

5. DYNAMICAL THEORY AND ITS APPLICATIONS

$$\mathbf{K}_h = \mathbf{K}_o - \mathbf{h}. \quad (5.1.2.16) \quad \text{part,}$$

Expression (5.1.2.15) shows that the solution of the propagation equation can be interpreted as an infinite sum of plane waves with amplitudes \mathbf{D}_h and wavevectors \mathbf{K}_h . This sum is a *wavefield*, or *Ewald wave*. The same expression is used to describe the propagation of any wave in a periodic medium, such as phonons or electrons in a solid. Expression (5.1.2.14) was later called a *Bloch wave* by solid-state physicists.

The wavevectors in a wavefield are deduced from one another by translations of the reciprocal lattice [expression (5.1.2.16)]. They can be represented geometrically as shown in Fig. 5.1.2.1(b). The wavevectors $\mathbf{K}_o = \mathbf{OP}$; $\mathbf{K}_h = \mathbf{HP}$ are drawn *away* from reciprocal-lattice points. Their common extremity, P , called the *tie point* by Ewald, characterizes the wavefield.

In an absorbing crystal, wavevectors have an imaginary part,

$$\mathbf{K}_o = \mathbf{K}_{or} + i\mathbf{K}_{oi}; \quad \mathbf{K}_h = \mathbf{K}_{hr} + i\mathbf{K}_{hi},$$

and (5.1.2.16) shows that all wavevectors have the same imaginary part,

$$\mathbf{K}_{oi} = \mathbf{K}_{hi}, \quad (5.1.2.17)$$

and therefore undergo the same absorption. This is one of the most important properties of wavefields.

5.1.2.3. Boundary conditions at the entrance surface

The choice of the ‘ \mathbf{o} ’ component of expansion (5.1.2.15) is arbitrary in an infinite medium. In a semi-infinite medium where the waves are created at the interface with a vacuum or air by an incident plane wave with wavevector $\mathbf{K}_o^{(a)}$ (using von Laue’s notation), the choice of \mathbf{K}_o is determined by the boundary conditions.

This condition for wavevectors at an interface demands that their tangential components should be continuous across the boundary, in agreement with Descartes–Snell’s law. This condition is satisfied when the difference between the wavevectors on each side of the interface is parallel to the normal to the interface. This is shown geometrically in Fig. 5.1.2.2 and formally in (5.1.2.18):

$$\mathbf{K}_o - \mathbf{K}_o^{(a)} = \mathbf{OP} - \mathbf{OM} = \overline{MP} \cdot \mathbf{n}, \quad (5.1.2.18)$$

where \mathbf{n} is a unit vector normal to the crystal surface, oriented towards the inside of the crystal.

There is no absorption in a vacuum and the incident wavevector $\mathbf{K}_o^{(a)}$ is real. Equation (5.1.2.18) shows that it is the component normal to the interface of wavevector \mathbf{K}_o which has an imaginary

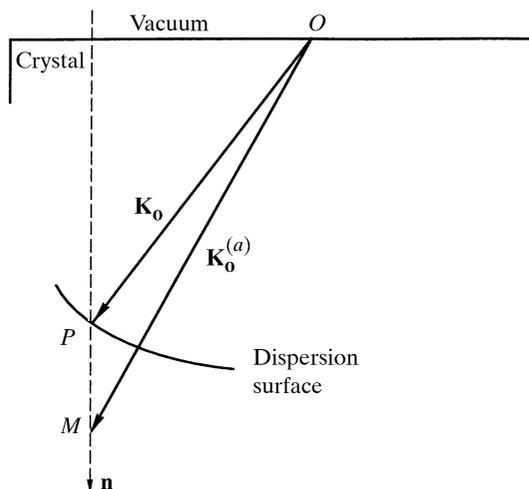


Fig. 5.1.2.2. Boundary condition for wavevectors at the entrance surface of the crystal.

$$\mathbf{K}_{oi} = \mathcal{I}(\overline{MP}) \cdot \mathbf{n} = -\mu\mathbf{n}/(4\pi\gamma_o), \quad (5.1.2.19)$$

where $\mathcal{I}(f)$ is the imaginary part of f , $\gamma_o = \cos(\mathbf{n} \cdot \mathbf{s}_o)$ and \mathbf{s}_o is a unit vector in the incident direction. When there is more than one wave in the wavefield, the effective absorption coefficient μ can differ significantly from the normal value, μ_o , given by (5.1.2.13) – see Section 5.1.5.

5.1.2.4. Fundamental equations of dynamical theory

In order to obtain the solution of dynamical theory, one inserts expansions (5.1.2.15) and (5.1.2.4) into the propagation equation (5.1.2.2). This leads to an equation with an infinite sum of terms. It is shown to be equivalent to an infinite system of linear equations which are the *fundamental equations* of dynamical theory. Only those terms in (5.1.2.15) whose wavevector magnitudes K_h are very close to the vacuum value, k , have a non-negligible amplitude. These wavevectors are associated with reciprocal-lattice points that lie very close to the Ewald sphere. Far from any Bragg reflection, their number is equal to 1 and a single plane wave propagates through the medium. In general, for X-rays, there are only two reciprocal-lattice points on the Ewald sphere. This is the so-called *two-beam* case to which this treatment is limited. There are, however, many instances where several reciprocal-lattice points lie simultaneously on the Ewald sphere. This corresponds to the *many-beam* case which has interesting applications for the determination of phases of reflections [see, for instance, Chang (1987) and Hümmer & Weckert (1995)]. On the other hand, for electrons, there are in general many reciprocal-lattice points close to the Ewald sphere and many wavefields are excited simultaneously (see Chapter 5.2).

In the two-beam case, for reflections that are not highly asymmetric and for Bragg angles that are not close to $\pi/2$, the fundamental equations of dynamical theory reduce to

$$\begin{aligned} 2X_o D_o - kC\chi_h D_h &= 0 \\ -kC\chi_h D_o + 2X_h D_h &= 0, \end{aligned} \quad (5.1.2.20)$$

where $C = 1$ if \mathbf{D}_h is normal to the $\mathbf{K}_o, \mathbf{K}_h$ plane and $C = \cos 2\theta$ if \mathbf{D}_h lies in the plane; this is due to the fact that the amplitude with which electromagnetic radiation is scattered is proportional to the sine of the angle between the direction of the electric vector of the incident radiation and the direction of scattering (see, for instance, *ITC*, Section 6.2.2). The polarization of an electromagnetic wave is classically related to the orientation of the electric vector; in dynamical theory it is that of the electric displacement which is considered (see Section A5.1.1.3 of the Appendix).

The system (5.1.2.20) is therefore a system of four equations which admits four solutions, two for each direction of polarization. In the *non-absorbing* case, to a very good approximation,

$$\begin{aligned} X_o &= K_o - nk, \\ X_h &= K_h - nk. \end{aligned} \quad (5.1.2.21)$$

In the case of an *absorbing crystal*, X_o and K_h are complex. Equation (5.1.5.2) gives the full expression for X_o .

5.1.2.5. Dispersion surface

The fundamental equations (5.1.2.20) of dynamical theory are a set of linear homogeneous equations whose unknowns are the amplitudes of the various waves which make up a wavefield. For the solution to be non-trivial, the determinant of the set must be set equal to zero. This provides a secular equation relating the magnitudes of the wavevectors of a given wavefield. This equation is that of the locus of the tie points of all the wavefields that may

5.1. DYNAMICAL THEORY OF X-RAY DIFFRACTION

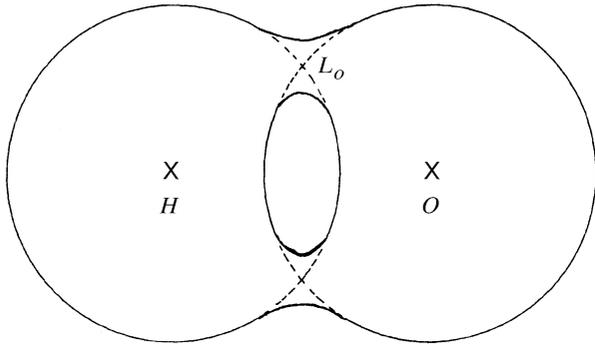


Fig. 5.1.2.3. Intersection of the dispersion surface with the plane of incidence. The dispersion surface is a connecting surface between the two spheres centred at reciprocal-lattice points O and H and with radius nk . L_o is the Lorentz point.

propagate in the crystal with a given frequency. This locus is called the *dispersion surface*. It is a constant-energy surface and is the equivalent of the index surface in optics. It is the X-ray analogue of the constant-energy surfaces known as Fermi surfaces in the electron band theory of solids.

In the two-beam case, the dispersion surface is a surface of revolution around the diffraction vector \mathbf{OH} . It is made from two spheres and a connecting surface between them. The two spheres are centred at O and H and have the same radius, nk . Fig. 5.1.2.3 shows the intersection of the dispersion surface with a plane passing through \mathbf{OH} . When the tie point lies on one of the two spheres, far from their intersection, only one wavefield propagates inside the crystal. When it lies on the connecting surface, two waves are excited simultaneously. The equation of this surface is obtained by equating to zero the determinant of system (5.1.2.20):

$$X_o X_h = k^2 C^2 \chi_h \chi_{\bar{h}} / 4. \quad (5.1.2.22)$$

Equations (5.1.2.21) show that, in the zero-absorption case, X_o and X_h are to be interpreted as the distances of the tie point P from the spheres centred at O and H , respectively. From (5.1.2.20) it can be seen that they are of the order of the vacuum wavenumber times the Fourier coefficient of the dielectric susceptibility, that is five or six orders of magnitude smaller than k . The two spheres can therefore be replaced by their tangential planes. Equation (5.1.2.22) shows that the product of the distances of the tie point from these planes is constant. The intersection of the dispersion surface with the plane passing through \mathbf{OH} is therefore a hyperbola (Fig. 5.1.2.4) whose diameter [using (5.1.2.5) and (5.1.2.22)] is

$$\overline{A_{o2}A_{o1}} = |C|R\lambda(F_h F_{\bar{h}})^{1/2} / (\pi V \cos \theta). \quad (5.1.2.23)$$

It can be noted that the larger the diameter of the dispersion surface, the larger the structure factor, that is, the stronger the interaction of the waves with the matter. When the polarization is parallel to the plane of incidence ($C = \cos 2\theta$), the interaction is weaker.

The asymptotes T_o and T_h to the hyperbola are tangents to the circles centred at O and H , respectively. Their intersection, L_o , is called the *Lorentz point* (Fig. 5.1.2.4).

A wavefield propagating in the crystal is characterized by a tie point P on the dispersion surface and two waves with wavevectors $\mathbf{K}_o = \mathbf{OP}$ and $\mathbf{K}_h = \mathbf{HP}$, respectively. The ratio, ξ , of their amplitudes D_h and D_o is given by means of (5.1.2.20):

$$\xi = \frac{D_h}{D_o} = \frac{2X_o}{kC\chi_{\bar{h}}} = \frac{-2\pi V X_o}{R\lambda C F_{\bar{h}}}. \quad (5.1.2.24)$$

The hyperbola has two branches, 1 and 2, for each direction of polarization, that is, for $C = 1$ or $\cos 2\theta$ (Fig. 5.1.2.5). Branch 2 is

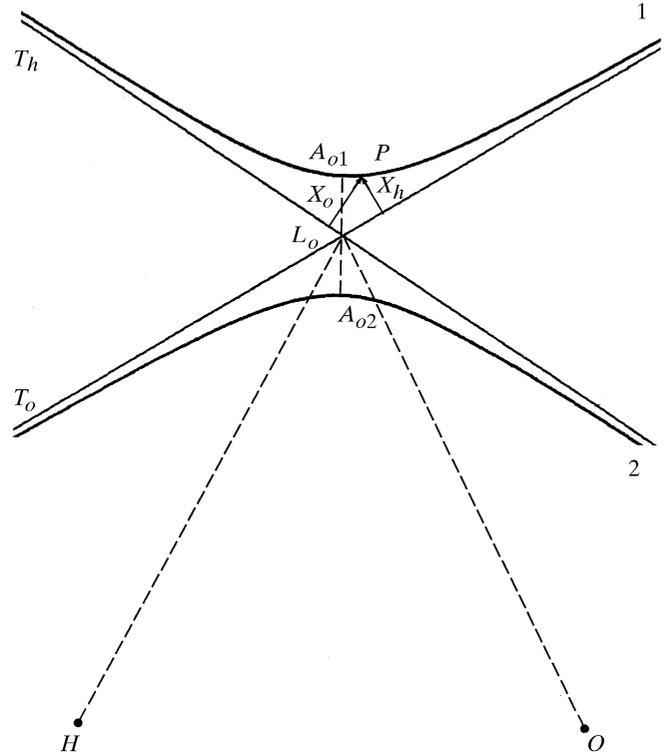


Fig. 5.1.2.4. Intersection of the dispersion surface with the plane of incidence shown in greater detail. The Lorentz point L_o is far away from the nodes O and H of the reciprocal lattice: $OL_o = HL_o = 1/\lambda$ is about 10^5 to 10^6 times larger than the diameter $A_{o1}A_{o2}$ of the dispersion surface.

the one situated on the same side of the asymptotes as the reciprocal-lattice points O and H . Given the orientation of the wavevectors, which has been chosen away from the reciprocal-lattice points (Fig. 5.1.2.1b), the coordinates of the tie point, X_o and X_h , are positive for branch 1 and negative for branch 2. The phase of ξ is therefore equal to $\pi + \varphi_h$ and to φ_h for the two branches, respectively, where φ_h is the phase of the structure factor [equation (5.1.2.6)]. This difference of π between the two branches has important consequences for the properties of the wavefields.

As mentioned above, owing to absorption, wavevectors are actually complex and so is the dispersion surface.

5.1.2.6. Propagation direction

The energy of all the waves in a given wavefield propagates in a common direction, which is obtained by calculating either the group velocity or the Poynting vector [see Section A5.1.1.4, equation (A5.1.1.8) of the Appendix]. It can be shown that, averaged over time and the unit cell, the Poynting vector of a wavefield is

$$\mathbf{S} = (c/\varepsilon_0) \exp(4\pi\mathbf{K}_{oi} \cdot \mathbf{r}) \left[|D_o|^2 \mathbf{s}_o + |D_h|^2 \mathbf{s}_h \right], \quad (5.1.2.25)$$

where \mathbf{s}_o and \mathbf{s}_h are unit vectors in the \mathbf{K}_o and \mathbf{K}_h directions, respectively, c is the velocity of light and ε_0 is the dielectric permittivity of a vacuum. This result was first shown by von Laue (1952) in the two-beam case and was generalized to the n -beam case by Kato (1958).

From (5.1.2.25) and equation (5.1.2.22) of the dispersion surface, it can be shown that the propagation direction of the wavefield lies along the normal to the dispersion surface at the tie point (Fig. 5.1.2.5). This result is also obtained by considering the group velocity of the wavefield (Ewald, 1958; Wagner, 1959). The angle α