3. It is a multiple-solution method.
4. The calculations can easily be done on a desk calculator, and are also well adaptable to computer programming.

The method proved successful for a projection of the structure of L-asparagine monohydrate (De Vries, 1963), for projections of the structures of three modifications of mesotartaric acid (Bootsma & Schoone, 1965) and for a projection of the structure of tartronic acid (Kroon, Kanters & Van Eyck, 1965).

APPENDIX

The value 23·1 obtained in § 3 for the P^*_+/P^*_- ratio for S_1 is actually somewhat too high, since three of the triple products involved are not independent, because S_3 and S_8 are each present in two different triple products. The correct value of P^*_+/P^*_-, which can be obtained by calculating the probabilities for the various sign combinations in a way analogous to that followed in Table 1, is 21·7.

For each of the signs S_3 and S_8 separately, its influence may easily be calculated as is shown here for S_3. Instead of

$$S_2S_3S_4 = S_3S_8S_9$$

we write

$$S_3 \times (S_2S_4 = S_8S_9) .$$

The term between brackets may now be considered as one single sign, the P_+/P_- ratio of which is equal to the product of the P_+/P_- ratios of S_2S_4 and S_8S_9. From here on the calculation of P^*_+/P^*_- for S_1 proceeds as in § 3.

Since S_3 and S_8 occur together in one triple product, this trick can not be used for the calculation of the influence of the two signs together. However, in most cases the error made when the procedure of § 3 is followed (which ignores the effect of the interrelation between the correlation equations) is rather small.

It is a pleasure to thank Prof. J. M. Bijvoet and Prof. A. F. Peerdeman for their continuous interest and valuable criticism.

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On the Proper Modes of Propagation of X-rays*

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The modes of propagation of an X-ray wave field for single diffraction are evaluated at the Bragg angle as an illustration of a method which can be applied to simultaneous diffraction.

Introduction

The methods of the dynamical theory of X-ray diffraction are presented extensively in established texts (James, 1954; Zachariasen, 1945) and recently have been subjected to some reviews (James, 1963). The general theoretical treatment deals with cases when any number of reciprocal lattice points enter the Ewald sphere. However, applications of the general results of the dynamical theory to situations observed in practice are almost exclusively limited to two fields, *i.e.* to the case when only two reciprocal lattice points (one of which is the origin) are on the sphere of reflection. Such theory has been worked out in detail. The starting and most important part is the evaluation of the equations of the dispersion sheets.

When considering a larger number of fields, say three or four, the usual procedure is unwieldy. It is rather difficult to obtain the dispersion sheets. Considerable information can, however, be obtained when one works exclusively under conditions when Bragg's law is fulfilled exactly. These conditions correspond to the diameter points of the dispersion sheets.

In addition, it seems to us that in the case of a larger number of fields it is necessary to evaluate the normal modes of vibration of the electric field vector of the X-ray wave fields. Considerable insight

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is then gained into the whole field of multiple X-ray diffraction theory (Zajac, Saccocio & Bindell, 1964).

We will illustrate the method of evaluating the modes of vibration in the two field case. The purpose of this paper is not the determination of the modes in the two field case, since this is really known, but rather our aim is the illustration of a method which can be used in cases of a larger number of fields.

Theory

Let $\mathbf{k}_0$ and $\mathbf{k}_H$ be the propagation vectors of an X-ray wave field inside a crystal pointing towards the two points 0 and H of the reciprocal lattice; $\mathbf{k}_0$ and $\mathbf{k}_H$ indicate the directions of incidence and diffraction respectively (Fig. 1). Because the waves are entirely transverse, the vibrations of the electric vector will be confined to planes perpendicular to the propagation vector. For reference directions in these planes we will choose the directions specified by pairs of orthogonal unit vectors $\boldsymbol{\sigma}_0$, $\boldsymbol{\pi}_0$ and $\boldsymbol{\sigma}_H$, $\boldsymbol{\pi}_H$ respectively as shown in Fig. 1, where the angles α and β are with reference to the plane of incidence. As can be seen from the figure the choice is quite arbitrary.

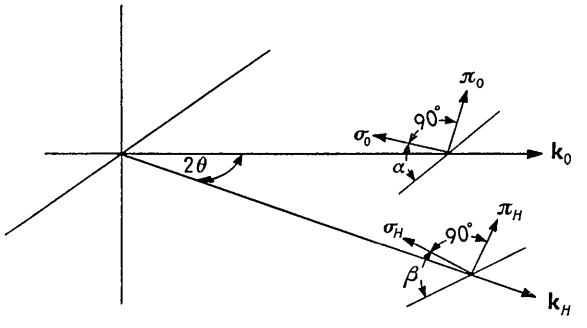

Fig. 1. Wave vectors $\mathbf{k}_0$ and $\mathbf{k}_H$ with directions of polarization σ and π chosen arbitrarily.

Following Kato's (1958) general formulation and writing the component equations we have

$$\begin{aligned} -x_0 D_0^{\sigma} &= \varphi_{\bar{H}} D_H^{\sigma H}\gamma_1 + \varphi_{\bar{H}} D_H^{\pi H}\gamma_3 \\ -x_0 D_0^{\pi} &= \varphi_{\bar{H}} D_H^{\sigma H}\gamma_4 + \varphi_{\bar{H}} D_H^{\pi H}\gamma_2 \\ -x_H D_H^{\sigma H} &= \varphi_H D_0^{\sigma}\gamma_1 + \varphi_H H_0^{\pi}\gamma_4 \\ -x_H D_H^{\pi H} &= \varphi_H D_0^{\sigma}\gamma_3 + \varphi_H D_0^{\pi}\gamma_2 \end{aligned} \tag{1}$$

where

$$k^2 x_n = k_n^2 - k^2(1-\varphi_0)\,, \tag{2}$$

and

$$\begin{aligned} \gamma_1 &= \cos 2\theta \cos\alpha \cos\beta + \sin\alpha \sin\beta = \boldsymbol{\sigma}_0 \,.\, \boldsymbol{\sigma}_H \\ \gamma_2 &= \cos 2\theta \sin\alpha \sin\beta + \cos\alpha \cos\beta = \boldsymbol{\pi}_0 \,.\, \boldsymbol{\pi}_H \\ \gamma_3 &= -\cos 2\theta \cos\alpha \sin\beta + \sin\alpha \cos\beta = \boldsymbol{\sigma}_0 \,.\, \boldsymbol{\pi}_H \\ \gamma_4 &= -\cos 2\theta \sin\alpha \cos\beta + \cos\alpha \sin\beta = \boldsymbol{\pi}_0 \,.\, \boldsymbol{\sigma}_H \,. \end{aligned} \tag{3}$$

We will determine the modes of propagation only for the points on the dispersion sheets which satisfy Bragg's law exactly. Following the usual procedure, from the equations (1) we formulate the secular equation in which, because we work with the diameter points only, we set

$$x_0 = x_H = x\,. \tag{4}$$

Thus

$$\begin{vmatrix} x & 0 & \gamma_1\varphi_{\bar{H}} & \gamma_3\varphi_{\bar{H}} \\ 0 & x & \gamma_4\varphi_{\bar{H}} & \gamma_2\varphi_{\bar{H}} \\ \gamma_1\varphi_H & \gamma_4\varphi_H & x & 0 \\ \gamma_3\varphi_H & \gamma_2\varphi_H & 0 & x \end{vmatrix} = 0 \tag{5}$$

for which, in terms of the γ's, the four roots may be shown to be

$$\begin{aligned} x^2 = \tfrac{1}{2}\varphi_H\varphi_{\bar{H}}\{&(\gamma_1^2+\gamma_2^2+\gamma_3^2+\gamma_4^2) \\ &\pm[(\gamma_1^2+\gamma_2^2+\gamma_3^2+\gamma_4^2)^2 - 4(\gamma_1\gamma_2-\gamma_3\gamma_4)^2]^{\frac{1}{2}}\}. \end{aligned} \tag{6}$$

From the definitions (3), these roots, or eigenvalues, reduce to

$$x_{1,2} = \pm(\varphi_H\varphi_{\bar{H}})^{\frac{1}{2}}$$

and

$$x_{3,4} = \pm\cos 2\theta\,(\varphi_H\varphi_{\bar{H}})^{\frac{1}{2}}. \tag{7}$$

To find the corresponding modes of propagation we consider the ratios D_0^{π}/D_0^{σ} and $D_H^{\pi H}/D_H^{\sigma H}$ for each value in (7). Thus

$$D_0^{\pi}/D_0^{\sigma} = -\frac{\begin{vmatrix} x & \gamma_1\varphi_{\bar{H}} & \gamma_3\varphi_{\bar{H}} \\ \gamma_1\varphi_H & x & 0 \\ \gamma_3\varphi_H & 0 & x \end{vmatrix}}{\begin{vmatrix} 0 & \gamma_1\varphi_{\bar{H}} & \gamma_3\varphi_{\bar{H}} \\ \gamma_4\varphi_H & x & 0 \\ \gamma_2\varphi_H & 0 & x \end{vmatrix}}$$

or

$$D_0^{\pi}/D_0^{\sigma} = \frac{x^2-(\gamma_1^2+\gamma_3^2)\gamma_H\varphi_{\bar{H}}}{\varphi_H\varphi_{\bar{H}}(\gamma_1\gamma_4+\gamma_2\gamma_3)}. \tag{8}$$

Using (3) this equation becomes

$$D_0^{\pi}/D_0^{\sigma} = \frac{x^2-(\sin^2\alpha+\cos^2\alpha\cos^2 2\theta)\varphi_H\varphi_{\bar{H}}}{\varphi_H\varphi_{\bar{H}}\sin\alpha\cos\alpha(1-\cos^2 2\theta)}. \tag{9}$$

For eigenvalues

$$x_{1,2} = \pm(\varphi_H\varphi_{\bar{H}})^{\frac{1}{2}},$$

expression (9) becomes

$$D_0^{\pi}/D_0^{\sigma} = \cot\alpha \tag{10}$$

and for

$$x_{3,4} = \pm\cos 2\theta\,(\varphi_H\varphi_{\bar{H}})^{\frac{1}{2}}$$

we obtain

$$D_0^{\pi}/D_0^{\sigma} = -\tan\alpha\,. \tag{11}$$

The ratios $D_H^{\pi H}/D_H^{\sigma H}$ can be determined in the same manner.

Conclusion and discussion

The meaning of the results will be made clear with the help of Fig. 2(*a*) and (*b*). For the ratio given by equation (10), we see from Fig. 2(*a*) that the vector $\mathbf{D}_0$, as described by its components, oscillates perpendicular to the plane of incidence. When equation (11) is interpreted we obtain the result shown in Fig. 2(*b*), namely that $\mathbf{D}_0$ vibrates in the plane of incidence.

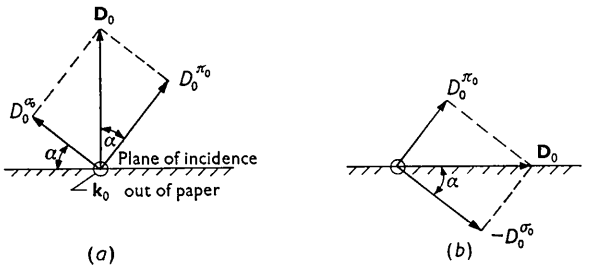

Fig. 2. Directions of vibration of $\mathbf{D}_0$.

The complete set of the normal modes of propagation consists of four modes all of which are shown in Fig. 3(*a*), (*b*), (*c*) and (*d*).

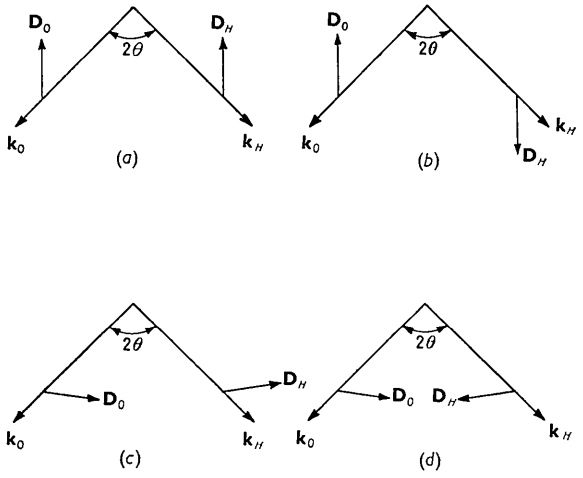

Fig. 3. Proper modes of propagation.

We have presented a method of evaluating the normal modes of propagation in the two-field case for the situation when Bragg's law is fulfilled exactly. In this case four modes exist. For q fields we would expect $2q$ modes of propagation. The same method can be used for three, four and a larger number of fields.

APPENDIX*

In the two field case it can readily be shown for every point of the dispersion sheet that the states of vibration of the eigenvectors are confined to the plane of incidence and perpendicular to it. In this general case equation (5) takes the form

$$\begin{vmatrix} x_0 & 0 & \gamma_1\varphi_{\bar{H}} & \gamma_3\varphi_{\bar{H}} \\ 0 & x_0 & \gamma_4\varphi_{\bar{H}} & \gamma_2\varphi_{\bar{H}} \\ \gamma_1\varphi_H & \gamma_4\varphi_H & x_H & 0 \\ \gamma_3\varphi_H & \gamma_2\varphi_H & 0 & x_H \end{vmatrix} = 0 . \tag{12}$$

The solution of this equation is

$$x_0x_H = \tfrac{1}{2}\varphi_H\varphi_{\bar{H}}\{\gamma_1^2+\gamma_2^2+\gamma_3^2+\gamma_4^2 \pm \sqrt{((\gamma_1^2+\gamma_2^2+\gamma_3^2+\gamma_4^2)^2-4(\gamma_1\gamma_2-\gamma_3\gamma_4)^2)}\} \tag{13}$$

which gives

$$x_0x_H = \varphi_H\varphi_{\bar{H}} \tag{14}$$

and

$$x_0x_H = \cos^2 2\theta\varphi_H\varphi_{\bar{H}} . \tag{15}$$

The determinant of equation (12) contains the general polarization conditions and there is no restriction on the x's. The two equations (14) and (15) indicate that the two allowed wave vectors (related to x_0 and x_H) lie on two separate hyperbolic sheets irrespective of the choice of the polarization direction.

The determination of the directions in which the electric vectors vibrate for any point of the dispersion surface readily follows. We obtain

$$D_0^\pi/D_0^\sigma = \frac{-\begin{vmatrix} x_0 & \gamma_1\varphi_{\bar{H}} & \gamma_3\varphi_{\bar{H}} \\ \gamma_1\varphi_{\bar{H}} & x_{\bar{H}} & 0 \\ \gamma_3\varphi_H & 0 & x_H \end{vmatrix}}{\begin{vmatrix} 0 & \gamma_1\varphi_{\bar{H}} & \gamma_3\varphi_{\bar{H}} \\ \gamma_4\varphi_H & x_H & 0 \\ \gamma_2\varphi_H & 0 & x_H \end{vmatrix}}$$

$$= \frac{x_0x_H-\gamma_3\varphi_{\bar{H}}\gamma_3\varphi_H-\gamma_1\varphi_{\bar{H}}\gamma_1\varphi_H}{\gamma_3\varphi_{\bar{H}}\gamma_2\varphi_H+\gamma_1\varphi_{\bar{H}}\gamma_4\varphi_H} . \tag{16}$$

For one branch $x_0x_H=\varphi_H\varphi_{\bar{H}}$, and

$$D_0^\pi/D_0^\sigma = \cot\alpha . \tag{17}$$

i.e. the electric vector is perpendicular to the plane of incidence (the usual σ state). For the other branch $x_0x_H=\cos^2 2\theta\varphi_H\varphi_{\bar{H}}$, and

$$D_0^\pi/D_0^\sigma = -\tan\alpha , \tag{18}$$

which means that the electric displacement vector oscillates in the plane of incidence.

The foregoing shows that the general solution to the linear equations describing the fields inside the crystal (two-beam case) separates into two pairs of dispersion surfaces, one for each of the usual polarization states; and this is independent of the initial choice of polarization directions. Hence, this separation is valid for all incident conditions, not only at the exact Bragg angle.

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* This appendix has been written almost in the form suggested by Boris W. Batterman. The authors acknowledge this fact and thank him for other critical remarks concerning the paper.

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