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

Study on the Absorption of Electron Wave Fields in Ideal Crystals by Interference Double Refraction Experiments

G. LEHMPFUHL AND K. MOLIÈRE
Fritz-Haber-Institut der Max-Planck-Gesellschaft
Berlian-Dahlem, Deutschland

This article reports a method of quantitative determination of absorption coefficients by measuring half widths of double refraction spots. The absorption coefficients are evaluated for the 220 reflection of a MgO crystal as function of the energy in the range 30–70 kV. They turn out to be of the order of 5 to 7 times 10^5 cm^{-1} and show a minimum below 50 kV. The imaginary parts of the structure potentials obtained from the half widths are in good agreement with the calculations of Yoshioka not taking into account the thermal scattering.

1. Introduction

Evidence of the absorption of electron wave fields (anomalous absorption) was observed some years ago in the experiments on interference double refraction. There exists an angular spread of the diffraction spots perpendicularly to the crystal edge. The spread is different for the two coponents of a diffraction doublet (Altenhein and Molière, 1954[1]) and the intensities of the two spots are different. (Honjo and Mihama, 1954[2]). These facts are due to the absorption, and can not be explained by the dynamical theory neglecting absorption.

This article gives an account of a method of quantitative determination of absorption coefficients by measuring half widths of double refraction spots. The first idea of this possibility is due to Niehrs.[3] The absorption coefficients are evaluated for the 220 reflection from MgO crystal as function of the energy in the range 30–70 kV. The imaginary parts of the structure potentials obtained from the half widths are in good agreement with the calculations of Yoshioka[4] not taking into account the thermal scattering.

2. Theory of diffraction by an absorbing crystal wedge

M. von Laue[4] emphasized in 1952 the fact that, in the wave-mechanical treatment of particle motion in a crystal field, the Bloch wave fields have to be considered as physical entities. According as the distribution of the stationary current density of a special wave field in the crystal lattice, there should exist a certain amount of inelastic interaction between the particles and the crystal atoms, giving rise to a characteristic absorption coefficient of the wave field.

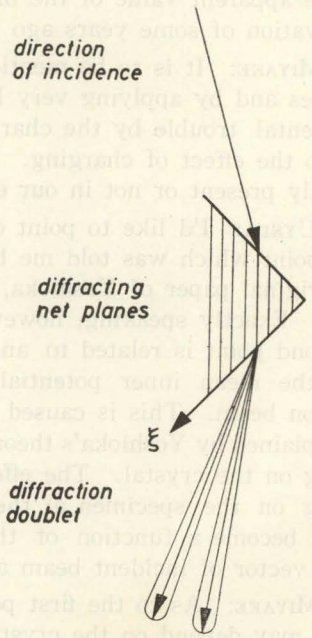

Fig. 1. Interference double refraction at a crystal wedge.

For the sake of simplicity, let us consider a crystal wedge limited by two surfaces perpendicular to each other (Fig. 1: $\xi=0$ entrance surface, $\eta=0$ exit surface), and the two-beam case. If we neglect the waves reflected at the surfaces as usual, the wave vectors $\boldsymbol{k}_h^j$ and the amplitudes $u_h^j(h=0=(0,0,0)$ or $g=(g_1g_2g_3)$; $j=1, 2)$ may be calculated by the well known dynamical procedure from the conditions of incidence. It is known

that

$$|u_g^1|=|u_g^2| \ . \qquad (1)$$

We have to assume that each wave field is attenuated in the direction ξ, or the inner normal of the entrance surface, by an exponential factor. Then the amplitude at the exit surface becomes a sum of the partial wave functions

$$u_h^j \exp\left(2\pi i k_{h\xi}^j - \frac{1}{2}\mu_j\right)\xi \cdot f(\zeta) \ . \qquad (2)$$

$f(\zeta)$ needs not be discussed here; it is sufficient to assume that $f(\zeta)\neq 0$ only in a limited range of ζ. The factor 1/2 in the second term of the exponential means that μ_j is the absorption coefficient of the mean current density of the wave field.

According to diffraction theory the vacuum field at a large distance on the exit side may be represented by a Fourier transform of the amplitudes on the exit surface F:

$$\psi(\boldsymbol{S})=C\iint_F \exp\left(-\frac{2\pi \mathrm{i}}{\lambda}\boldsymbol{S}, \boldsymbol{r}\right)\psi(\boldsymbol{r})dF \ . \qquad (3)$$

$\boldsymbol{S}$ is the unit vector in the direction to the point of observation. By inserting (2) for $\psi(\boldsymbol{r})$ into (3), $\psi(\boldsymbol{S})$ turns out to be a sum of terms, each of which contains the following factor originating from the integration over ξ:

$$\left[(S_\xi-\beta_{h\xi}^j)+i\frac{\lambda}{4\pi}\mu_j\right]^{-1} , \qquad (4)$$

where

$$S_\xi=\cos(\boldsymbol{S}, \xi) \ , \qquad (5)$$

and

$$\beta_{h\xi}^j=\lambda k_{h\xi}^j \ . \qquad (6)$$

If there is no overlapping of the functions (4), the intensity becomes a sum of terms proportional to

$$J_h^j=|u_h{}^j|^2\cdot\left[(S_\xi-\beta_{h\xi}^j)^2+\left(\frac{\lambda}{4\pi}\mu_j\right)^2\right]^{-1} . \qquad (7)$$

Formula (7) should be a good approximation for the intensity distribution of the diffracted spots with $h=g$.

From the consideration of eqs. (7) and (1) we obtain the following relations:

1) The intensity is maximum for

$$S_\xi=\beta_g^1, \beta_g^2 \ . \qquad (8)$$

(As to the wave field indices we assume $\beta_g{}^1<\beta_g{}^2$.)

2) The ratio of the maxima is

$$\frac{J_{g,\max}^2}{J_{g,\max}^1}=\left(\frac{\mu_1}{\mu_2}\right)^2 . \qquad (9)$$

3) The integrated intensities are proportional to $\mu_j{}^{-1}$:

$$\frac{J_{g,\mathrm{int}}^2}{J_{g,\mathrm{int}}^1}=\frac{\mu_1}{\mu_2} \qquad (10)$$

4) The half width of a spot is proportional to μ_j:

$$\frac{H_j}{L}=\frac{\lambda}{2\pi}[1-(\beta_g^j)^2]^{-1/2}\cdot\mu_j \ . \qquad (11)$$

(H_j=half width measured on the plate, L=camera length.)

By the the usual dynamical calculation for non-absorbing crystals we have[5]

$$\beta_g{}^{1,2}=\beta_g+(2E\beta_g)^{-1}[E\rho_g+\phi_0\cdot c\mp\Delta| \ . \qquad (12)$$

The meanings of the symbols are: E=acceleration potential, ϕ_0, ϕ_g=structure potentials (eV), $\rho_g=\lambda\cdot|b_g|(\theta_0-\theta_g)$=excitation error ($\theta_0$=angle of the incident beam S_0 with the net planes, θ_g=Bragg angle), $c=(\beta_g/\beta_0)^{1/2}$ (equal to 1 within a few percent), $\beta_0=S_{0\xi}$, $\beta_g=S_{0\xi}+\lambda b_{g\xi}$ and

$$\Delta=\{[E\rho_g-\phi_0(c-1)]^2+\phi_g\phi_{\bar{g}}c^2\}^{1/2} \ . \qquad (13)$$

The absorption of wave fields may be formally represented by additional imaginary parts of the structure potentials[6),7)]. For centrosymmetrical crystals, substituting $\phi_0\rightarrow\phi_{0r}+i\phi_{0i}$, $\phi_g\rightarrow\phi_{gr}+i\phi_{gi}$, we have $\phi_g\phi_{\bar{g}}=(\phi_{gr}^2-\phi_{gi}^2)+2i\phi_{gr}\phi_{gi}$. On the left side of (12) we have to add an imaginary term $i(\lambda/4\pi)\mu_{1,2}$. Equating the real parts and the imaginary parts respectively, we obtain expressions for ϕ_{0i} and the imaginary part of Δ, analogously to v. Laue's treatment for X-rays[8)]. Using eq. (11) these two quantities can be expressed by the sum and difference of the half widths, respectively:

$$\phi_{0i}=c^{-1}\frac{E}{L}\beta_g[1-\beta_g^2]^{1/2}\frac{H_2+H_1}{2} \ , \qquad (14)$$

and

$$\Delta_i=\frac{E}{L}\beta_g[1-\beta_g{}^2]^{1/2}\frac{H_2-H_1}{2} \ , \qquad (15)$$

where the approximate value β_g is used for β_g^1 and β_g^2 in the square roots.

By evaluating Δ_r and Δ_i in terms of potetials and excitation error, we can calculate ϕ_{gi} by $\phi_{gr}, \phi_{0r}, \phi_{0i}$ and ρ_g.

3. Experiments

The experimental arrangement has been described in a previous paper[9)]. Using a

tungsten point cathode[10] and a two-lens condenser we obtained a focus diameter of 0,5μ at the specimen, and of 3–4μ at the plate. The camera length was 405 mm. Because the widths of the spots to be measured (30μ) were of the same order as the point resolution of photographic emulsions, we transformed the points into streaks by an electrostatic deflexion system as described in our previous paper[9], fed with a 50 cps saw-tooth emf. The streak resolution was found to be 4μ[11]. For measuring half widths we made two records simultaneously on the same

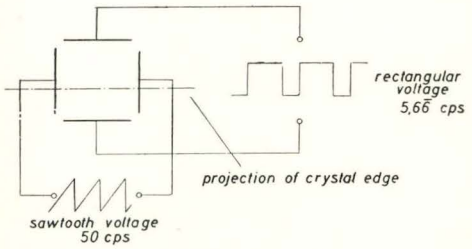

Fig. 2. Electrostatic deflexion system for double exposure with time ratio 1:2.

photographic plate with a ratio 1:2 of exposure time. This was achieved by a deflexion device with two pairs of plates perpendicular to each other, the second pair fed by a periodical change of two constant voltages with time ratio 1:2, as shown in Fig. 2. The two deflexion fields were synchronized.

The specimen was a crystal wedge of MgO with two fresh cleavage faces (100) and (010). It has been shown previously[9] that it is possible to detect portions of crystal wedges having a practically ideal character over ranges up to 20μ at the crystal edge.

The investigated reflections were 220 and $\bar{2}\bar{2}0$. The direction of the incident beam was nearly along the axis [1$\bar{1}$0], and was so adjusted that, with focussing at the crystal edge, only the desired reflection was visible on the fluorescent screen. The orientation of the crystal was determined from a Kikuchi-pattern taken with the same focus position[9].

For the measurement of the half widths, the beam was focussed on the screen and the crystal was cautiously dipped into the beam by lateral shifting until the doublet made its appearance The deflecting saw-tooth field adjusted parallel to the crystal edge. Fig. 3a shows a record of a refraction doublet consisting of two double streaks with ratio 1:2 of exposure times, and Fig. 3c the curve of the corresponding microphotometry, giving an example for the determination of the half widths H_1 and H_2. A second streak record

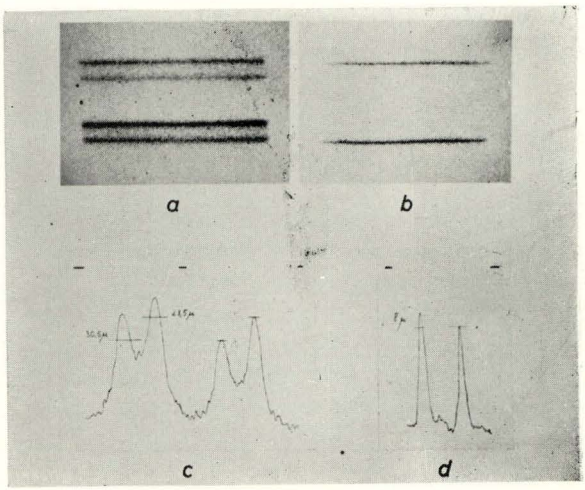

Fig. 3. Photographs and microphotograms for a 220-doublet of MgO.

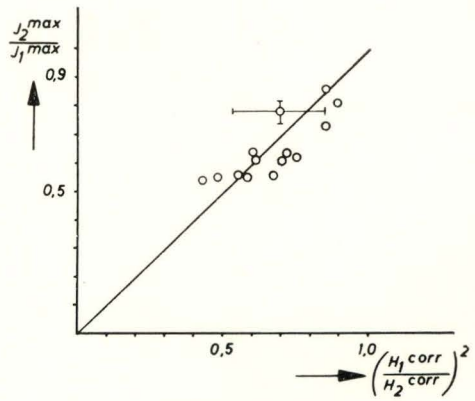

Fig. 4. Test of formula (9).

(Fig. 3b and d) obtained with the saw-tooth field perpendicular to the crystal edge gives the "natural half width" H_0, which is not due to absorption. The "corrected half widths" $H_j^{corr}=(H_j^2-H_0^2)^{1/2}$ are assumed to give the part of the half widths due to absortion.

The measured half widths have been found to be reproducible with a mean error of $\pm 2\mu$, mainly due to the resolution of the photographic plates.

By evaluation of blackening curves in some of the experiments we convinced ourselves that the intensities are in accordance with the requirements of eqs. (9)-(11). Fig. 4 shows a test proving that the maximal intensities are inversely proportional to the square of the half widths within the experimental error, which is indicated by the cross drawn for one of the measure points.

4. Results

A plot of the absorption coefficients as function of the energy, calculated from the half widths according to eq. (11) is shown in Fig. 5. The five pairs of dots and circles

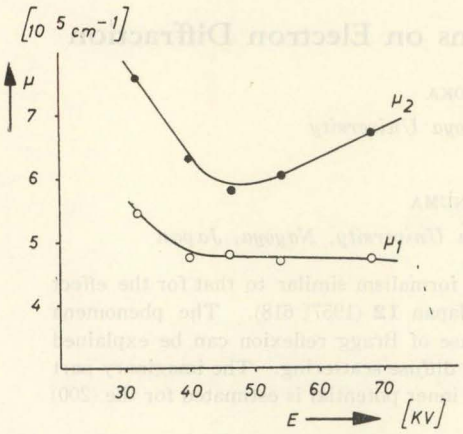

Fig. 5. Absorption coefficients as function of the energy.

represent only a selected example of the experiments. The mean error of a single measurement is about 7%.

The arithmetical mean of the two curves gives the normal absorption coefficient μ_0 for the case where the Laue condition is satisfied. μ_0 is proportional to ϕ_{0i} (cf. eq. (14)).

We have no explanation at present for the fact that μ_0 seems to have a minimum just below 50 kV.

Fig. 6 gives the imaginary structure potentials ϕ_{0i} and $\phi_{220,i}$ as function of the energy, calculated by the formulae (13)-(15) with the assumption of the values $\phi_{gr}=5{,}1$ V, $\phi_{0r}=14{,}9$ V[9].

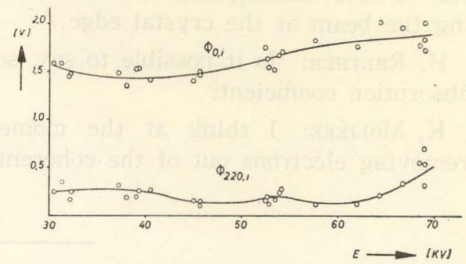

Fig. 6. Imaginary structure potential as function of the energy.

The mean errors of a single measurement are about 7% for the upper curve (ϕ_{0i}) and about 50% for the $\phi_{210,i}$ curve. We dare not to say whether the maxima and minima of the lower curve are real, but at present we have no argument for the assumption that the curve must be represented by a smooth function.

It is remarkable that our values agree excellently with the calculations* of Yoshioka[4]. For 40 kV our values are $\phi_{0i}=1{,}4$ V and $\phi_{220,i}=0{,}25$ V. The first figure may be directly compared with the value $-C^i_{00}=1{,}47$ V of Table I in Yoshioka's paper, the second one is equal within the experimental error to his value $C^i_{h0}=0.14$ V.

References

1 H. J. Altenhein and K. Molière: Z. Phys. **139** (1954) 113.
2 G. Honjo and K. Mihama: J. Phys. Soc. Japan **9** (1954) 195.
3 H. Niehrs: Phys. Verh. **5** (1954) 175.
4 H. Yoshioka: J. Phys. Soc. Japan **12** (1957) 618.
5 K. Molière and H. Niehrs: Z. Phys. **137** (1954) 445.
6 M. v. Laue: Acta Cryst. **6** (1953) 217.
7 K. Molière: Ann. Phys. Lpz. **34** (1939) 461.
8 M. v. Laue: Acta Cryst. **2** (1949) 106.
9 G. Lehmpfuhl and K. Molière: Z. Phys. **164** (1961) 389.
10 S. Maruse: *Proc. Eur. Reg. Conf. on Electron Microscopy (Delft 1960)* Vol. I, 73.
11 G. Lehmpfuhl: Z. Naturforsch. **16A** (1961) 716.

DISCUSSION

G. Honjo: I think the effect of the anomalous absorption effect depends on the resonance error. How did you determine the resonance error?

* In these calculations, the thermal scattering, which is taken into account in his paper given in this volume, is not considered.

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

H. RRETHER: 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[1]. Similar phenomena to this have been observed in electron diffraction[2]. 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[3] 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