

5.1. DYNAMICAL THEORY OF X-RAY DIFFRACTION

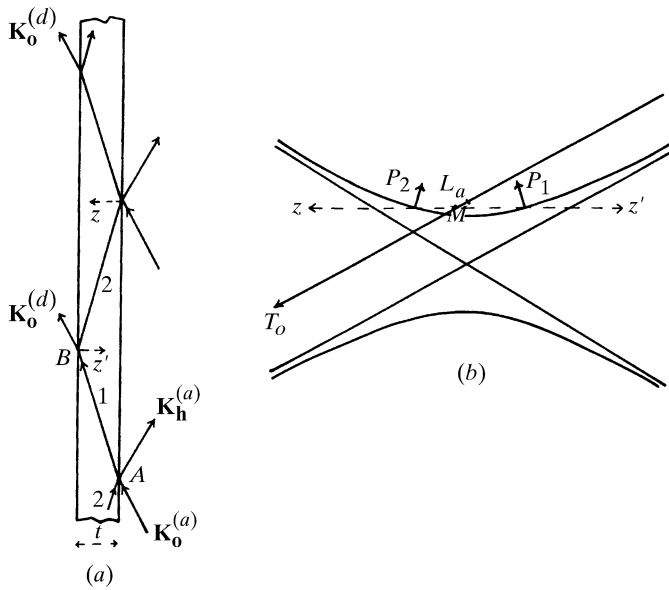


Fig. 5.1.7.4. Bragg case: thin crystals. Two wavefields propagate in the crystal. (a) Direct space; (b) reciprocal space.

points towards the inside. The wavefield propagating along AB will therefore generate at B :

(i) a partially transmitted wave outside the crystal, $D_o^{(d)} \exp(-2\pi i \mathbf{K}_o^{(d)} \cdot \mathbf{r})$;

(ii) a partially reflected wavefield inside the crystal.

The corresponding tiepoints are obtained by applying the usual condition of the continuity of the tangential components of wavevectors (Fig. 5.1.7.4b). If the crystal is a plane-parallel slab, these points are M and P_2 , respectively, and $\mathbf{K}_o^{(d)} = \mathbf{K}_o^{(a)}$.

The boundary conditions are then written:

(i) entrance surface:

$$D_{o1} + D_{o2} = D_o^{(a)}, \quad D_{h1} + D_{h2} = D_h^{(a)};$$

(ii) back surface:

$$D_{o1} \exp(-2\pi i \mathbf{K}_{o1} \cdot \mathbf{r}) + D_{o2} \exp(-2\pi i \mathbf{K}_{o2} \cdot \mathbf{r}) = D_o^{(a)} \exp(-2\pi i \mathbf{K}_o^{(d)} \cdot \mathbf{r}),$$

$$D_{h1} \exp(-2\pi i \mathbf{K}_{h1} \cdot \mathbf{r}) + D_{h2} \exp(-2\pi i \mathbf{K}_{h2} \cdot \mathbf{r}) = 0.$$

Rocking curve. Using (5.1.3.10), it can be shown that the expressions for the intensities reflected at the entrance surface and transmitted at the back surface, I_h and I_o , respectively, are given by different expressions within total reflection and outside it:

(i) $|\eta| < 1$:

$$I_h = |\gamma| \frac{|D_h^{(a)}|^2}{|D_o^{(a)}|^2} = \frac{\cosh^2[\pi(t/\Lambda_B)(1 - \eta^2)^{1/2}] - 1}{\cosh^2[\pi(t/\Lambda_B)(1 - \eta^2)^{1/2}] - \eta^2} \quad (5.1.7.7a)$$

$$I_o = \frac{|D_o^{(d)}|^2}{|D_o^{(a)}|^2} = \frac{1 - \eta^2}{\cosh^2[\pi(t/\Lambda_B)(1 - \eta^2)^{1/2}] - \eta^2},$$

where Λ_B is the value taken by Λ_o [equation (5.1.3.8)] in the Bragg case.

There is no longer a total-reflection domain but the extinction effect still exists, as is shown by the hyperbolic cosine term. The maximum height of the rocking curve decreases as the thickness of the crystal decreases.

(ii) $|\eta| > 1$:

$$I_h = \frac{1 - \cos^2[\pi(t/\Lambda_B)(\eta^2 - 1)^{1/2}]}{\eta^2 - \cos^2[\pi(t/\Lambda_B)(\eta^2 - 1)^{1/2}]}, \quad (5.1.7.7b)$$

$$I_o = \frac{\eta^2 - 1}{\eta^2 - \cos^2[\pi(t/\Lambda_B)(\eta^2 - 1)^{1/2}]}.$$

The cosine terms show that the two wavefields propagating within the crystal interfere, giving rise to *Pendellösung* fringes in the rocking curve. These fringes were observed for the first time by Batterman & Hildebrandt (1967, 1968). The angular positions of the minima of the reflected beam are given by

$$\eta = \mp (K^2 \Lambda_B^2 t^{-2} + 1)^{1/2},$$

where K is an integer.

Integrated intensity. The integrated intensity is

$$I_{hi} = \pi \delta \tanh[\pi t / \Lambda_B], \quad (5.1.7.8)$$

where t is the crystal thickness. When this thickness becomes very large, the integrated intensity tends towards

$$I_{hi} = \pi \delta. \quad (5.1.7.9)$$

This expression differs from (5.1.7.3) by the factor π , which appears here in place of $8/3$. von Laue (1960) pointed out that because of the differences between the two expressions for the reflecting power, (5.1.7.2) and (5.1.7.7b), perfect agreement could not be expected. Since some absorption is always present, expression (5.1.7.3), which includes the factor $8/3$, should be used for very thick crystals. In the presence of absorption, however, expression (5.1.7.8) for the reflected intensity for thin crystals does tend towards that for thick crystals as the crystal thickness increases.

Comparison with geometrical theory. When t/Λ_B is very small (thin crystals or weak reflections), (5.1.7.8) tends towards

$$I_{hi} = R^2 \lambda^2 t |F_h|^2 / (V^2 \gamma_o \sin 2\theta), \quad (5.1.7.10)$$

which is the expression given by geometrical theory. If we call this intensity $I_{hi}(\text{geom.})$, comparison of expressions (5.1.7.8) and (5.1.7.10) shows that the integrated intensity for crystals of intermediate thickness can be written

$$I_{hi} = I_{hi}(\text{geom.}) \frac{\tanh(\pi t / \Lambda_B)}{(\pi t / \Lambda_B)}, \quad (5.1.7.11)$$

which is the expression given by Darwin (1922) for primary extinction.

5.1.7.2.2. Absorbing crystals

Reflected intensity. The intensity of the reflected wave for a thin absorbing crystal is

$$I_h = |\gamma| \left| \frac{D_h^{(a)}}{D_o^{(a)}} \right|^2$$

$$= \left| \frac{F_h}{F_h'} \right| \frac{\cosh 2b - \cos 2a}{L \cosh 2b + (L^2 - 1)^{1/2} \sinh 2b - \cos(2a + 2\Psi')}, \quad (5.1.7.12)$$

where

$$2a = [\pi t / \Lambda_B] \rho \cos(\beta + \omega),$$

$$2b = [\pi t / \Lambda_B] \rho \sin(\beta + \omega).$$

L , ρ and ψ' are defined in (5.1.7.5), β is defined in (5.1.3.7) and ω is the phase angle of $(\eta^2 - 1)^{1/2}$.

Comparison with geometrical theory. When t/Λ_B decreases towards zero, expression (5.1.7.12) tends towards $[\sin(\pi t \eta / \Lambda_B) / \eta]^2$; using (5.1.3.5) and (5.1.3.8), it can be shown that expression (5.1.7.12) can be written, in the non-absorbing symmetric case, as

$$I_h = \frac{R^2 \lambda^2 C^2 |F_h|^2 t^2}{V^2 \sin^2 \theta} \left\{ \frac{\sin[2\pi k \cos(\theta) t \Delta\theta]}{[2\pi k \cos(\theta) t \Delta\theta]} \right\}^2, \quad (5.1.7.13)$$

where d is the lattice spacing and $\Delta\theta$ is the difference between the angle of incidence and the middle of the reflection domain. This expression is the classical expression given by geometrical theory [see, for instance, James (1950)].

5.1.8. Real waves

5.1.8.1. Introduction

The preceding sections have dealt with the diffraction of a plane wave by a semi-infinite perfect crystal. This situation is actually never encountered in practice, although with various devices, in particular using synchrotron radiation, it is possible to produce highly collimated monochromated waves which behave like pseudo plane waves. The wave from an X-ray tube is best represented by a spherical wave. The first experimental proof of this fact is due to Kato & Lang (1959) in the transmission case. Kato extended the dynamical theory to spherical waves for non-absorbing (1961*a,b*) and absorbing crystals (1968*a,b*). He expanded the incident spherical wave into plane waves by a Fourier transform, applied plane-wave dynamical theory to each of these components and took the Fourier transform of the result again in order to obtain the final solution. Another method for treating the problem was used by Takagi (1962, 1969), who solved the propagation equation in a medium where the lateral extension of the incident wave is limited and where the wave amplitudes depend on the lateral coordinates. He showed that in this case the set of fundamental linear equations (5.1.2.20) should be replaced by a set of partial differential equations. This treatment can be applied equally well to a perfect or to an imperfect crystal. In the case of a perfect crystal, Takagi showed that these equations have an analytical solution that is identical with Kato's result. Uragami (1969, 1970) observed the spherical wave in the Bragg (reflection) case, interpreting the observed intensity distribution using Takagi's theory. Saka *et al.* (1973) subsequently extended Kato's theory to the Bragg case.

Without using any mathematical treatment, it is possible to make some elementary remarks by considering the fact that the divergence of the incident beam falling on the crystal from the source is much larger than the angular width of the reflection domain. Fig. 5.1.8.1(*a*) shows a spatially collimated beam falling on a crystal in the transmission case and Fig. 5.1.8.1(*b*) represents the corresponding situation in reciprocal space. Since the divergence of the incident beam is larger than the angular width of the dispersion surface, the plane waves of its Fourier expansion will excite every point of both branches of the dispersion surface. The propagation directions of the corresponding wavefields will cover the angular range between those of the incident and reflected beams (Fig. 5.1.8.1*a*) and fill what is called the *Borrmann triangle*. The intensity distribution within this triangle has interesting properties, as described in the next two sections.

5.1.8.2. Borrmann triangle

The first property of the Borrmann triangle is that the angular density of the wavefield paths is inversely proportional to the

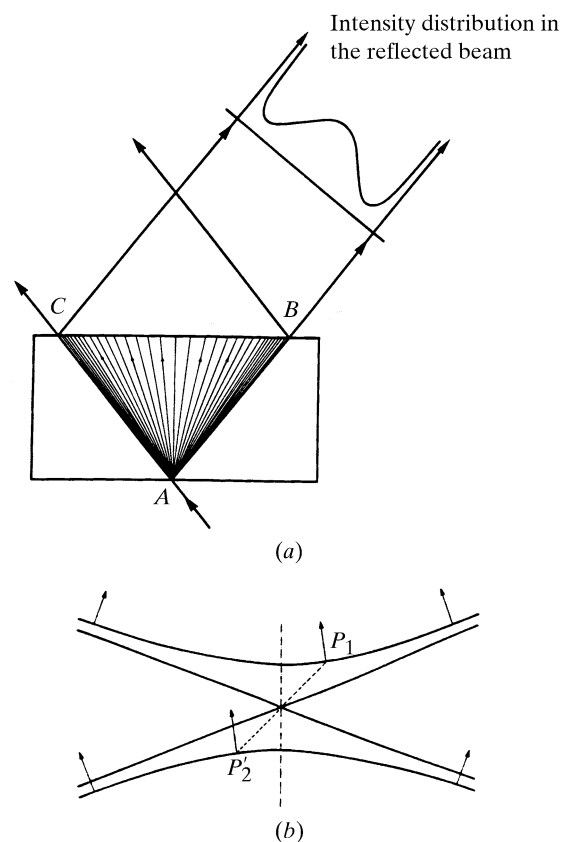


Fig. 5.1.8.1. Borrmann triangle. When the incident beam is divergent, the whole dispersion surface is excited and the wavefields excited inside the crystal propagate within a triangle filling all the space between the incident direction, AC , and the reflected direction, AB . Along any direction Ap within this triangle two wavefields propagate, having as tie points two conjugate points, P and P' , at the extremities of a diameter of the dispersion surface. (*a*) Direct space; (*b*) reciprocal space.

curvature of the dispersion surface around their tie points. Let us consider an incident wavepacket of angular width $\delta(\Delta\theta)$. It will generate a packet of wavefields propagating within the Borrmann triangle. The angular width $\Delta\alpha$ (Fig. 5.1.8.2) between the paths of the corresponding wavefields is related to the radius of curvature \mathcal{R} of the dispersion surface by

$$\mathcal{A} = \Delta\alpha / \delta(\Delta\theta) = k \cos \theta (\mathcal{R} \cos \alpha), \quad (5.1.8.1)$$

where α is the angle between the wavefield path and the lattice planes [equation (5.1.2.26)] and \mathcal{A} is called the amplification ratio. In the middle of the reflecting domain, the radius of curvature of the dispersion surface is very much shorter than its value, k , far from it (about 10^4 times shorter) and the amplification ratio is therefore very large. As a consequence, the energy of a wavepacket of width $\delta(\Delta\theta)$ in reciprocal space is spread in direct space over an angle $\Delta\alpha$ given by (5.1.8.1). The intensity distribution on the exit surface BC (Fig. 5.1.8.1*a*) is therefore proportional to I_h / \mathcal{A} . It is represented in Fig. 5.1.8.3 for several values of the absorption coefficient:

(i) Small values of $\mu_0 t$ (less than 2 or 3) (Fig. 5.1.8.3*a*). The intensity distribution presents a wide minimum in the centre where the density of wavefields is small and increases very sharply at the edges where the density of wavefields is large, although it is the reverse for the reflecting power I_{hj} . This effect, called the *margin effect*, was predicted qualitatively by Borrmann (1959) and von Laue (1960), demonstrated experimentally by Kato & Lang (1959), and calculated by Kato (1960).