

K. MOLIÈRE: The excitation error and other parameters of the crystal orientation can be determined precisely from an analysis of the Kikuchi-pattern taken by focusing the beam at the crystal edge.

H. RRETHET: Is it possible to say something about the physical meaning of the absorption coefficient?

K. MOLIÈRE: I think at the moment that absorption includes all processes of removing electrons out of the coherent system of wave fields.

JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN

VOL. 17, SUPPLEMENT B-II, 1962

PROCEEDINGS OF INTERNATIONAL CONFERENCE ON MAGNETISM AND CRYSTALLOGRAPHY, 1961, VOL. II

## The Effect of Thermal Vibrations on Electron Diffraction

HIDE YOSHIOKA

*Physical Institute, Nagoya University*

AND

YOSHIRO KAINUMA

*College of General Education, Nagoya University, Nagoya, Japan*

The effect of thermal vibrations is treated in a formalism similar to that for the effect of inelastic waves (H. Yoshioka, *J. Phys. Soc. Japan* **12** (1957) 618). The phenomena of anomalous transmission of electrons in the case of Bragg reflexion can be explained largely in terms of the damping by the thermal diffuse scattering. The imaginary part of the correction for Fourier coefficients of the inner potential is estimated for the (200) reflexion of aluminium.

It is well known that the intensity of X-rays transmitted through a thick perfect crystal increases markedly when the crystal is set at a position satisfying the Bragg condition<sup>1)</sup>. Similar phenomena to this have been observed in electron diffraction<sup>2)</sup>. For example, anomalous transmission was observed in the Hillier pattern from a thick film of molybdenite. The mechanism for the X-ray case is explained as follows. According to the dynamical theory of diffraction, the wave field inside the crystal is approximated by a superposition of two components corresponding to the two points on the dispersion surface. One of them is a standing wave which has nodes on atomic planes parallel to the relevant net plane, whereas the other component has loops on atomic planes. Therefore, the former wave interacts only weakly with the electrons of the crystal, having very small probability of photo-ionization process. The slow decay of the former wave is the cause of the anomalous transmission. A similar

mechanism may be proposed for the case of electron diffraction. However, here the role of the photo-ionization must be replaced by scattering process. The author studied previously<sup>3)</sup> the effect of inelastic scattering on electron diffraction. According to the calculation, the apparent absorption by the process of inelastic scattering decreases indeed, when the crystal satisfies the Bragg condition. However, the calculated degree of the decrease is very small. This is because the range of the interaction between the incident electron and the electrons in the crystal is not so small as the interatomic distance. Therefore the standing wave nature of the incident electron has little effect on the inelastic scattering. The above consideration suggests that the dominant cause of the anomalous transmission may possibly be the thermal scattering of electrons.

The inner potential depends upon the state of lattice vibrations. We consider the fluctuating part of the potential



$$\Delta V(\mathbf{r}, q_k) = \sum_{lj} U_j(\mathbf{r} - \mathbf{R}_{lj} - \mathbf{u}_{lj}) - \langle a | U_j(\mathbf{r} - \mathbf{R}_{lj} - \mathbf{u}_{lj}) | a \rangle, \quad (1)$$

where  $\mathbf{r}$  is the coordinates of the incident electron,  $q_k$  is the coordinates of the thermal vibrations,  $U_j$  is the potential due to the  $j$ th atom,  $\mathbf{R}_{lj} = \mathbf{R}_l + \mathbf{r}_j$  is the equilibrium position of the  $j$ th atom of the  $l$ th cell,  $\mathbf{u}_{lj}$  is the displacement of this atom. The second term of the above expression is the expectation value of the potential in the state  $a$  of the

lattice vibration. The fluctuating part of the potential is confined mainly to the neighborhood of atomic nuclei. Therefore, the effect of the fluctuating part is expected to depend on the standing wave pattern. We can represent the effect due to this fluctuating part as correction terms  $C_{hg}$  to the Fourier coefficients  $V_h$  of the inner potential, in quite a similar way to that treated by us for the case of inelastic scattering. The expression is

$$C_{hg} = -\frac{2m}{(2\pi)^3 \hbar^2 V} \int \int \frac{\langle a | \Delta V(\mathbf{r}, q_k) \Delta V(\mathbf{r}', q_k) | a \rangle \exp\{-i(\mathbf{k}_h - \mathbf{k}')\mathbf{r} + i(\mathbf{k}_g - \mathbf{k}')\mathbf{r}'\} d\mathbf{r} d\mathbf{r}'}{k'^2 - k_0^2 - i\varepsilon} d\mathbf{k}', \quad (2)$$

where  $\varepsilon$  is a positive infinitesimal quantity. The fundamental equations of the dynamical theory modified are

$$\left\{ \frac{\hbar^2}{2m} (\mathbf{k}_h^2 - k_0^2) + V_0 + C_{hh} \right\} \chi_h + \sum_g (V_{h-g} + C_{hg}) \chi_g = 0. \quad (3)$$

The Fourier coefficient  $V_h$  is an expectation value in the state  $a$  as  $C_{hg}$  is. We replace the expectation values in the state  $a$  by the statistical averages. Strictly speaking, we must take the statistical average in the final expression of the intensity of transmitted or reflected beams. However, our procedure of taking averages may be allowed and lead to a good approximation, because the number of phonons in the state is very large. Then, using the relation

$$\langle \exp(-i\mathbf{k}\mathbf{u}_{lj}) \rangle_{AV} = \exp\{-\langle (\mathbf{k}, \mathbf{u}_{lj})^2 \rangle_{AV}/2\} \quad (4)$$

the integral over the coordinates  $\mathbf{r}, \mathbf{r}'$  in the expression of  $C_{hg}$  becomes

$$\sum_{lmjk} E_j^t(\mathbf{k}_h - \mathbf{k}') E_k^t(\mathbf{k}_g - \mathbf{k}')^* \langle (\mathbf{k}_h - \mathbf{k}', \mathbf{u}_{lj})(\mathbf{k}_g - \mathbf{k}', \mathbf{u}_{mk}) \rangle_{AV} \exp\{-i(\mathbf{k}_h - \mathbf{k}')\mathbf{R}_{lj} + i(\mathbf{k}_g - \mathbf{k}')\mathbf{R}_{mk}\}, \quad (5)$$

where  $E_j^t$  is the atom form factor at the temperature  $T$ ,

$$E_j^t = \exp(-M_j) E_j = \exp(-M_j) m_j^{-\frac{1}{2}} \int U_j(\mathbf{r}) \exp\{-i\mathbf{k}\mathbf{r}\} d\mathbf{r}, \quad (6)$$

where  $M_j$  is half of the exponent of the Debye-Waller factor,  $M_j = \langle (\mathbf{u}_j, \mathbf{k})^2 \rangle_{AV}/2$ , and  $m_j$  is the mass of the  $j$ th atom. Representing  $\mathbf{u}_{lj}$  in terms of normal coordinates, we have

$$C_{hg} = -\frac{2m}{(2\pi)^3 \hbar^2 v} \int \frac{d\mathbf{k}'}{k'^2 - k_0^2 - i\varepsilon} \left[ \sum_{i=1,2,3} \frac{\hbar(2n_{\tau s} + 1)}{2\omega_{\tau s}} (\mathbf{e}_{\tau s}, \mathbf{k}_h - \mathbf{k}') (\mathbf{e}_{\tau s}, \mathbf{k}_g - \mathbf{k}') F^t(\mathbf{k}_h - \mathbf{k}') F^t(\mathbf{k}_g - \mathbf{k}')^* \right], \quad (7)$$

where  $v$  is the volume of unit-cell,  $n_{\tau s}$  is the number of phonons with the frequency  $\omega_{\tau s}$  and the polarization  $\mathbf{e}_{\tau s}$ ,  $\tau$  is the wave vector of the phonon satisfying

$$\mathbf{k} - \mathbf{k}' + \boldsymbol{\tau} = 2\pi \mathbf{b}. \quad (8)$$

where  $\mathbf{b}$  is a reciprocal lattice vector;  $F^t(\mathbf{k}_h - \mathbf{k}')$  is given by

$$F^t(\mathbf{k}_h - \mathbf{k}') = \sum_j E_j^t(\mathbf{k}_h - \mathbf{k}') \exp\{-i(\mathbf{k}_h - \mathbf{k}', \mathbf{r}_j)\}. \quad (9)$$

The decay of the two wave fields in the case of  $\mathbf{h}$  reflexion is determined by  $C_{00}^i \pm C_{0h}^i$ , where the suffix  $i$  means the imaginary part. This part is expressed as a surface integral over the Ewald sphere,



$$C_{hg}^i = -\frac{m}{(2\pi)^3 \hbar^2 v} \frac{\pi}{k_0} \int dS \left[ \sum_{i=1,2,3} \frac{\hbar(2n_{\tau s}+1)}{2\omega_{\tau s}} (e_{\tau s}, \mathbf{k}_h - \mathbf{k}') (e_{\tau s}, \mathbf{k}_g - \mathbf{k}') F^t(\mathbf{k}_h - \mathbf{k}') F^t(\mathbf{k}_g - \mathbf{k}')^* \right]. \quad (10)$$

If we assume that a unit-cell contains only one atom and that the three modes of acoustic waves have polarizations perpendicular to one another and have equal velocities,  $C_{0h}^i$  is given by

$$C_{0h}^i = -\frac{m}{\hbar^2 v (2\pi)^3 k_0} \int dS \frac{\kappa T}{c^2 \tau^2} (x^2 + y^2 - p^2) F^t(x) F^t(y)^*, \quad (11)$$

where  $c$  is the velocity of acoustic waves, and  $x = |\mathbf{k} - \mathbf{k}'|$ ,  $y = |\mathbf{k}_h - \mathbf{k}'|$  and  $p = |\pi \mathbf{h}|$ . We draw a circle with a radius  $p$  on the Ewald sphere so that the origin and the lattice point  $2\pi \mathbf{h}$  are diametrical. Then, (11) shows that the diffuse scattering which occurs outside the circle favours the anomalous transmission, whereas the diffuse scattering inside the circle cancels the effect.

The values of  $C_{hg}$  were estimated for the case of (200) reflexion of aluminium, assuming  $\lambda = 0.06 \text{ \AA}$  and using the Debye temperature  $\theta = 398^\circ \text{K}$ . In the calculation, the minimum value of the wave vector  $\tau/2\pi$  was put equal to the inverse of the linear dimension of the crystal  $1/L$ ,  $L$  being assumed to be  $400 \text{ \AA}$ . If  $\tau_{\min} = 0$ , the integrals diverge. In the first column of the Table 1 are shown the values which are calculated assuming that only the diffuse scattering due to the "systematic" interactions (diffuse scattering corresponding to the regions close to the lattice points of  $(h00)$  type of the reciprocal space) takes place. The second column shows the values for the case where the incident beam is parallel to the  $(010)$  plane of the crystal. It is evident that the contribution from the "accidental" diffuse scattering can be very large. Therefore, the values of  $C_{hg}^i$  should be strongly dependent on the azimuth of the incident beam.

Table I. Values of  $C_{hg}^i$  for aluminium.

	$C_{00}^i$ (ev)	$C_{0h}^i$ (ev) ( $h$ for (200))
syst.	0.20	0.084
total	0.64	0.47

### References

- 1 G. Borrmann: *Z. Phys.* **127** (1950) 297.
- 2 G. Honjo and K. Mihama: *J. Phys. Soc. Japan* **9** (1954) 184.
- 3 H. Yoshioka: *J. Phys. Soc. Japan* **12** (1957) 618.
- Phys. Soc. Japan **14** (1959) 1119, **16** (1961) 580.
- H. Hashimoto, A. Howie and M. J. Whelan: *Phil. Mag.* **5** (1960) 967.

### DISCUSSION

S. MIYAKE: I should like to give a comment which is related to both the papers of Drs. Yoshioka and Gjønness. I had recently an opportunity to make some discussion with Prof. Ewald, when he told me: In the dynamical theory of X-ray diffraction, the generation of diffracted waves and, at the same time, the change of the state of the scattering system are considered, and thus the theory can be called "the dynamical theory" in its true sense. The dispersion effect is due to the change of the scattering system. The current dynamical theory of electron diffraction, on the other hand, assumes a static electric potential in the crystal at the beginning, and the change of the state of the scattering system is not taken into account explicitly. Prof. Ewald pointed out in this that there might still remain a lack of the thorough correspondence between both the dynamical theories of X-ray and electron diffraction.

After this discussion, I became to realize the fact that the theory originated by Yoshioka, which was derived by taking account of the possibility of changes in states of the scattering system, may be the treatment filling up the pointed-out gap. I discussed about relevant things with Dr. Fujiwara, Prof. Kato and Prof. Kainuma, and we came to consider that the sought correspondence between the dynamical theory of X-ray diffraction and that of electron diffraction seems to have already been established.

H. YOSHIOKA: I almost agree with Prof. Miyake's opinion.