JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 17, SUPPLEMENT B-II, 1962
PROCEEDINGS OF INTERNATIONAL CONFERENCE ON MAGNETISM AND CRYSTALLOGRAPHY, 1961, VOL. II

Recent Developments of the Matrical and Semi-Reciprocal Formulation in the Field of Dynamical Theory

M. TOURNARIE
Centre d'Etudes Nucléaires de Saclay, France

Using a new formulation, we can compare Fujiwara's and Fujimoto's theories with the author's theory. The initial behaviours (for thin specimens with increasing thickness) are different. We cannot yet know if these discrepancies are reduced for thicker specimens. Numerical results of the author's theory for many beams are given for some models.

1. Propagation equation

We start from the Schrödinger equation. Here we understand by "potential" a quantity which has the same dimension as energy, which is a function of space coordinates and whose modulus and argument are defined by the amplitude and the phase of the wave elastically scattered by an infinitely thin sheet of the crystal. The potential defined in this way should be complex if inelastic scattering occurs.

The crystal is assumed to be infinite in transverse directions. If the incident beam is a plane wave, the diffracted wave can be developed in a Fourier series in any plane parallel to the entrance face. The amplitudes of the Fourier coefficients are functions of the position of the plane and given by

$$\Psi(x, \boldsymbol{y})=\sum_{\boldsymbol{h}} \varphi_{\boldsymbol{h}}(x) \exp(2\pi i \boldsymbol{y}\boldsymbol{D}(\boldsymbol{h}+\boldsymbol{\varepsilon})) \qquad (1)$$

where $\boldsymbol{y}$ is a row-vector with 2 components y and z, $\boldsymbol{D}$ a 2×2 matrix taking into account the geometry of the crystal, $\boldsymbol{h}$ a column-vector with 2 components k and l, and $\boldsymbol{\varepsilon}$ the transverse components of the direction of the incident beam.

2. Semi-reciprocal formulation

A Fourier transformation in the yz plane gives the amplitudes of the components of the wave field

$$\left[K_{\boldsymbol{h}}^2+\frac{d^2}{dx^2}\right]\varphi_{\boldsymbol{h}}(x)=\sum_{\boldsymbol{h}'} v_{\boldsymbol{h}-\boldsymbol{h}'}(x)\varphi_{\boldsymbol{h}'}(x) , \qquad (2)$$

where $v_{\boldsymbol{h}}$ is $2m/\hbar$ times the coefficient of the development in the Fourier series of the "potential" in the plane at a depth x, and $K_{\boldsymbol{h}}$ is the x-component of the wave vector of the beam $\boldsymbol{h}$ in the vacuum.

3. Matrical and semi-reciprocal formulation

If $\boldsymbol{K}$ is a diagonal matrix with N elements on the diagonal (N: the number of possible difffracted beams) and $\boldsymbol{V}(x)$ a potential matrix with $N\times N$ elements as follows,

$$[\boldsymbol{K}]_{\boldsymbol{h}\boldsymbol{h}'}=\delta_{\boldsymbol{h}\boldsymbol{h}'}K_{\boldsymbol{h}} \qquad [\boldsymbol{V}(x)]_{\boldsymbol{h}\boldsymbol{h}'}=v_{\boldsymbol{h}-\boldsymbol{h}'}(x) ,$$

the equation (2) can be written

$$\frac{d}{dx} \mid \varphi(x) > = \mid \varphi'(x) >$$

$$\frac{d}{dx} \mid \varphi'(x) > = [-\boldsymbol{K}^2+\boldsymbol{V}(x)] \mid \varphi(x) >$$

or

$$\left[\boldsymbol{K}^2+\frac{d^2}{dx^2}\right] \mid \varphi(x) > = \boldsymbol{V}(x) \mid \varphi(x) > \qquad (3)$$

4. Green matrix

The equation (3) can be solved by use of a "Green matrix" $\boldsymbol{G}(x, x')$ satisfying

$$\left[\boldsymbol{K}^2+\frac{d^2}{dx^2}\right]\boldsymbol{G}(x, x')=\boldsymbol{I}\delta(x-x') .$$

Of two independent solutions, one corresponds to the emerging waves

$$\boldsymbol{G}(x, x')=-\frac{i}{2}\boldsymbol{K}^{-1} \exp(i|x-x'|\boldsymbol{K}) .$$

5. Integral equation

By use of a Green matrix, eq. (3) can be written as

$$|\varphi(x)> = \exp(ix\boldsymbol{K})|a_0> - i\int_0^1 \exp(i|x-x'|\boldsymbol{K}) \times\frac{1}{2}\boldsymbol{K}^{-1}\boldsymbol{V}(x') \mid \varphi(x') > dx' .$$

6. Born development

The integral equation shows that $\mid \varphi(x)>$ can be written in the form:

$$\mid \varphi(x) > = \sum_m \mid \varphi^{(m)}(x) >$$

where

$$|\varphi^{(m)}(x)> = -i\int_0^1 \exp(i|x-x'|K) \times \frac{1}{2}K^{-1}V(x')\,|\varphi^{(m-1)}(x')>dx'$$

and

$$|\varphi^{(0)}(x)> = \exp(ixK)|a_0>$$

7. Spatial frequencies appearing in the solution

If we observe certain spatial frequencies in an n-th term of the Born development, we observe in the $(n+1)$-th term the frequencies which result from the differences (and the sums) between (and of) these frequencies and all the K_h.

So, the number of the spatial frequencies involved for a given order of Born approximation increases very rapidly with the order and with the number of the beams.

8. Solution

The solution of the equation (3) as function of the initial conditions can be written simply with a $2N$-components super-vector consisting in the sequence of the components of $|\varphi(x)>$ and those of $d/dx\,|\varphi(x)>$:

$$\begin{bmatrix} |\varphi(x)> \\ |\varphi'(x)> \end{bmatrix} = Q(x)\begin{bmatrix} |\varphi_0> \\ |\varphi'_0> \end{bmatrix}$$

where

$$Q(x) = I + \int_0^x M(w)dw + \int_0^x M(w)\int_0^w M(v)dvdw + \cdots = \begin{bmatrix} Q_{11} & Q_{12} \\ Q_{21} & Q_{22} \end{bmatrix},$$

$$M(x) = \begin{bmatrix} O & I \\ -K^2+V(x) & O \end{bmatrix}.$$

Taking into account the entering fields, we obtain the reflected field $|b_0>$ and the transmitted field $|\varphi_1>$:

$$|b_0> = [A+B]^{-1}[B-A]\,|a_0>$$

$$|\varphi_1> = 2[Q_{11}[A+B]^{-1}B + iQ_{12}K[A+B]^{-1}A]\,|a_0>$$

with

$$A = KQ_{11} + iQ_{21} \text{ and } B = Q_{22}K - iKQ_{12}K.$$

9. Fujiwara's approximation

If one assumes that the back-travelling field is very small, the backward reflexion for each term of the Born development is weak, and the integral equation can be written as

$$|\varphi(x)> = \exp(ixK)[|a_0> - i\int_0^x \exp(-ix'K) \times \frac{1}{2}K^{-1}V(x')\,|\varphi(x')>dx']$$

and the differential equation is reduced to the first order equation

$$\frac{d}{dx}|\varphi(x)> = i\left[K - \frac{1}{2}K^{-1}V(x)\right]|\varphi(x)>.$$

Furthermore, if one assumes that $d/dx\,V(x)=O$ the solution is

$$|\varphi(x)> = \exp\left[ix\left(K - \frac{1}{2}K^{-1}V\right)\right]|a_0>.$$

In the general case $(d/dx\,V(x) \neq O)$,

$$|\varphi(x)> = Q(x)\,|a_0>$$

where

$$Q(x) = I + i\int_0^x \left[K - \frac{1}{2}K^{-1}V(w)\right]dw - \int_0^x \left[K - \frac{1}{2}K^{-1}V(w)\right]\int_0^w \left[K - \frac{1}{2}K^{-1}V(v)\right]dvdw + \cdots.$$

10. Fujimoto's approximation

We must remark that the commutator $[K, 1/2K^{-1}V]$ is different from zero, but small when $\varepsilon = o$ (normal case). In this case, we can find again Fujimoto's approximation, putting

$$K = kI.$$

We obtain

$$|\varphi(x)> = \exp(ixk)\exp\left(-\frac{i}{2k}xV\right)|a_0>.$$

Nnmerical applications

The matrical and semi-reciprocal formulation has been programmed recently on IBM 7090 computor for 31 beams, a column potential $(d/dx\,V(x)=O)$, an absorbing specimen and asymmetric structures.

11. Comparison with Fujiwara's theory

In the calculation, we use Fujiwara's theory as a first approximation for a thin crystal, so we can make comparisons if the thickness is below 6.4 Å.

We find that the *phases* of the diffracted beams out of the Bragg position are quite different between the two theories. It seems then that the theory of image formation is sensitive to the mode of approximation.

When the thickness increases, the beam *intensities* remain the same order of magnitude, but the differences increase between the results given by the different theories.

But it seems hazardous to extrapolate these discrepancies to thick specimens, because, for thin specimens these discrepancies perhaps result from the interferences between the perturbations of the waves on the entrance face and the exit face.

12. Two strong beams

If a reflexion is at the Bragg position, the beam corresponding to this reflexion interferes strongly with the direct beam and one obtains oscillations analogous to the pendulum solution.

The period is the same as that in Bethe's theory.

But, 1) the extinction of the direct beam is never complete,

2) the amplitudes of the weak beams are not a linear combination of the amplitudes of the two strong beams. These amplitudes show weak ripples about a mean value. As amplitudes of the ripples decrease, their period becomes short.

13. Three strong beams

If two reflexions are in Bragg position, we do not obtain a pendulum solution but a more complicated oscillating solution without complete extinction. At about 100 Å, the amplitudes seem to be stabilized as pointed out by Fengler.

14. Increase of the accelerating voltage

If we increase the accelerating voltage, the intensities do not tend uniformly to the kinematical intensities.

15. Tilt of the crystal

By tilting the crystal, we can obtain the condition of maximum for the forbidden reflexions. The forbidden reflexions are maximum at their Bragg position. In this position, the non forbidden reflexions from which the forbidden ones are caused are not maximum.

All the amplitudes and phases vary slowly with the tilting, even for the beams for which the excitation error remains the same.

16. Effect of the limitation of the number of beams

The suppression of the beams for which the structure factor is zero has no important effect on the values calculated for the amplitudes and phases of the strong and medium reflexions.

The calculation must be made with the pairs of beams having symmetric indices about 000, when their structure factors are not zero even if a beam is very weak. If this condition is not fulfilled, the results will be strongly non conservative for the current.

References

J. M. Cowley and A. F. Moodie: Acta Cryst. **12** (1959) 360.

H. Fengler: Dissertation, Berlin, 1960.

F. Fujimoto: J. Phys. Soc. Japan **14** (1959) 1558.

K. Fujiwara: J. Phys. Soc. Japan **14** (1959) 1513.

H. Niehrs: Z. Phys. **156** (1959) 446.

M. Tournarie: Bull. Soc. franç. Minér. Crist. **83** (1960) 179.

M. Tournarie: C. R. Acad. Sci. **252** (1961) 2862

DISCUSSION

L. STURKEY: As Dr. Tournarie has calculated, the intensity of a forbidden reflexion becomes maximum when the crystal is set for Bragg reflexion from the forbidden plane. I have made some experimental observations confirming this for Si: As the crystal is tilted so that the Bragg conditions for the (111) plane are satisfied, this reflexion is strongest. As the crystal is turned further, toward the (222) reflexion, this (222) reflexion becomes strongest, and with further tilting toward (333), the (333) becomes strongest. In this way, it was shown the forbidden reflexion (222) acts like a regular reflexion.