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

Absorption and Diffraction Effects of Electron Waves Observed in 300 kV Electron Microscopic Images

H. HASHIMOTO AND K. TANAKA,
Department of Physics, Kyoto University
Kyoto, Japan

K. KOBAYASHI AND E. SUITO,
Institute for Chemical Research, Kyoto University
Kyoto, Japan

AND

S. SHIMADZU AND M. IWANAGA
Shimadzu Seisakusho, Co. Ltd.
Kyoto, Japan

By using 300 kV electron microscope, some quantitative observations of the energy dependence of dynamical extinction distance and transmissive power of electron waves in crystalline material have been carried out. The results show the relativistic correction of mass of electron must not be disregarded both for the elastic and inelastic scattering. Energy dependence of the visibility of thickness fringe is also discussed. A few electron micrographs taken at 300 kV are illustrated.

1. Introduction

It is expected that the character of transmission electron microscopic images can be improved by elevating the accelerating voltage, i.e. the extinction distance and transmissive power of electron wave will increase with increasing accelerating potential of the incident electron wave. By using 300 kV electron microscope in Kyoto University, the energy dependence of these values in aluminum single crystal were measured.

2. Energy dependence of extinction distance

According to the dynamical theory of electron diffraction developed by Bethe, electron waves entering a crystal set at the Bragg condition have a periodic intensity both in primary and diffracted waves. This extinction distance is given by

$$\xi_g = \frac{\kappa}{U_g} = \frac{h^2}{2meV_g}\frac{1}{\lambda} \tag{1}$$

where κ is wave vector given by $1/\lambda$, λ is wavelength and U_g is $(2meV_g/h^2)$, V_g being Fourier coefficient of crystal lattice potential.

Fujiwara[1] pointed out to one of the present authors that eq. (1) should be replaced by

$$\xi_g = \frac{h^2}{2meV_g}\frac{1}{\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)}} \tag{2}$$

if the relativistic effect is taken into account. Later, Howie and Whelan[2] have informed to one of the present authors that they have also obtained the same conclusion independently.

If a crystal has a cross section of wedge shape and is in the Bragg angle, the thickness fringes corresponding to the periodic intensity of the electron wave can be seen in its transmission electron microscopic images.

Fig. 1 is an electron microscopic image of electro-polished aluminum foil. The thickness of the specimen increases gradually from its edge (indicated by an arrow) to the upper part of the figure. As the crystal bends cylindrically along the edge, only the portion along a line perpendicular to the edge is in exact Bragg condition for a net plane. One of the two white bands in Fig. 1 which is perpendicular to the edge and is indicated by an arrow mark is due to (111) Bragg reflection.

Near the edge of the crystal, equally spaced and dark broad lines can be seen along the band. These lines are the thickness contour fringes due to the Bragg reflection. From the spacings of the fringes the extinction distance of electron wave in crystal can be measured if the local thickness of the crystal is known. As it is very difficult to know

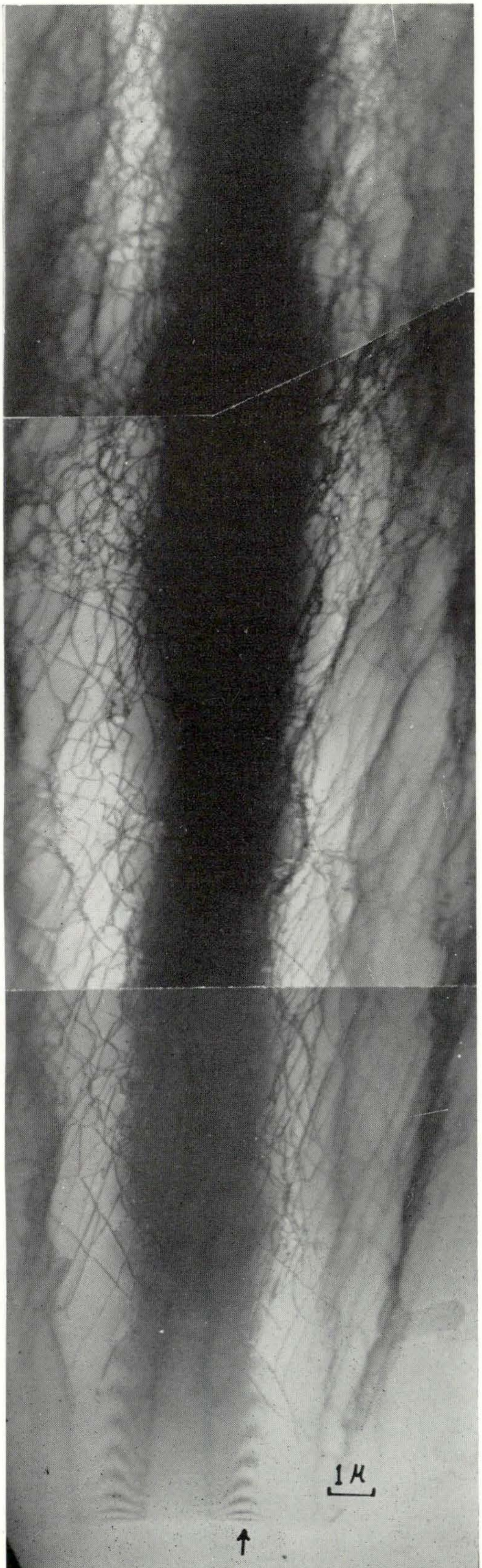

Fig. 1. Electro-polished Al-foil, (300 kV)

the local thickness of the crystal, the theoretical value of the extinction distance at 100 kV was used as a scale to measure the local thickness. After the measurement of the local thickness of the crystal, the extinction distances at 200 kV and 300 kV were measured and are shown in table 1

Table I.

	100 kV	200 kV	300 kV	
ξ_g	(584 Å)	738±10 Å	817±13 Å	observed
	(584 Å)	(740 Å)	(828 Å)	calculated
v_g	(5.72 V)	5.74±0.08 V	5.80±0.09 V	observed

These values were compared with theoretical values which are corrected by the theory of relativity. The agreement between them is fairly good. The value V_g which is calculated from the observed values of extinction distance ξ_g at different voltages gives nearly constant value.

3. Energy dependence of transmissive power

As well known, in the dynamical theory of diffraction, absorption is represented by the addition of a small periodic imaginary part $V_i(\boldsymbol{r})$ to the crystal lattice potential $V(\boldsymbol{r})$, i.e. the absorption coefficient of electron wave is given by

$$\varepsilon=\frac{2\pi}{\kappa}\left(U_{0i}\mp\frac{U_{gi}}{\sqrt{1+x^2}}\right)=\varepsilon_0\mp\Delta\varepsilon$$

where U_{0i} and U_{gi} are the o-th and g-th Fourier coefficient of $(2meV_i(\boldsymbol{r})/h^2)$, and x is the parameter representing the deviation from the Bragg angle. At the large deviation from the Bragg angle, ε tends to ε_0 which is the mean absorption coefficient. From the decrease in intensity of extinction contour lines which are indicated by an arrow in Fig. 1, the absorption coefficients of electron wave were measured.

According to the theory of absorption[3], the amplitude A of periodic intensity of thickness fringe is given by

$$A=\tfrac{1}{2}\exp-(\varepsilon_0 Z)$$

and the background of the intensity B is given by

$$B=\tfrac{1}{2}\exp(-\varepsilon_0 Z)\cdot\cosh(\Delta\varepsilon\cdot Z)$$

if the crystal is at the Bragg condition. Then, ε_0 and $\Delta\varepsilon$ are obtained by measuring the intensities A and B and the local thickness of the crystal Z. As is pointed out by Kamiya and Uyeda[4], the contrast of the image may be influenced by the size of the aperture of objective lens. The observations, therefore, have been carried out at the voltages of 100 kV, 200 kV and 300 kV, and with two apertures of objective lens. The reciprocal of observed values of ε_0 and $\Delta\varepsilon$ are shown in Table II. The effective aperture sizes are also shown in Table.

Table II.

	Size of aperture	100 kV	200 kV	300 kV
$1/\varepsilon_0$	large	1420 Å	2320 Å	2840 Å
	small		2260 Å	2840 Å
$1/\Delta\varepsilon$	large	2300 Å	3350 Å	3800 Å
	small		4730 Å	4630 Å
$2\theta/\lambda$	large	0.471 Å^{-1}	0.628 Å^{-1}	0.816 Å^{-1}
	small		0.356 Å^{-1}	0.463 Å^{-1}

In a private discussion, Miyake[5] suggested to one of the authors that the relativistic correction of mass should not be disregarded also for the inelastic scattering. (See also the paper of Miyake, Fujiwara and Suzuki in this volume[6]). Hirsch, Howie and Whelan[7] have pointed out in their private communication to one of the authors that the transmissive power should be proportional to $(v/c)^2$ from their theoretical point of view. $(v/c)^2$ is proportional to $(1/\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)})^2$.

Reciprocal of absorption coefficient, which gives the transmissive power of electron wave, are plotted against

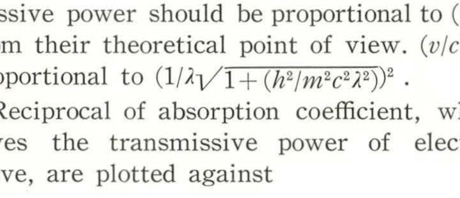

Fig. 2. Transmissive power of electron wave.

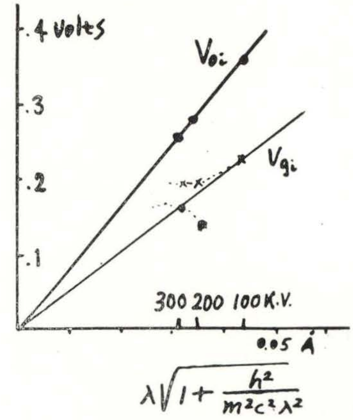

Fig. 3. Energy dependence of imaginary potential.

$$(1/\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)})^2$$

as shown in Fig. 2. $1/\varepsilon_0$ does not depend on the size of aperture of objective lens and is proportional to $(1/\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)})^2$. $1/\Delta\varepsilon$ gives different values for the different apertures, but when the effective aperture is the same it is proportional to $(1/\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)})^2$.

V_{0i} and V_{gi} which are calculated from the observed values ε_0 and $\Delta\varepsilon$ are plotted against

$$\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)}$$

as shown in Fig. 3. V_{0i}, V_{gi} are proportional to $\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)}$ or (c/v), where v is the velocity of electron.

4. Discussion

As was observed in the present experiment, the theoretical values of the extinction distance of electron wave at different voltages which are corrected by the theory of relativity coincide with the observed values. The extinction distance increases with increasing $1/\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)}$. Therefore it may be concluded that the present observation gives an experimental support to the theory of relativity.

As is shown in Fig. 3, V_{0i} and V_{gi} are proportional to $\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)}$.

The amount of absorption is given by

$$\varepsilon Z=2\pi n\left(V_{0i}\mp\frac{V_{gi}}{\sqrt{1+x^2}}\right)\Big/ V_g=n\cdot A\cdot\lambda\sqrt{1+\frac{h^2}{m^2c^2\lambda^2}},$$

where Z is the thickness of the crystal, and n is the number of the thickness fringe and A is a constant. The visibility of the thickness fringe is given by

$$1/(nA\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)}) \propto (v/c)\,.$$

The number of visible fringes increases proportionally to

$$1/(A\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)}) \text{ or } (v/c)\,.$$

5. Conclusion

The extinction distance increases with increasing $1/(\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)})$ or (v/c) and the transmissive power of electron wave increases with increasing

$$(1/\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)})^2 \quad \text{or} \quad (v/c)^2\,.$$

The visibility of the thickness fringe increases with increasing

$$(1/\lambda\sqrt{1+(h^2/m^2c^2\lambda^2)}) \text{ or } (v/c)\,.$$

Fig. 4 shows the intensity of primary electron wave in crystal in different accelerating voltages. The ratio of (v/c) and $(v/c)^2$ are shown in Table III for 100 kV, 200 kV and 300 kV.

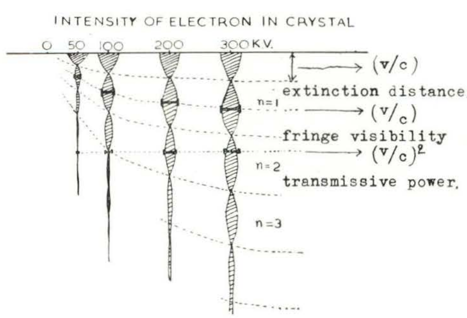

Fig. 4. Intensity of electron wave in crystal.

Table III.

		100 kV	200 kV	300 kV
v/c	$1/\lambda\sqrt{1+\frac{h^2}{m^2c^2\lambda^2}}$	1	1.26	1.42
$(v/c)^2$	$\left(1/\lambda\sqrt{1+\frac{h^2}{m^2c^2\lambda^2}}\right)^2$	1	1.60	2.01

6. Pictures taken with 300 kV electron microscope

Since the transmissive power of electron wave increases with increasing the accelerating voltages, the structure inside the thick crystal can be seen by the 300 kV electron microscope. Some illustrations are shown in Figs. 1, 5, 6, 7 and 8.

In Fig. 1, the spacing of thickness fringe corresponds to the thickness 828 Å, then the thickness of the film at the portion where the thickness fringe almost disappears is about 1 micron. Thickness increases along the direction which is indicated by an arrow. Fig. 5 shows the thickest part of the same specimen which is shown in Fig. 1. The thickness of the film was measured by micrometer as 10 microns. Fig. 6 is a picture of electropolished stainless steel film. A fringe due to a stacking fault is observed. The thickness of the film can be measured from the number of the fringe as 8400 Å. Fig. 7 shows a grain boundary in stainless steel

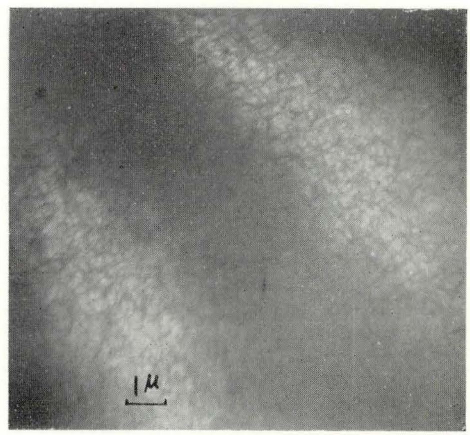

Fig. 5. Electro-polished Al-foil 10 microns. (300 kV)

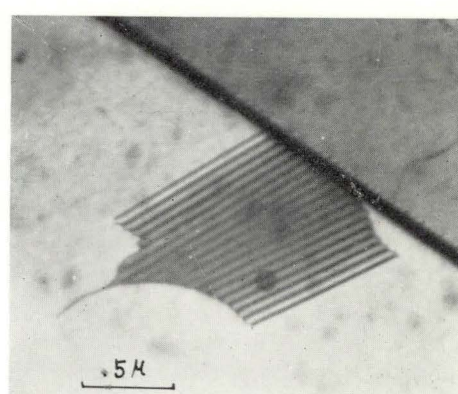

Fig. 6. Stacking fault in stainless steel. (300 kV)

film. From the spacing and the number of the fringes which appeared in the boundary, the thickness of the film can be estimated to be 8800 Å. Dislocations in this grain boundary do not show the dotted structure though they are inclined to the crystal surface and their contrast decreases near the portion where the fringe appears. Such contrast of dislocation has been explained in terms of absorption of electron waves. Fig. 8 is an electron micrograph of polyoxymethylene crystal taken at 250 kV. Rotation moiré pattern due to the several layers which are grown in spiral can be seen. The damage due to the electron irradiation of 250 kV electron was very small for the organic material such as polyoxymethylene and revealed crystalline structure.

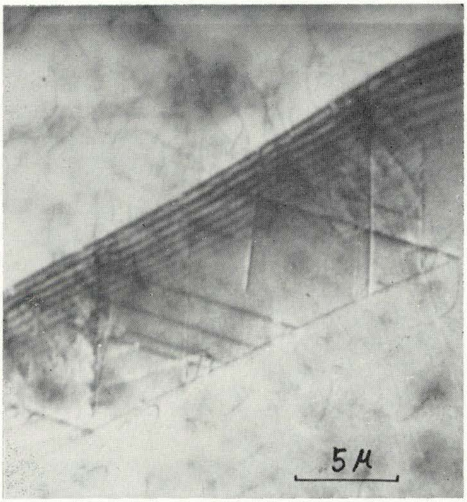

Fig. 7. Grain boundary in stainless steel. (300 kV)

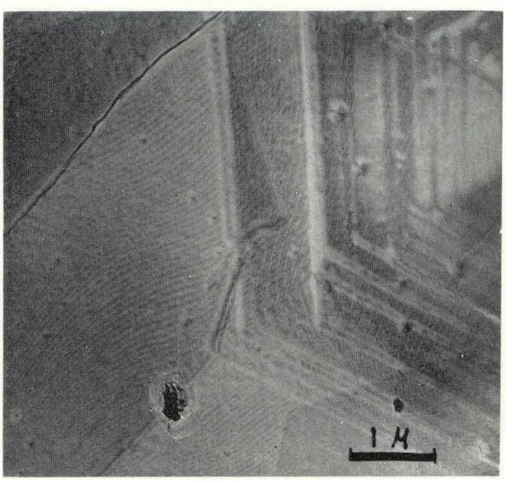

Fig. 8. Polyoxymethylene. (250 kV)

7. Acknowledgment

The authors would like to express their sincere thanks to Messrs. Fujita, Iwasa and Marukawa for preparing the specimens and to Prof. Uyeda, Drs. Kohra and Watanabe for their helpful discussions. The authors would also like to acknowledge their thanks to Dr. Fujiwara and Prof. Miyake for sending their theoretical conclusion and to Dr. Whelan for his helpful discussions based on the theory developed by his group.

References

1 K. Fujiwara: Read at the meeting of Phys. Soc. Japan 31st March, 1961.
2 A. Howie and M. J. Whelan: Private communication 15th Aug., 1961.
3 K. Kohra and H. Watanabe: J. Phys. Soc. Japan **16** (1961) 580. H. Hashimoto, A. Howie and M. J. Whelan: Phil. Mag. **5** (1960) 967.
4 Y. Kamiya and R. Uyeda: J. Phys. Soc. Japan **16** (1961) 1361.
5 S. Miyake: Private discussion at the meeting of Phys. Soc. Japan 31st March, 1961.
6 S. Miyake, K. Fujiwara and K. Suzuki: In this Volume, p. 124.
7 P. B. Hirsch, A. Howie and M. J. Whelan: Private communication 15th Aug., 1961.

DISCUSSION

P. B. HIRSCH: Is it possible that the dislocations you referred to as lying in the boundary are actually lying at the surface? The contrast is typical of dislocations at the surface.

H. HASHIMOTO: The dislocation contrast which I showed has special character. This contrast is observed superposed on the thickness fringe, and is very weak at the portion where the thickness fringe is observed, and very strong at the portion where the contrast of the thickness fringe becomes very weak. Furthermore, this contrast changes considerably by small variation of the orientation. I think such a kind of character can not be observed in the contrast of a dislocation at the surface.