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

Variation of Electron Diffraction Intensities with Tilting Angle and λH

SHIGEYA KUWABARA

Department of Physics, Faculty of Science
Hiroshima University, Hiroshima, Japan

The variations of electron diffraction intensities of thin Au, NaCl and BiOCl polycrystalline films with more or less preferred orientation were examined with change of the tilting angle around axes lying in film layers, crystal size of the specimens and wave-length, and the results were compared with those of the two-wave and many-wave theories.

In many of the electron diffraction investigations the dynamical effect cannot be neglected because of strong interactions between electrons and matter. According to Blackman's theory[1], which is based on the two-wave theory, the kinematical values of diffraction intensities are expected when λH tends to zero, where λ is the wavelength of the electrons and H the thickness of the single crystals. For several metals or compounds containing light atoms, Blackman's theory was verified[2,3]. Since then, such a dynamical effect (primary extinction effect) has been corrected by the extrapolation method[3] or the statistical method[4] for the evaluation of kinematical values in the structure analyses by electron diffraction.

However, Miyake[5] reported recently that if the higher order approximation is used, the two-wave theory does not give kinematical values for the limiting case of $\lambda H \to 0$, which contradicts Blackman's theory of lower approximation. To elucidate such a contradiction he pointed out that many-wave theory should be used for the crystals which are currently used in electron diffraction structure analyses. Fujiwara[6] and Fujimoto[7] developed the many-wave theory and both of them got the same result which was obtained by Cowley and Moodie[8,9] by their physical optics theory.

If the many-wave theories are used it is expected that the intensities show the kinematical values for $\lambda H \to 0$ and moreover they change when the crystals are tilted around axes lying in the film layers even if they are measured along the axes of tilting, because the contribution of weak waves changes with the tilting angle.

It has already been verified that the diffraction intensities change with the tilting angle for BiOCl polycrystalline film at constant accelerating voltages[10]. It was also shown by calculation that if the thin crystals are composed of one kind of atom or several light atoms, where the effect of phase-shift can be neglected in the scattering process, the intensity variation with λH will show the same tendency as Blackman's theory, but if they are composed of both heavy and light atoms it will give quite different tendency. The aim of the present experiment is to examine these theoretical predictions.

A vibrating type d.c. amplifier was used to measure the intensities. The evaporated Au and NaCl polycrystaline-films and precipitated BiOCl polycrystalline-films were used as the specimens. The diffracted intensities along the axes of tilting were measured for various values of tilting and λH, and the results were compared with the two-wave and many-wave theories.

The results are shown in Figs. 1, 2 and 3, for Au, NaCl and BiOCl, respectively. The relative intensities are given in the ordinates (the intensity values of (220) reflections were normalized to 100 for Au and NaCl and to 10 for BiOCl), and the λH in the abscissas. The circles are the experimental plots and the vertical line segments mean the error of the intensity measurement. The full lines in Figs. 1 and 2 and the chain lines in Fig. 3 are the curves calculated from Blackman's theory with the temperature factors $B=0.6$, 1.5 and 1.0Å^2 for Au, NaCl and BiOCl, respectively.

Specimens with no preferred orientation were used for Au. In Fig. 1, the black circles correspond to the crystal size 35Å (film thick-

ness 15Å) and the open ones 42Å (60Å). Generally the experimental values agree with the curves very well.

The results for NaCl films are shown in Fig. 2. The black circles correspond to the crystal size 68Å (film thickness 60Å) with very poor preferred orientation, for which the intensity values were averaged out for 0, 30 and 60°. The open circles correspond to the crystal size 100Å (600Å). The values for the tilting angles 0, 30 and 60° are plotted in the vertical line for each λH value. The highest values are for 30° and the lowest ones for 60°. When all the values for each tilting angle are averaged out, the variation of the intensities shows the same tendency as Blackman's formula. However, the variation of the intensities with tilting angle at the same λH values cannot be explained by this theory.

Fig. 3 shows the results for BiOCl films with zero tilting angle. The intensity variation with the tilting angle at the fixed accelerating voltage gave the same result as before[10]. The black circles in the figure are for the crystal size 62Å and the open ones

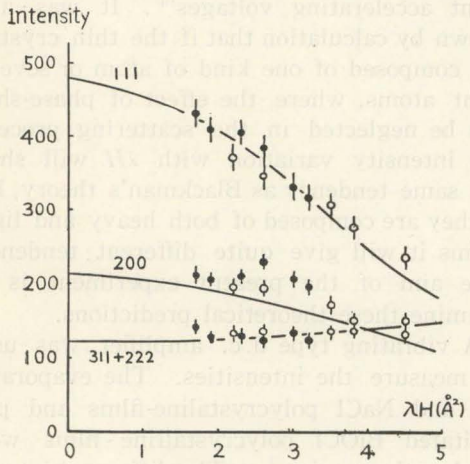

Fig. 1. Au.

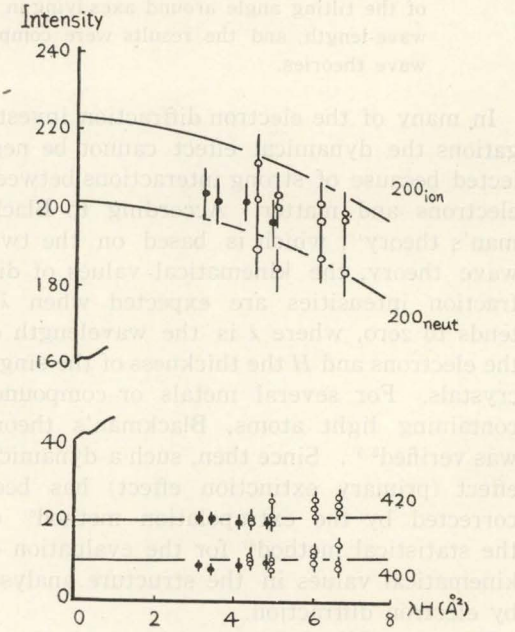

Fig. 2. NaCl.

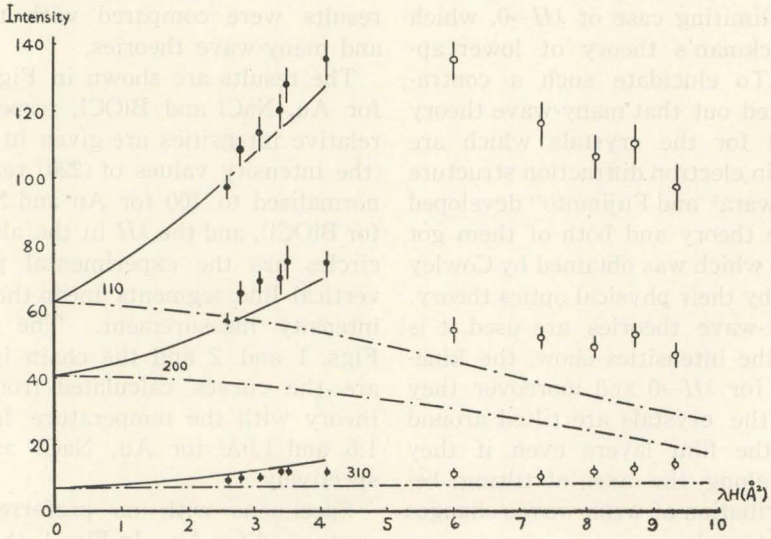

Fig. 3. BiOCl.

for 160Å. The variation of the diffraction intensities with λH for thinner crystal (62Å) shows quite different tendency from the curves of Blackman's theory. However, it agrees with the curves calculated from the phase-grating theory[9] (full lines in this figure). The intensity variations of open circles is similar to Blackman's formula in trend but different in magnitude. For the thicker single crystals or polycrystals with large crystal size the two-wave theory will, in principle, become a better approximation. The discrepancy in magnitude may partly explained if the phase-shift in the scattering process is taken into account[11]. However, the higher order approximation of many-wave theories is necessary to explain the intensity variation for such a thick crystal quantitatively[12].

In conclusion, though Blackman's formula based on the two-wave theory gives an explanation of the intensity variations with λH for the crystals containing only one kind of atom or several light atoms, it was only fortunate, and the many-wave theories should be used for thin crystals especially when they are composed of both heavy and light atoms.

References

1 M. Blackman: Proc. Roy. Soc. **173** (1939) 68.
2 S. Kuwabara: J. Phys. Soc. Japan **10** (1955) 416; **12** (1957) 637.
3 G. Honjo and N. Kitamura: Acta Cryst. **10** (1957) 533.
4 S. Nagakura: Acta Cryst. **10** (1957) 601.
5 S. Miyake: J. Phys. Soc. Japan **14** (1959) 1347.
6 K. Fujiwara: J. Phys. Soc. Japan **14** (1959) 1513.
7 F. Fujimoto: J. Phys. Soc. Japan **14** (1959) 1558: **15** (1960) 859.
8 J. M. Cowley and A. F. Moodie: Acta Cryst. **10** (1957) 609.
9 J. M. Cowley and A. F. Moodie: Acta Cryst. **12** (1959) 360.
10 J. M. Cowley and S. Kuwabara: Acta Cryst. In Press.
11 J. A. Hoerni: Phys. Rev. **102** (1956) 1534.
12 J. M. Cowley: Private communication.

DISCUSSION

F. FUJIMOTO: In BiOCl, it may not be suitable to take the 220-reflection as standard, because the 220-reflection is a higher order reflection of the strong 110-reflection. In the case of higher order reflection, the effect of other reflections is generally large and the two-wave approximation is not applicable according to my results, which were obtained by the intensity calculation only at the Bragg position. If you take a higher order reflection as standard, we cannot say whether the discrepancy between the values obtained from two-wave approximation and your experiment comes from the circumstance that the crystal includes heavy atoms, or not. We should take a high index reflection, such as 310, as standard.

S. KUWABARA: I can agree with your opinion.

M.J. WHELAN: Has any allowance been made in the interpretation of your results for the effects of absorption? Absorption must have some important effect on integrated intensities particularly in gold, and probably also in BiOCl since it contains heavy atoms. You could obtain some idea of the importance of absorption simply by looking at your specimens in an electron microscope.

S. KUWABARA: Although I did not pay any attention to the absorption effect, it may be involved in the present experiment. In fact, Prof. Honjo *et al.* found a deviation of the experimental plots from the theoretical curves at lower accelerating voltage, when they studied the correction of the primary extinction effect.