Short Communications

New methods of determining Fourier coefficients of crystal potential from thickness fringes in electron micrographs. By AYAHIKO ICHIMIYA, TATSUO ARII and RYOZI UYEDA, *Department of Applied Physics, Faculty of Engineering, Nagoya University, Nagoya, Japan* and AKIRA FUKUHARA, *Hitachi Central Research Laboratory, Kokubunji, Tokyo, Japan*

Two methods are described for the determination of Fourier coefficients V_h and V_{2h} with a high accuracy. In the first method, the fringe distances are at (1) the first-order Bragg position, (2) the second-order Bragg position and (3) the symmetric position. A measurement for magnesium oxide gave $V_{111} = 1\cdot78 \pm 0\cdot05$ and $V_{222} = 3\cdot90 \pm 0\cdot10$ volt. In the second method, which is proposed for the case where the second-order fringes are too broad for an accurate measurement, the same is done from fringe distances at (1) and (3) measured at least at two different accelerating voltages. A 15-beam calculation has shown that this method enables us to obtain highly accurate values, provided that the measurement is done at a conventional voltage around 100 kV and an extremely high voltage over 1000 kV.

Several methods are known for the determination of Fourier coefficients of crystal potential by the use of dynamical effects in electron diffraction (Hibi, Kambe & Honjo, 1955; Goodman & Lehmpfuhl, 1967; Watanabe, Uyeda & Fukuhara, 1968; Cowley, 1969; Gjønnes & Høier, 1971; Kreutle & Meyer-Ehmsen, 1971; Lehmpfuhl, 1972). For example, the use of thickness fringes in electron micrographs has long been known (Hibi, Kambe & Honjo, 1955). However, a high accuracy has not been attained with this method because of the difficulty in calibrating the magnification of the electron microscope with an accuracy better than a few percent.

In the first of the present methods, micrographs of thickness fringes are taken at the following three positions: (1)
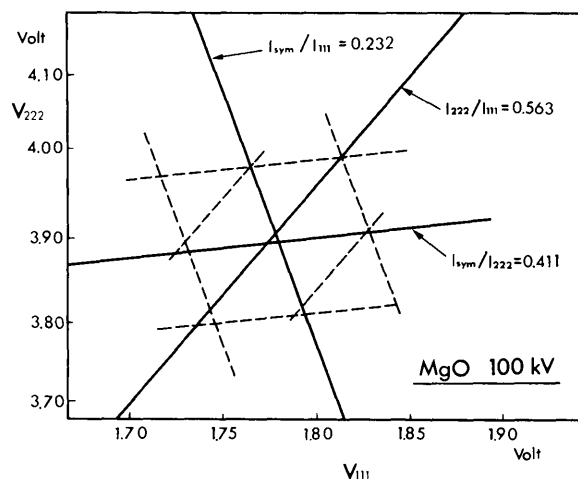

Fig. 1. Determination of V_{111} and V_{222} of magnesium oxide by the first method. Three curves are drawn with experimental values of ratios I_{sym}/I_{111}, I_{sym}/I_{222} and I_{222}/I_{111}. A pair of broken lines is drawn corresponding to the errors of each ratio. The hexagon bounded by the three pairs may give an estimate of the errors in V_{111} and V_{222}.
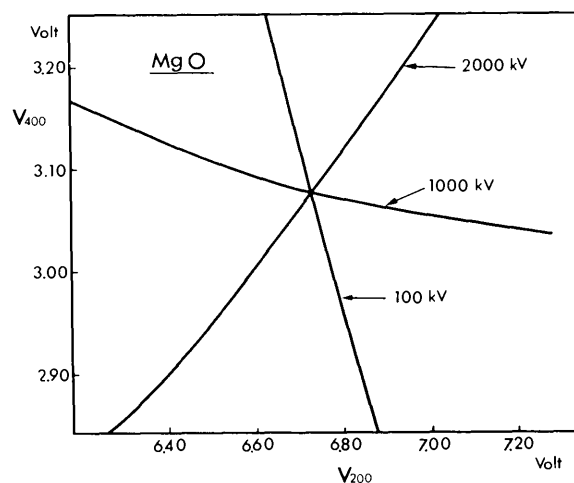

Fig. 2. Determination of V_{200} and V_{400} of magnesium oxide by the second method. No experimental value is yet available. The three curves show a result of 15-beam calculation.

The exact Bragg position of the *h*th order, (2) that of the 2*h*th order and (3) the symmetric position for the *h*th plane. It should be noted that distinct fringes are always obtained at position (3). The fringe distances I_h, I_{2h} and I_{sym} are measured for (1), (2) and (3) respectively. The ratios I_{sym}/I_h, I_{sym}/I_{2h} and I_{2h}/I_h are calculated. It is necessary that the three micrographs should be taken at the same magnification, but it is not necessary to know this value. According to the dynamical theory, each of the ratios is a function of Fourier coefficients V_h, V_{2h}, V_{3h}, *etc.* and the accelerating voltage E, provided that the crystal is so oriented that no accidental interaction takes place. In practice, however, each ratio is essentially a function of three parameters, V_h, V_{2h} and E, because V_{3h} and V's of higher order are known from other data and moreover, their small variation has no significant effect on the values of the ratios. Thus,

provided that E is known, V_h and V_{2h} can be calculated from the experimental values of the three ratios.

Fig. 1 gives an experimental result for magnesium oxide. The abscissa is V_{111} and the ordinate, V_{222}. The accelerating voltage is 100 kV. From each experimental value of the ratios, a curve is drawn on the diagram. The values of V_{111} and V_{222} are determined from the intersection of the curves. The error can also be estimated as shown by broken lines in the Figure. The values obtained were $V_{111} = 1\cdot78 \pm 0\cdot05$ and $V_{222} = 3\cdot90 \pm 0\cdot10$ volt. The former value is in good accord with that of Lehmpfuhl (1972).

In many cases, the fringe distance is too large at the second-order position for an accurate value of I_{2h} to be determined. The second method is proposed for such a case In this method, no fringes at the second order are required, while those at the first-order position and the symmetric position must be taken at least at two different accelerating voltages. Then, V_h and V_{2h} can be calculated from the ratios I_{sym}/I_h at these voltages. No experiment has yet been carried out with this method. A calculation was done to estimate the accelerating voltages adequate for the measurement.

Fig. 2 shows a result obtained by assuming the values of I_{sym}/I_{200} at 100, 1000 and 2000 kV. The three curves intersect at approximately 60'. This means that a high-accuracy measurement will be possible if the measurement is done at a conventional voltage around 100 kV and at least at an extremely high voltage over 1000 kV.

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A note on the higher-moment test for space-group determination.
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Theoretical expressions for the second, third and fourth moments of normalized intensity z and the fourth moment of the intensity (I) scattered by an asymmetric unit are given for crystals in which all atoms are in general positions. The expression for the fourth moment of I applicable to crystals containing atoms in both general and different types of special position is also given.

Foster & Hargreaves (1963b) have given in Table 1 of their paper the expressions for the first three moments of the intensity (I) scattered by the asymmetric unit and these results are applicable to crystals (containing atoms at general positions in the unit cell) belonging to all but two of the 74 space groups and the nine related plane groups in the triclinic, monoclinic and orthorhombic systems. They have also shown how these results could be used to derive the theoretical expressions for the second and third moments of the normalized intensity (z) when the crystal contains atoms in both general and special positions. In this note we shall list the expressions for the fourth moment of the intensity (I), since in some cases the tests based on the second and third moments of z may not be very effective. For example, for crystals containing a few (*i.e.* one or two) dominating heavy atoms besides a large number of light atoms, it may be useful to employ the fourth moment of z. This is clear from Table 1 (computed from the results of Parthasarathy, 1966) which lists the higher moments of z for crystals containing one or two dominating atoms besides a large number of light atoms (for brevity referred to as the one-atom case and two-atom case respectively) in the space groups $P1$ and $P\bar{1}$ in terms of the parameter σ_1^2 (which is the fractional contribution to the local mean intensity from the heavy atoms in the unit cell.)

Another purpose of this note is to list the *explicit expressions* for the second, third and fourth moments of the normalized intensity z applicable for crystals containing all atoms at general positions in the unit cell, since such crystals are of frequent occurence. As the expressions for $\langle I^4 \rangle$ for crystals containing atoms in both general and special positions and for $\langle z^n \rangle$, $n = 2$, 3 and 4, for crystals containing all atoms in general positions could be derived from the theoretical results of Foster & Hargreaves (1963a) we shall give only the final results, omitting all the intermediate steps. The notation followed in this paper is the same as that used by Foster & Hargreaves (1963a, b).

Table 1. *Higher moments of z for the one-atom and two-atom cases when the heavy-atom contributions are* $0\cdot7$, $0\cdot8$ *and* $0\cdot9$

The tabulated values have been calculated from the results of Parthasarathy (1966). Note the inefficiency of $\langle z^2 \rangle$ and the distinction of $\langle z^4 \rangle$ in all the cases.

σ_1^2	Space group	One-atom case $\langle z^2 \rangle$	$\langle z^3 \rangle$	$\langle z^4 \rangle$	Two-atom case $\langle z^2 \rangle$	$\langle z^3 \rangle$	$\langle z^4 \rangle$
0·7	$P1$	1·51	2·96	7·07	1·76	4·14	11·94
	$P\bar{1}$	2·02	5·79	21·17	2·27	7·40	30·93
0·8	$P1$	1·36	2·29	4·55	1·68	3·63	9·31
	$P\bar{1}$	1·72	3·99	11·51	2·04	5·72	19·89
0·9	$P1$	1·19	1·63	2·50	1·60	3·08	6·75
	$P\bar{1}$	1·38	2·37	4·79	1·79	4·07	10·92

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