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Measurement of Mean and Anomalous Absorption Coefficients of Electrons in MgO Crystals by the Use of Electron Micrographic Images

H. WATANABE AND A. FUKUHARA
Hitachi Central Research Laboratory, Kokubunji
Tokyo, Japan

AND

K. KOHRA
Institute of Physics, College of General Education
University of Tokyo, Komaba
Tokyo, Japan

The intensity distribution of the equal thickness fringe in dark field images of MgO crystallites obtained by using the (200) reflection is measured and analysed on the basis of dynamical theory taking account of absorption by assuming the imaginary potential. Some inconsistent results of the analysis and large fluctautions in observed values seem to be attributed mainly to the effect of inelastic scattering. The observation on extra fringe pattern which appears outside the crystal image by chromatic aberration supports this view.

1. It has been now generally accepted that anomalous absorption phenomena take place in the case ofelectr on diffraction too, in a similar way to the case of X-ray diffraction. Difference in absorption coefficients of two wave fields was observed for double diffraction spots from MgO crystallites[1], and anomalous absorption effect was observed in the so-called Hillier diffraction pattern of thin films of molybdenite crystal.[2] More recently, quantitative measurements of absorption coefficients have been made using electron micrographs from thin films of Al or stainless steel[3] and MgO crystallites.[4] On the other hand, Yoshioka[5] formulated that the effect of absorption can be theoretically dealt with by assuming an imaginary potential.

In the previous report on measurement for MgO crystals, the bright field image was used. In this report the dark field image is used and the results are discussed. It is pointed out that the simple theory taking account of absorption often fails in analysing the observed intensity curve and that the main reason will be the effect of inelastically scattered electrons.

2. The procedure of obtaining the absorption coefficients is in principle the same for both bright and dark field images. An example of photographs of dark field image is shown in Fig. 1, where the diffrated beam from the net plane (200) is used. At first the intensity is measured along the line normal to the fringe. Next, the intensity curve

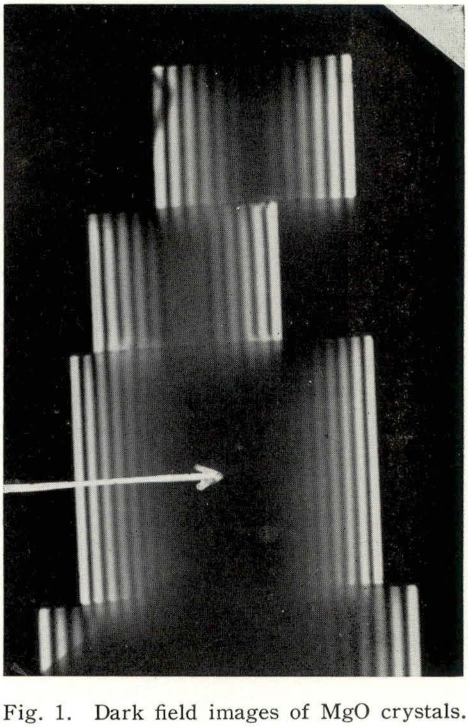

Fig. 1. Dark field images of MgO crystals.

is analysed using the dynamical theory taking account of the imaginary potential.

Bright and dark field images are formed by transmitted and diffracted electrons, respectively. The intensities are given by the dynamical theory as follows in the analogous way to the case of X-ray diffraction:

$$I_t=\frac{I_0}{4}\left\{\frac{2\cos(\sqrt{1+W^2}\,|v_h|\,\lambda t/2\pi)}{1+W^2}e^{-\mu_0 t}+\left(1+\frac{W}{\sqrt{1+W^2}}\right)^2 e^{-(\mu_0-\Delta\mu)t}+\left(1-\frac{W}{\sqrt{1+W^2}}\right)^2 e^{-(\mu_0+\Delta\mu)t}\right\}, \quad (1)$$

and

$$I_h=\frac{I_0}{4}\left\{-\frac{2\cos(\sqrt{1+W^2}\,|v_h|\,\lambda t/2\pi)}{1+W^2}e^{-\mu_0 t}+\frac{1}{1+W^2}e^{-(\mu_0-\Delta\mu)t}+\frac{1}{1+W^2}e^{-(\mu_0+\Delta\mu)t}\right\}, \quad (2)$$

with

$$\mu_0=\frac{v_{0i}\lambda}{2\pi},\ \Delta\mu=\frac{v_h}{|v_h|}\cdot\frac{v_{hi}\lambda}{2\pi\sqrt{1+W^2}} \quad (3)$$

where the crystal is assumed to have the center of symmetry, and I_0 is the intensity of the incident wave, t the thickness of specimen, W a parameter indicating the deviation from the Laue-Bragg condition, v_h the h-th Fourier coefficient of the crystal potential, and v_{0i} and v_{hi} the 0-th and h-th coefficients of the imaginary potential, respectively.

It must be noticed here that the expressions (1) and (2) are concerned only with elastically scattered electrons. If the contribution of inelastic scatterring is negligible, we can obtain the values of the absorption coefficients by analysing the observed intensity curve using eqs. (1) and (2).

The intensity curve of a dark field image expected from the theoretical formula, (2), is shown schematically in Fig. 2. Abscissa shows the thickness of the crystal. The fringe pattern, a, corresponds to the first term, and the background, b, is composed of the second and third terms. The thickness is given as the function of the distance from the edge of the image and the inclination of the crystal, the latter of which is determined from the external shape of the image. W is determined from the spacing of the fringes. To find μ_0 and $\Delta\mu$, curves a and b are plotted on a logarithmic scale as shown in Fig. 3. From the theory the followings are expected: (i) the curve a is a straight line, from the slope of which μ_0 is determined; (ii) the curve b

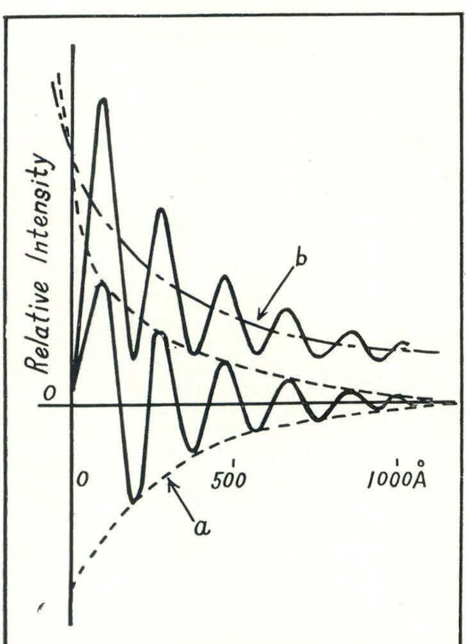

Fig. 2. Schematic intensity curve expected from the theory.

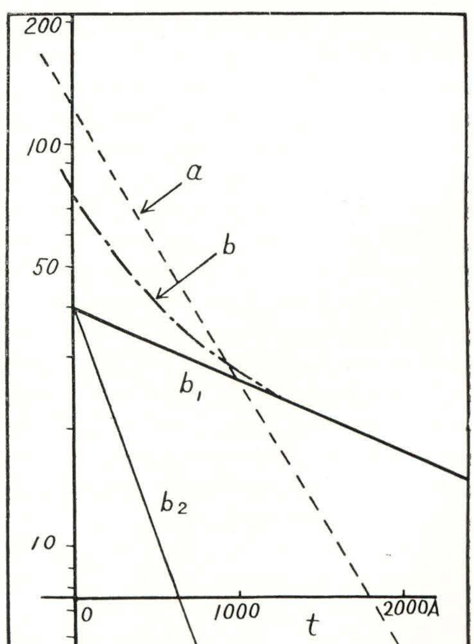

Fig. 3. Plot of the intensity curves on a logarithmic scale.

Table I.

Acc. Volt. (kv)	Period (Å)	μ_0^{-1} (Å)	v_{0i} (ev)	$(\mu_0-\Delta\mu)^{-1}$ (Å)	$(\mu_0+\Delta\mu)^{-1}$ (Å)	v_{hi} (ev)	$\sqrt{1+W^2}$
50	353	382	1.27 v				1.20
	328	405	1.20				1.29
75	368	592	1.03				1.42
	368	585	1.04				1.42
100	462	781	0.85	2860	453	0.81	1.30
	457	746	0.89	2700	437	0.84	1.31

To estimate the value of $\sqrt{1+W^2}$ from the period of the fringe, use is made of the observed value $v_h=7.0$ ev by Honjo and Mihama[1].

is composed of two straight lines b_1 and b_2, corresponding to the second and third terms in eq. (2), respectively, from the slopes of which $\mu_0-\Delta\mu$ and $\mu_0+\Delta\mu$ are determined, respectively. As additional condition, (iii) both b_1 and b_2 start from a point on y-axis, the ordinate of which is a quarter of the one of the cross point of the line a and y-axis.

In the bright field image the condition corresponding to (iii) is different from that in the above case. The ordinates of the cross points of curve a, b_1 and b_2 with y-axis are given by $\frac{1}{2}(1+W^2)$, $\frac{1}{4}\{1+(W/(1+W^2))\}^2$ and $\frac{1}{4}\{1-(W/(1+W^2))\}^2$, respectively. Their relation is not so simple as in the above case. However, not only the magnitude of W but also its sign could be determined from this relation independently of the measurement of the spacing of the fringe, although this situation makes, in practice, the analysis of the curves more difficult or ambiguous.

3. Observed intensity curves, however, do not always behave in the way expected from the theory, namely, all the conditions mentioned above are not always satisfied.

First of all, the curve b is sometimes nearly straight, and sometimes is curved even in the opposite direction to the one predicted by the theory. As seen from the condition (i) and (ii), the values μ_0 and $\Delta\mu$ may be determined from slopes of the three curves, but consistent values are not always obtained. Even if the analysis is made rather well, pretty large fluctuations are found among observed values.

In Table I values picked up from many observations are given. The reciprocal of mean absorption coefficient, μ_0^{-1}, corresponds to mean free path of inelastic scattering, and the reciprocals of other two absorption coefficients, $(\mu_0-\Delta\mu)^{-1}$ and $(\mu_0+\Delta\mu)^{-1}$, correspond to mean free paths of two wave fields, respectively. For 50 and 75 kv the values of anomalous absorption coefficients are not given because the experimental curves corresponding to b are mostly nearly straight. As noticed above, we must be careful in making detailed discussions on the obtained values, but we can know the order of the absorption coefficients for the electron micrographs.

4. As causes for the contradictions between the theory and observations, the followings are to be considered: (i) effect of inelastic scattering, (ii) effect of weak reflections, (iii) deviation from the rigorous cubic shape, and (iv) imperfection of the crystal.

In the present experiment, (i) seems to be the most important. Now let us consider the effect of inelastic scattering a little. In Fig. 1 it may be noticed, as Kamiya suggested to

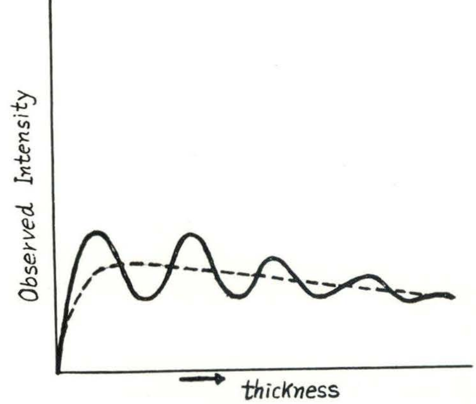

Fig. 4. Observed intensity curve of an outside fringe pattern.

us[6], that an additional, extra fringe pattern appears outside of the lower edge of each crystal image. This is due to chromatic aberration, in other words, due to inelastic scattering. An example of the intensity curve of the additional fringe pattern of the outside is given in Fig. 4. It is to be noted that the inelastically scattered electrons contribute also to the formation of the fringe pattern as Kamiya and Uyeda pointed out.[7] Let us notice, next, the behaviour of the background. It increases with thickness, attains a maximum and then decreases. This tendency is a strong contrast to that expected from the theory. The behaviour of observed curve *a* contradicting the theory will be probably or mainly due to this situation. According to Fujimoto and Kainuma,[8] the intensity distribution of inelastic scattering depends on W and minimum for $W=0$. Variety in behaviour of observed curves *b* or large fluctuation of observed values of the absorption coefficients might be explained by this fact. Although the effect of inelastic scattering varies with W, this will make smaller the magnitudes of the observed absorption coefficients than those expected from the theory used here.

The second factor, namely, the effect of weak reflections will change sensitively with different orientation of the crystal, and will give a complicated effect on the observed value.

If we make observations for a certain value of W, for example, for $W=0$, using a special specimen holder, we can expect less fluctuation in observed values of the absorption coefficients, but the effect of inelastic scattering will remain to exist. For comparison of the experiment with the theory, it is necessary either to remove the inelastic part in the experiment, or to take account of the inelastic part in the theory.

The authors should like to express their thankfulness to Prof Uyeda, Dr. Kamiya and Prof. Fujimoto for their valuable discussions.

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DISCUSSION

K. MOLIÈRE: 1) I think that the consideration of inelastic scattering in your experiment is very important. But I should like to emphasize your second point (*weak* interactions) too. In the case of 200 reflection of Mg0, we have to consider the systematic interactions $\bar{2}00$ and 400 which are pretty strong. It would therefore be better to perform experiments for the case of 220 reflection.

2) How large is the experimental error in your measurement of the absorption coefficients?

K. KOHRA: As to your question (1), I agree with your opinion, and we have also measured the absorption coefficients for 220 reflection, using the bright field image (reported at the Meeting of Phys. Soc. Japan at Osaka in October of 1960). In the case of 220 reflection, however, it is not possible to see the effect of inelastic scattering through chromatic aberration.

H. WATANABE: The experimental error in our intensity measurement is less than 5%. The linearity of the photographic plate was calibrated by the double exposure method.

K. KOHRA: In our measurement there are still some sources of the error, for example, the period of the fringe, excitation error and thickness. These quantities are estimated by assuming that the shape of the crystal is cubic, but this will not always hold. The determination of the coefficients from the intensity curve plotted on the logarithmic scale may also be a source of errors.

N. KATO: Is there any possibility that the effect of defocussing results in different behaviours of the intensity distribution you mentioned?

H. WATANABE: The Fresnel fringe appears in the neighbourhood of the edge when the image is defocussed, which makes the intensity distribution different. In our measurement, therefore, only well focussed images were used.

G. HONJO: It seems that the method of Professor Molière is superior to your method in determining the absorption coefficients.

K. KOHRA: I admit that their experiment may be more accurate and reproducible. I would like to notice, however, that our measurement is not concerened with the electron diffraction image but with the electron micrographic image. I think the study on the electron micrograph is also important especially in relation to inelastic scattering, as pointed out in this report.

H. HASHIMOTO: I saw in your electron micrograph that the number of visible fringes is different even in one crystal, that is, the number of visible fringes is larger in the lower part of the crystal than in the upper part. This difference will give a different absorption coefficient. Which part of the micrograph did you measure?

K. KOHRA: We have measured the intensity at the middle region where the number of visible fringes is nearly constant. The fact you have pointed out is related to the extra fringe which is observed outside of the lower edge as mentioned in the report. The inelastically scattered electrons shift downwards because of chromatic aberration. It is conceived that the image at the upper edge is made of only elastically scattered electrons and the fringe image in outside is made of inelastically scattered ones, while the image at the middle part is made of both electrons scattered elastically and inelastically. Measurements at various positions have been done very recently by Kamiya, Uyeda and us, which will be reported elsewhere.