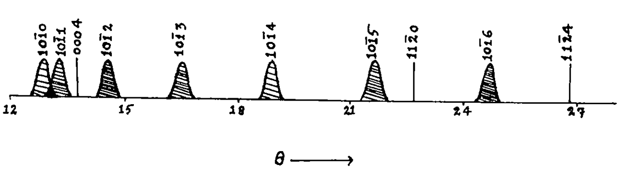

Fig. 4. Schematic powder pattern for faulted crystals ($\alpha = 0{\cdot}1$, $0{\cdot}9$).

The reciprocal lattice and the powder pattern (schematic) for deformation-faulted d.h.c.p. crystals are illustrated in Figs. 3 and 4. The nature of the powder pattern suggests a possible method of determining α in practice. Consider the pair of reflexions 0004 and $10\bar{1}2$, the first of which is unaffected by faulting, while the second is affected. If we assume that the broadening of the two reflexions by all factors other than faulting is equal, then the broadening resulting from these other factors can be removed from the total broadening of the $10\bar{1}2$ reflexion by the method due to Stokes (1948), yielding the fault broadening and thus α. Alternatively, α can be evaluated, although not so accurately, from a comparison of the ratios of the integrated intensities of reflexions with different values of L (Table 2).

The rare earth metals lanthanum, cerium (below −10 °C), praseodymium, neodymium and americium are known to exhibit the d.h.c.p. structure (Barrett & Massalski, 1966) besides many intermetallic compounds. Interesting results may be expected from X-ray diffraction studies of these metals and alloys after deformation.

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The Effects of *n*-Beam Dynamical Diffraction on Electron Diffraction Intensities from Polycrystalline Materials

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Calculations based on *n*-beam dynamical diffraction theory have shown that the intensities of rings and arcs in diffraction patterns from polycrystalline materials are strongly dependent on the experimental conditions. For arc patterns from BiOCl, calculations confirm a revised formula for the intensities from very thin crystals and indicate dependences on the distributions of orientation, thickness and bending of the crystals which are sufficient to account for the wide deviations of recent experimental results from the predictions of the primary extinction formula, based on the two-beam approximation. Inferences regarding the possible errors in electron diffraction analysis of crystal structures, especially for materials containing heavy atoms, are drawn from calculations relating to the intensities of arc patterns from $AgTlSe_2$. It is suggested that *n*-beam calculations should be made in order to avoid serious error in the refinement stages of a structure analysis.

Introduction

The presence of dynamical diffraction effects in the intensities of electron diffraction ring patterns from polycrystalline materials has been recognized for many years through the success of the primary extinction formula of Blackman (1939); see, *e.g.*, Kuwabara (1957). However, the use of accurate electronic recording equipment and of filters to remove inelastically scattered electrons has revealed significant discrepancies between experimental results and the theoretical values based on the assumption of two-beam dynamical conditions. Thus Horstmann & Meyer (1962) found that the 400 and 222 intensities in ring patterns from polycrystalline aluminum did not lie on the primary extinction curve, and Wedel (1963) reported similar, but larger discrepancies in the case of silver. Recently Kuwabara, Turner & Cowley (1966) and Kuwabara (1967) have shown that intensities in arc patterns from

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polycrystalline BiOCl differ substantially from the two-beam values. In the case of light elements such as aluminum, a partial account of the deviations is given by using the dynamic potentials of Bethe (1928), as described by Horstmann (1965). In BiOCl and similar substances dynamical effects are expected to be strong because of the presence of heavy atoms in addition to light ones (Cowley & Kuwabara, 1962).

In the first part of this paper it is shown that the discrepancies between experimental values and the primary extinction curve can be explained by including multiple-beam interactions in the calculations of intensities, as stated by Kuwabara *et al.* (1966). A consequence of the dynamical nature of the electron diffraction process is the great sensitivity of the intensities to the characteristics of the specimen, and of the diffraction camera.

The failures of the simple kinematical and two-beam dynamical formulae have contributed to the widely held doubts about the usefulness of electron diffraction for the analysis of crystal structures. In a recent review, Cowley (1967) has examined the conditions under which the standard methods of structure analysis by electron diffraction from polycrystalline materials, as described by Vainshtein (1964), may be expected to be in error because of the neglect of n-beam effects. In particular, such effects cannot be ignored in the commonly occurring cases when at least one constituent of the material is of high atomic number. In the second section of this paper an estimate of these errors has been made for a typical case, and they are found to be important. A modification of the usual process of structure analysis refinement is suggested to enable such errors to be reduced.

Dynamical intensities from polycrystalline specimens

The problem of evaluating intensities from polycrystalline specimens is complicated by the need to take into account the variations in the thickness and orientation of the many crystals, within the illuminated area of the specimen, which contribute to the diffracted intensity. The specimens of greatest interest and use are those for which a preferred orientation exists so that, on tilting the supporting film, a textured diffraction pattern is obtained (Vainshtein, 1964). The recorded intensity is then a sum of contributions from each crystal, and can be computed by first finding the intensity of a reflexion from a single crystal as a function of crystal thickness and orientation and then summing intensities weighted by distribution functions chosen to represent the distributions of crystal thickness and orientation actually present in the sample.

In a specimen having a preferred orientation, one particular axis of all the crystals lies on, or close to, the 'texture axis' of the specimen. In the following we consider the example of BiOCl which crystallizes in platelets with their c axes perpendicular to the support film.

The variations in crystal thickness and orientation, which have a strong influence on the diffracted amplitudes, may be important within an individual crystal as well as between crystals, since crystals may be bent and have irregular shape. The question then arises as to whether it is the amplitudes or intensities of diffracted beams which should be averaged over the range of these variables. This will depend on the rates of their variation relative to the range of the lateral coherence of the electron beam at the specimen position, which is normally a few hundred Å but may be increased in high-resolution instruments to as much as 1000 Å (Cowley, 1961). For many specimens the close spacing of extinction contours in electron micrographs suggests that some crystals may be bent through appreciable angles within a distance of a few hundred Å.

While for most specimens it would seem to be appropriate to average incoherently over intensities, the possibility that averaging over amplitudes may be necessary for particular cases must be kept in mind.

The effects of crystal defects other than bending are not considered in the present work. It is assumed that they may be included in the functions describing the distribution of thickness and orientation within the specimen.

Integrated $hk0$ systematic intensities for BiOCl

The 2-beam approximation is based on the assumption that the crystal can be orientated so that only one beam other than the transmitted beam diffracts with appreciable magnitude. It is now generally accepted that this is rarely possible, and that the systematic interactions (Hoerni, 1956) are always significant. On the other hand, it is usually possible to avoid strong interactions with the non-systematic reflexions, provided that the incident beam is tilted well away from any principal direction in the crystal. This argument is the basis of our assumption of systematic 'one-dimensional' diffraction conditions for all crystals within a textured specimen which has been tilted to give an oblique-texture diffraction pattern.

According to Vainshtein (1964), the intensity J_{hk0}, recorded by traversing a microdensitometer slit of length Δ across an $hk0$ arc in a recording of an oblique-texture pattern, is given by

$$J_{hk0} = Cp_{hk0}\Delta J'_{hk0}\,, \tag{1}$$

where C is a scale factor, constant for all reflexions, p_{hk0} is the multiplicity, d_{hk0} the spacing of the $hk0$ planes, and

$$J'_{hk0} = \iint I_{hk0}T(\beta)H(D)d\beta dD\,.$$

Here I_{hk0} is the $hk0$ intensity, a function of tilt β in the hkl plane, and of crystal thickness D; $T(\beta)$ and $H(D)$ are functions representing the distributions of angle and thickness in the specimen.

We consider in detail the case where all crystals are of equal thickness, and have a gaussian distribution of width α about the texture axis; *i.e.*

$$T(\beta) = \exp(-\beta^2/\alpha^2)\,,$$
$$H(D) = \delta(D)\,,$$

and

$$J'_{hkl} = \int I_{hk0} \exp(-\beta^2/\alpha^2) d\beta\,. \tag{2}$$

(See Fig. 1). It is usually assumed that the crystal thickness is sufficiently large to ensure that the angular width of the distribution $T(\beta)$ is large compared with that of I_{hk0} resulting from to the size effect, so that a constant value of $T(\beta)$ may be assumed for each crystal. The formula of Blackman (1939) is then obtained by substitution of the 2-beam form of I_{hk0} (see Vainshtein, 1964) in equation (2), so that

$$J_{\mathbf{h}} = C|V_{\mathbf{h}}|^2 D d_{\mathbf{h}} p_{\mathbf{h}} \Delta \mathscr{D}_{\mathbf{h}} \tag{3}$$

where $\mathscr{D}_{\mathbf{h}}$ is the primary extinction factor defined by

$$\mathscr{D}_{\mathbf{h}} = \frac{1}{\sigma V_{\mathbf{h}} D} \int_0^{\sigma V_{\mathbf{h}} D} J_0(2x) dx\,,$$

h has been used for $hk0$, $V_{\mathbf{h}}$ is the $hk0$ Fourier potential (in volts), $\sigma = 2\pi m e \lambda / h^2$ and $J_0(x)$ is the zero order Bessel function. When $\sigma V_{\mathbf{h}} D$ is small ($<0{\cdot}25$), $\mathscr{D}_{\mathbf{h}}$ is unity and equation (3) then reduces to the kinematical formula.

The assumption that $\exp(-\beta^2/\alpha^2)$ is constant may be reasonable even for inner reflexions of specimens containing thick crystals, since α is often large (Fig. 1). In the limit $D \to 0$ the equation (3) then gives the ratio of intensities of two reflexions **h**, **g** as

$$\frac{J_{\mathbf{h}}}{J_{\mathbf{g}}} = \frac{|V_{\mathbf{h}}|^2 d_{\mathbf{h}}^2 p_{\mathbf{h}}}{|V_{\mathbf{g}}|^2 d_{\mathbf{g}}^2 p_{\mathbf{g}}}\,. \tag{4}$$

However, the weighting factor cannot be neglected for thin crystals, since for thin crystals the regions of scattering power around the reciprocal lattice points are extended into long spikes, and $I_{\mathbf{h}}(\beta) \to I_{\mathbf{h}}(0)$ for all values of β for which $\exp(-\beta^2/\alpha^2)$ has appreciable amplitude. We then obtain from equation (2)

$$\frac{J_{\mathbf{h}}}{J_{\mathbf{g}}} = \frac{|V_{\mathbf{h}}|^2 d_{\mathbf{h}} p_{\mathbf{h}}}{|V_{\mathbf{g}}|^2 d_{\mathbf{g}} p_{\mathbf{g}}}\,. \tag{5}$$

For $d_{\mathbf{g}} < d_{\mathbf{h}}$ the intensity ratio should therefore be lower than would be expected on the basis of equation (4).

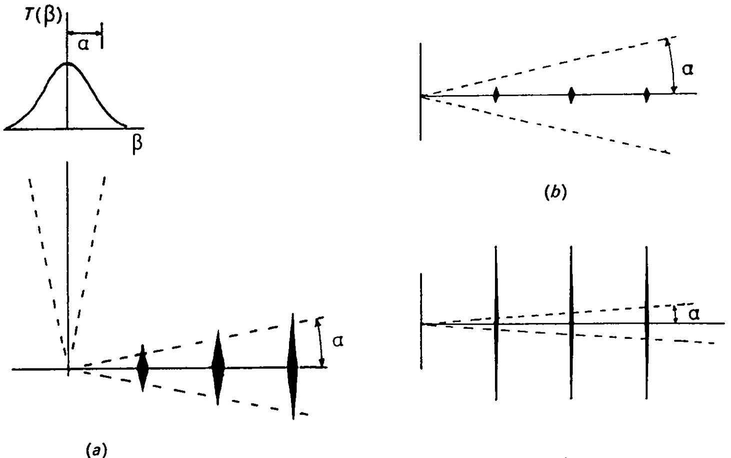
This is observed in the calculated results presented below. Intensities of ring patterns from very thin crystals of graphitic oxide measured by Aragon & Cowley (1963) were found to vary with $d_{\mathbf{h}}$ as in equation (5) rather than with $d_{\mathbf{h}}^2$ as in equation (4).

If the lateral coherence of the electron beam is appreciably larger than the regions over which crystal thickness or orientation change, equation (2) must be replaced by

$$J'_{\mathbf{h}} = \left| \int A_{\mathbf{h}}(\beta) \exp(-\beta^2/\alpha^2) d\beta \right|^2, \tag{2a}$$

where $A_{\mathbf{h}}(\beta)$ is the (complex) amplitude of the reflexion **h**.

In calculating n-beam dynamical intensities, the multi-slice numerical procedure developed by Goodman & Moodie (1965) (see also Cowley, 1967) from the dynamical theory of Cowley & Moodie (1957) was used. The matrix method of computation, developed by Howie & Whelan (1960) from the work of Neihrs & Wagner (1955), Fujimoto (1959) and Sturkey (1962) would also be suitable, although for the present case, where intensities for many tilts of thin crystals are required, the former may be somewhat faster (Turner, 1967). Values of $I_{hk0}(\beta)$ for the 110 and 310 lines of



(a)

(b)

(c)

Fig. 1. The effect of angular distribution of crystallites in the specimen on the distribution of scattering power in reciprocal space. The scattering power results from the convolution of the angular function $T(\beta)$ with the shape transform functions of the crystallites. (*a*) Schematic illustration of the distribution of reciprocal lattice points for a gaussian function $T(\beta)$ symmetrically placed about the incident beam direction. (*b*) For thick crystals the dynamical shape transform is usually much smaller than the distribution function, $T(\beta)$. (*c*) For very thin crystals the shape transform can extend far beyond $T(\beta)$.

reflexions in BiOCl were computed for the values of β needed in the numerical integrations of equations (2) and (2a). The results for J_{110}/J_{310} as a function of thickness are shown in Fig. 2 for three values of the angular spread α. The effect which highly coherent illumination could have on the ratio in the case of bent crystals is illustrated in Fig. 3, in which results obtained using equations (2) and (2a) are compared. An additional average over thickness smooths out these curves slightly, but a large dependence of J_{110}/J_{310} on the distribution functions and beam coherence remains.

Discussion

It is apparent that the large deviations from the two-beam curve of the intensities measured by Kuwabara (1967) can be interpreted in terms of n-beam interactions. By suitable choice of the parameters specifying specimen morphology it is possible to obtain calculated intensities which range on either side of the primary extinction curve. Qualitative arguments suggest that even greater deviations would be found for the cases where non-systematic interactions are strong, and two-dimensional calculations are required (Kuwabara, Turner & Cowley, 1966). However, the calculation time becomes unreasonably long for the large numbers of beams and orientations which must be included for such cases.

These results are hardly surprising in the light of the many calculations recently made which demonstrate that intensities from single crystals under n-beam conditions are extremely sensitive to orientation and thickness.

For polycrystalline materials containing fairly heavy atoms, good agreement between experimental and theoretical estimates of intensities can be expected only when the following points are observed:

(a) The specimen morphology should be examined by electron microscopy to ensure that the crystals are not bent to the extent that beam coherence will have important effects, and do not contain other defects which may appreciably affect the intensities.

(b) The ranges of thickness and orientation of the crystals should be determined, by electron microscopy or from the diffraction pattern obtained with an energy filter, and incorporated into n-beam systematic calculations.

(c) Intensities should be measured accurately, preferably using an energy filter to remove inelastically scattered electrons. The specimen tilts should be chosen in such a way that the assumption of systematic diffraction conditions is justified.

The recent general theory of X-ray diffraction presented by Zachariasen (1967) is based on the assumption of two-beam dynamic scattering in a mosaic crystal. Zachariasen suggests that the extension of this theory to the electron case is trivial. However such a treatment would have little relevance for electron diffraction because of the neglect of coherent n-beam interactions.

n-Beam diffraction and structure analysis

Electron diffraction structure analyses have for the most part relied on formulas such as equation (3) for the interpretation of intensity data (Vainshtein, 1964). The results presented above cast considerable doubt on the validity of this procedure for crystals in which n-beam effects are significant.

The quality of an electron diffraction structure analysis is usually still measured in terms of the 'reliability factor', defined as

$$R^x = \sum_{\mathbf{h}} \left| |V^x_{\mathbf{h}}| - |V_{\mathbf{h}}| \right| / \sum_{\mathbf{h}} |V^x_{\mathbf{h}}| ,$$

where the $V_{\mathbf{h}}$ are calculated values for the proposed structure and the superscript x denotes experimental values. By the use of photographic methods of recording intensities, R^x is typically about 15%. It will be shown below that a further contribution, R^n, due to n-beam effects, can double this value (Table 1). It is clear that attempts to refine the structure by adjusting the atomic parameters so as to optimize the agreement between $V^x_{\mathbf{h}}$ and $V_{\mathbf{h}}$ values cannot be justified if R^n is large.

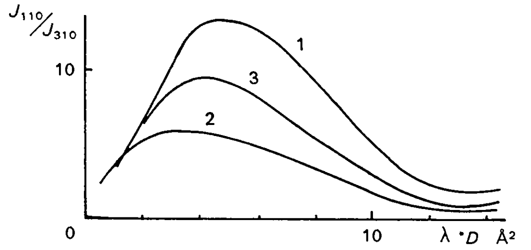


Fig. 2. Relative integrated intensities for BiOCl as a function of λ^*D and of angular half-width, α. (1) $\alpha = 1°$; (2) $\alpha = 2{\cdot}3°$; (3) $\alpha \to \infty$ (57° in the calculation). $\lambda^* = \lambda\sqrt{1 + h^2/m_0^2c^2\lambda^2}$ is the relativistic wavelength, λ, multiplied by the relativistic mass correction factor.

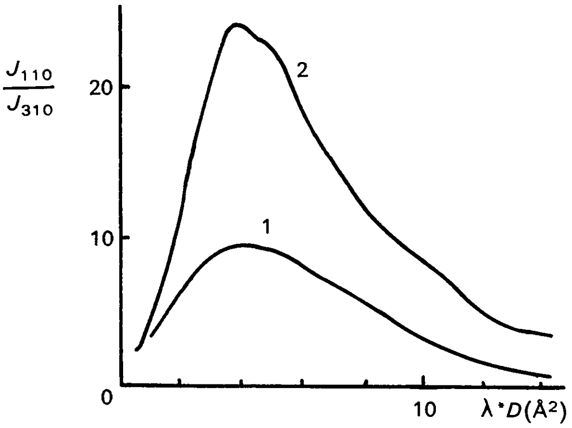


Fig. 3. Relative integrated intensities for BiOCl for large α and (1) integration over intensity; (2) integration over amplitude.

Since it is not possible to obtain values of V_h directly from experimental data, the basis of the analysis must be altered. We consider the experimental amplitudes,* $A_h^x = \sqrt{J_h^x}$ and attempt to optimize the agreement between these and realistic theoretical estimates, $A_h = \sqrt{J_h}$. where J_h is the computed integrated intensity. Only for the first step can the approximation $A_h = |V_h|$ be accepted if dynamic scattering is expected to be strong. With this approximation standard methods, such as Patterson projections, could be used to obtain an initial model of the structure which could then be refined using a better basis for the calculation of amplitudes.

The determination of the structure of $AgTlSe_2$ by Imamov & Pinsker (1965) may be taken as representative of the electron diffraction structure analyses carried out in recent years. In spite of the fact that Ag and Tl are quite heavy atoms, kinematic conditions were deduced from the observations that intensities appeared to decrease with angle roughly as the square of the scattering factors, and kinematical equations were used to obtain relative experimental values of V_h^x from the recorded intensities. Texture patterns were recorded from specimens with a preferred [001] axis perpendicular to the support film. From Patterson projections of the hexagonal unit cell the space group (No. 162: D_{3d}^1) and an initial set of atomic parameters were deduced. These parameters were refined by the use of electron density projections and by including a temperature factor until R^x had been reduced from 30% to 21%. The experimental values of the intensities were not listed, so it is not possible to attempt the alternative refinement procedure suggested above. However, the structure determined for $AgTlSe_2$ is suitable for our investigation of the significance of n-beam interactions.

Systematic n-beam calculations have been performed for the integration of the $h00$ reflexions of $AgTlSe_2$ using the structure parameters given by Imamov & Pinsker (1965). In Fig. 4 the results are plotted as a function of thickness, together with kinematic values. Dynamic effects become important for crystal thicknesses greater than 50 Å (6 unit cell layers). To compare these values with the experimental ones a scale factor is required.

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* In this section the geometric factors appearing in equation (2) are omitted.

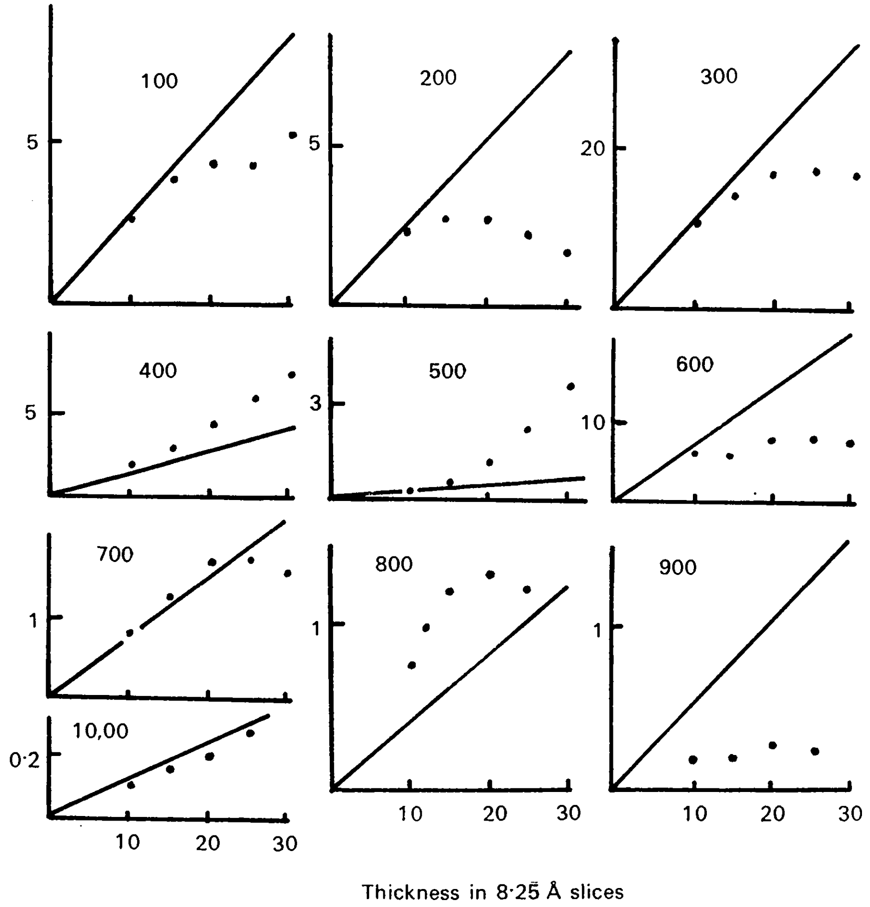

Fig. 4. Integrated intensities (arbitrary units) for $AgTlSe_2$ $h00$ systematic reflexions as a function of crystal thickness. Kinematical values are shown by the continuous lines; computed n-beam values by dots.

In X-ray diffraction structure analysis the refinement of the structure parameters is usually performed by use of a least-squares procedure to minimize the quantity

$$R_1 = \sum_{\mathbf{h}} W_{\mathbf{h}}(|F^o_{\mathbf{h}}| - b|F^c_{\mathbf{h}}|)^2 ,$$

where the $F^o_{\mathbf{h}}$ and $F^c_{\mathbf{h}}$ are the observed and calculated structure factors, b is an adjustable scale factor and the $W_{\mathbf{h}}$ are weighting factors determined from the experimental errors (see Lipson & Cochran, 1966). We define a similar quantity

$$P = \sum_{\mathbf{h}} (A^x_{\mathbf{h}} - bA_{\mathbf{h}})^2 / \sum_{\mathbf{h}} J^x_{\mathbf{h}} .$$

The scale factor b is chosen so that P is minimized, *i.e.*

$$b = \sum_{\mathbf{h}} A^x_{\mathbf{h}} A_{\mathbf{h}} / \sum_{\mathbf{h}} J^x_{\mathbf{h}} .$$

Approximate values of $A^x_{\mathbf{h}}$ for the $h00$ systematics of AgTlSe$_2$ were estimated from the graphical results given by Imamov & Pinsker (1965). In Fig. 5 these are compared with theoretical values based on kinematical ($P=6\%$) and n-beam calculations with the use of the final atom coordinates given by Imamov & Pinsker ($P=4\%$ for thickness $D=124$ Å). The agreement is significantly improved by including dynamical interactions, but the deviations of the latter theoretical results from the experimental values suggest that the atom coordinates may be in error.

This suggests that more reliable determinations of structures are likely if n-beam calculations are used since it can be shown that the kinematical process of refinement can lead to incorrect values of the structure parameters. A set of 'experimental' data was obtained by summing the theoretical results from n-beam calculations for three thicknesses ($D=82$ Å, $D=124$ Å, $D=165$ Å). The variables in the AgTlSe$_2$ structure which affect the $h00$ intensities are the $\mathbf{x}$ parameters of the Ag and Se atoms. Intensities were computed for the set of models in which $\mathbf{x}_{\mathrm{Ag}}$ and $\mathbf{x}_{\mathrm{Se}}$ were varied as shown in Table 1. These were then compared with the 'experimental' data by evaluating the factor P for both the dynamical amplitudes $A_{\mathbf{h}}$ and kinematical amplitudes (Table 1). It can be seen that in the dynamical case P is a minimum for the correct values of $\mathbf{x}_{\mathrm{Ag}}$ and $\mathbf{x}_{\mathrm{Se}}$, whereas in the kinematical case the minimum is shifted from the correct position by more than 0·1 Å.

Table 1. *Values of* $P(\%)$ *computed for* 13 *models of the* AgTlSe$_2$ *structure, in which the* $\mathbf{x}$ *parameters of the* Ag *and* Se *atoms were varied as shown*

The 'experimental' results were obtained for $\mathbf{x}_{\mathrm{Ag}}=0{\cdot}350$, $\mathbf{x}_{\mathrm{Se}}=0{\cdot}142$ as described in the text.

(*a*) Dynamical values of $A_{\mathbf{h}}$ were used; P shows a sharp minimum for $\mathbf{x}_{\mathrm{Ag}}=0{\cdot}350$, $\mathbf{x}_{\mathrm{Se}}=0{\cdot}142$ as expected.

(*b*) Using kinematical $A_{\mathbf{h}}$ values the minimum of P is shifted well away from the correct position. The contribution to the 'reliability factor' due to neglect of dynamical effects can be estimated for such cases, and is typically 10–15%.

(*a*) Dynamical $A_{\mathbf{h}}$'s

				$\mathbf{x}_{\mathrm{Ag}}$		
		0·330	0·340	0·350*	0·360	0·370
$\mathbf{x}_{\mathrm{Se}}$	0·134	1·59	–	1·40	–	2·57
	0·138	–	–	0·38	–	–
	0·142*	0·69	0·18	0·02	0·21	0·90
	0·146	–	–	0·27	–	–
	0·150	1·82	–	1·09	–	1·86

Table 1 (*cont.*)

(*b*) Kinematical $A_{\mathbf{h}}$'s

				$\mathbf{x}_{\mathrm{Ag}}$		
		0·330	0·340	0·350*	0·360	0·370
$\mathbf{x}_{\mathrm{Se}}$	0·134	6·2	–	2·7	–	4·0
	0·138	–	–	1·8	–	–
	0·142*	6·2	3·7	2·1	1·7	1·9
	0·146	–	–	6·1	–	–
	0·150	7·8	–	6·2	–	3·5

* Values determined by Imamov & Pinsker (1965).

The factor P is not particularly sensitive to changes in the temperature factor B (Fig. 6), and it seems likely that B could be kept constant until the final stages of refinement.

Conclusions

There is at present no alternative to the standard methods for the first stages of an analysis of a crystal structure by electron diffraction, to obtain an initial model of the positions of the atoms in the unit cell. Dynamical interactions and the effects of specimen morphology together cause a scatter of intensities about the primary extinction curve. For many crystals of interest, $(\sigma V_{\mathbf{h}} D)$ lies in the kinematical region of the

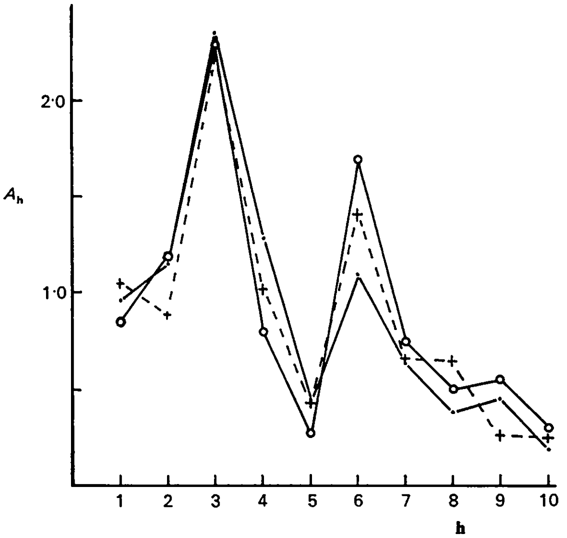

Fig. 5. Values of $A_{\mathbf{h}}$ (arbitrary units) for AgTlSe$_2$ $h00$ systematic reflexions: ——•—— experimental (after Imamov & Pinsker, 1965); --—○—— kinematical calculations (after Imamov & Pinsker; $P=6\%$); ——+----- n-beam calculations ($P=4\%$). The theoretical curves were scaled to the experimental one so that P was minimized.

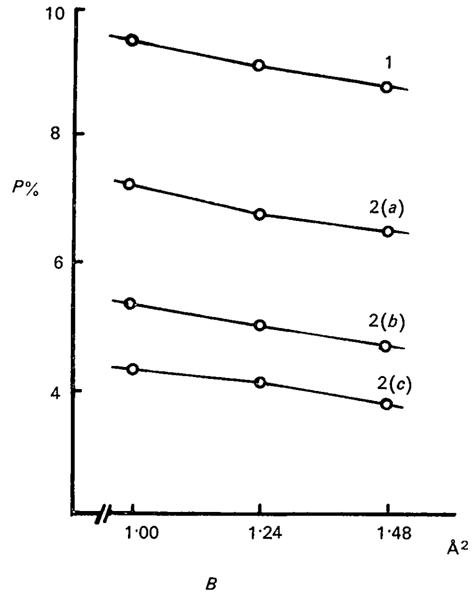

Fig. 6. Variation of P with temperature factor B for one of the structure models investigated. (1) Kinematical values of A_h. (2) Dynamical values for thicknesses: (a) $D=82$ Å; (b) $D=$: 124 Å; (c) $D=165$ Å.

Blackman curve, thus justifying the initial assumption of kinematical conditions.

Refinement of the atomic parameters by minimizing the differences between experimental amplitudes and V_h values is likely to lead to results significantly in error. However, it is possible to minimize instead the differences between experimental amplitudes and theoretical values based on dynamical calculations.

The success of such dynamical refinements will depend on the accuracy of the experimental data. The use of electronic recording systems and an energy filter seems advisable, together with some estimate of the distribution of thickness and orientation in the specimen. Electron microscopical examination of the specimens would be valuable for this and to check whether particular specimens are suitable for analysis.

We have attempted to demonstrate the inadequacies of the present methods of interpreting diffracted intensities from polycrystalline specimens when n-beam dynamical effects occur. It should be emphasized that the evidence presented is concerned with special cases and calculations for limited sets of data. A complete structure analysis along the lines indicated would be much more laborious but is desirable as a more searching test of our conclusions.

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