf{Q} = 0$ in (2), one can readily show that the spectral amplitude $\widehat{E}^t(\mathbf{k}_\parallel)$ of the forward-scattered wave field is

$$\widehat{E}^t(\mathbf{k}_\parallel) = \tau^t \widehat{E}^0(\mathbf{k}_\parallel), \tag{8}$$

where

$$\tau^t = 1 - ir_e F(0)N_{xy}\lambda/\sin\vartheta_B \approx \exp[-ir_e F(0)N_{xy}\lambda/\sin\vartheta_B]$$

is the transmission coefficient of an ideal atomic plane (Darwin, 1914). Thus, (8) describes refraction of plane waves transmitted by the atomic plane.

In a similar way, we can calculate the spectral amplitudes $\widehat{E}^s(\mathbf{k}_\parallel)$ and $\widehat{E}^t(\mathbf{k}_\parallel)$ for an X-ray beam incident onto the reverse side of the reflecting plane (see Fig. 1). In this case, the spectral amplitude $\widehat{E}^t(\mathbf{k}_\parallel)$ coincides with that given by (8), and the spectral amplitude $\widehat{E}^s(\mathbf{k}_\parallel)$ has the form

$$\widehat{E}^s(\mathbf{k}_\parallel) = \overline{\tau}^s \int \widehat{\overline{G}}(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel)\widehat{E}^0(\boldsymbol{\nu}_\parallel)\,\mathrm{d}\boldsymbol{\nu}_\parallel. \tag{9}$$

Here, $\overline{\tau}^s$ is the reflection coefficient for scattering from the reverse side of an ideal atomic plane (James, 1950) and $\widehat{\overline{G}}(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel)$ is the Fourier transform of the phase function $\exp[i\mathbf{g}\cdot\mathbf{u}(\boldsymbol{\rho})]$.

Let us proceed now to the problem of X-ray diffraction in distorted crystals. We restrict ourselves to the symmetric Bragg case. Let the $j$th atomic plane inside the crystal be characterized by the displacement function $\mathbf{u}_j(\boldsymbol{\rho})$, the transmission coefficient $\tau_j^t$, and the reflection coefficients $\tau_j^s$ and $\overline{\tau}_j^s$. The index $j$ varies from 0 to $N$, where $N$ is the total number of atomic planes in the crystal. The plane with $j = 0$ is the entrance surface of the crystal. We represent the wave fields just before the $j$th atomic plane as expansions with respect to plane waves,

$$E_j^{s,t}(\boldsymbol{\rho}) = \int \widehat{E}_j^{s,t}(\mathbf{k}_\parallel)\exp(i\mathbf{k}_\parallel\cdot\boldsymbol{\rho})\,\mathrm{d}\mathbf{k}_\parallel,$$

where $E_j^s(\boldsymbol{\rho})$ and $E_j^t(\boldsymbol{\rho})$ are the diffracted and transmitted wave fields, respectively. Then, taking into account the formulae (7)–(9), we can write the following recursion equations:

$$E_j^t(\mathbf{k}_\parallel) = \exp[i\psi_j(\mathbf{k}_\parallel)]\tau_{j-1}^t E_{j-1}^t(\mathbf{k}_\parallel) + \exp[i\psi_j(\mathbf{k}_\parallel)]\overline{\tau}_{j-1}^s \times \int \widehat{\overline{G}}_{j-1}(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel)\exp[i\psi_j(\boldsymbol{\nu}_\parallel)]E_j^s(\boldsymbol{\nu}_\parallel)\,\mathrm{d}\boldsymbol{\nu}_\parallel, \tag{10a}$$

$$E_j^s(\mathbf{k}_\parallel) = \tau_j^t\exp[i\psi_{j+1}(\mathbf{k}_\parallel)]E_{j+1}^s(\mathbf{k}_\parallel) + \tau_j^s\int \widehat{G}_j(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel)E_j^t(\boldsymbol{\nu}_\parallel)\,\mathrm{d}\boldsymbol{\nu}_\parallel, \tag{10b}$$

where $\psi_j(\mathbf{k}_\parallel) = (k_0^2 - \mathbf{k}_\parallel^2)^{1/2}d_j$ and $d_j$ is the mean distance between the $(j-1)$th and $j$th atomic planes. Equations (10) can be readily interpreted. For example, there are two terms on the right-hand side of (10$a$). The first term describes refraction and phase advance of transmitted waves propagating between the two adjacent planes. The second term describes the dynamical scattering of diffracted waves from the reverse side of the atomic plane and the contribution of the scattered wave field to the transmitted field. We can similarly analyze (10$b$). Equations (10) should be supplemented with the boundary conditions $E_0^t(\mathbf{k}_\parallel) = E_{\mathrm{in}}(\mathbf{k}_\parallel)$, $E_N^s(\mathbf{k}_\parallel) = 0$, where $E_{\mathrm{in}}(\mathbf{k}_\parallel)$ is the spectral amplitude of the incident wave field at the entrance surface. When

$$\widehat{G}_j(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel) = \widehat{\overline{G}}_j(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel) = \delta(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel)$$

and

$$E_{\mathrm{in}}(\boldsymbol{\nu}_\parallel) = \delta(\boldsymbol{\nu}_\parallel - \mathbf{k}_\parallel^0),$$

*i.e.* for a perfect crystal irradiated by a plane wave, equations (10) coincide with the well known Darwin equations for two-beam diffraction (Darwin, 1914; James, 1950). Note that (10)

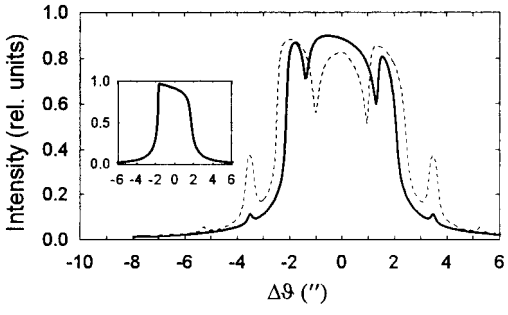


Fig. 2. Calculated rocking curves for a perfect silicon crystal under a SAW excitation (004 reflection, Cu $K\alpha_1$ radiation, $\sigma$ polarization). The SAW wavelength is $\lambda_s = 16\,\mu$m; the SAW amplitude is $a = 0.1$ Å (solid line), $a = 0.2$ Å (dashed line). $\Delta\vartheta$ is the deviation from the Bragg angle. The inset shows the rocking curve for the perfect crystal.

can also be written for the case when together with the deformation field $\mathbf{u}(\mathbf{r})$ there exists a three-dimensional spatial modulation of the structure factor $F(2\vartheta_0)$ in (1).

In the general case of an arbitrary displacement function $\mathbf{u}_j(\boldsymbol{\rho})$, the analytical solution of (10) can be obtained by means of various approximate methods only. For example, it is easy to solve these equations in the kinematical approximation. Neglecting the second term on the right-hand side of (10*a*), we derive the following expression for the spectral amplitude $E_0^s(\mathbf{k}_\parallel)$ of the diffracted field at the entrance surface:

$$E_0^s(\mathbf{k}_\parallel) = \sum_{j=0}^{N} \tau_j^s T_j(\mathbf{k}_\parallel) \int \widehat{G}_j(\mathbf{k}_\parallel - \boldsymbol{\nu}_\parallel) T_j(\boldsymbol{\nu}_\parallel) E_{\text{in}}(\boldsymbol{\nu}_\parallel)\, \mathrm{d}\boldsymbol{\nu}_\parallel, \qquad (11)$$

where

$$T_j(\mathbf{k}_\parallel) = \prod_{p=1}^{j} \{\exp[i\psi_p(\mathbf{k}_\parallel)]\tau_{p-1}^t\}, \quad T_0 \equiv 1.$$

A formula similar to (11) was obtained by Holý *et al.* (1994) on the basis of another approach.

As a numerical example, we show the numerical solution of (10) for dynamical diffraction under a surface acoustic wave (SAW) excitation in a perfect silicon crystal [see *e.g.* Zolotoyabko *et al.* (1993) for details of the experimental technique]. In this case, we can, in the first approximation, take $\mathbf{u}_j(x) = \mathbf{a}\sin(2\pi x/\lambda_s)$, where $\mathbf{a}$ and $\lambda_s$ are the SAW amplitude and wavelength, respectively. Fig. 2 illustrates the appearance of the satellites in the rocking curves.

In conclusion, the Takagi–Taupin equations are a powerful tool for the investigation of dynamical diffraction in distorted crystals. Equations (10) represent another approach to the problem. A comparison of these approaches will be the subject of future work.

The author thanks Professor A. V. Andreev for stimulating discussions.

## References

Andreev, A. V. & Prudnikov, I. R. (1998). *Kristallografiya*, **43**, 111–118.
Bartels, W. J., Hornstra, J. & Lobeek, J. W. (1986). *Acta Cryst.* A**42**, 539–545.
Belyaev, Yu. N. & Kolpakov, A. V. (1983). *Phys. Status Solidi A*, **76**, 641–646.
Borie, B. (1967). *Acta Cryst.* **23**, 210–216.
Caticha, A. (1994). *Phys. Rev. B*, **49**, 33–38.
Darwin, C. G. (1914). *Philos. Mag.* **27**, 675–690.
Holý, V., Wolf, K., Kastner, M., Stanzl, H. & Gebhardt, W. (1994). *J. Appl. Cryst.* **27**, 551–557.
James, R. W. (1950). *The Optical Principles of the Diffraction of X-rays.* London: Bell.
Kato, N. (1961). *Acta Cryst.* **14**, 526–532.
Penning, P. (1966). *Philips Res. Rep. Suppl.* **21**, 1–113.
Takagi, S. (1969). *J. Phys. Soc. Jpn*, **26**, 1239–1253.
Taupin, D. (1964). *Bull. Soc. Fr. Minéral. Cristallogr.* **87**, 469–511.
Vardanyan, D. M., Manoukyan, H. M. & Petrosyan, H. M. (1985). *Acta Cryst.* A**41**, 212–217.
Zolotoyabko, E., Jacobsohn, E., Shechtman, D., Kantor, B. & Salzman, J. (1993). *J. Appl. Phys.* **73**, 8647–8649.