Acta Cryst. (1982). A**38**, 41–48

Treatment for the Intensity Problem of *n*-Beam Kinematical Reflections in a Dynamical Formalism

BY SHIH-LIN CHANG*

Instituto de Física 'Gleb Wataguin', Universidade Estadual de Campinas, Campinas – 13100, *Sao Paulo, Brasil*

(*Received* 5 *January* 1981; *accepted* 17 *June* 1981)

Abstract

The use of exponential functions as an approximation for reflection powers leads the intensity problem of *n*-beam kinematical diffraction to an eigenvalue problem. It is solved in the way similar to an *n*-beam Borrmann dynamical diffraction problem. Besides the intensities of Bragg-reflected beams, the excitation of modes concerning the intensity attenuation is also calculated for highly absorbing infinitely thick crystals. Experiments of multiple reflection from GaAs, InAs and InP single crystals for Cu $K\alpha$ radiation were carried out. Comparison between the experimental and calculated reflected intensities for several 2-, 3-, 4-, 5- and 8-beam cases are given and discussed.

Introduction

Based on the secondary extinction theory of X-ray and neutron diffraction, Moon & Shull (1964) derived a formalism for neutron diffraction intensities of multiple reflection in single crystals. A similar treatment was given by Zachariasen (1965) for X-ray cases. Since then, only a few theoretical works concerning the intensity aspect of multiple reflection from single crystals, in terms of kinematical theory, have been reported. These include the articles of Caticha-Ellis (1969), Prager (1971) and Parente & Caticha-Ellis (1974). Recently, Chang (1981) has extended the same treatment for multilayer systems in which several thin single-crystal layers are involved. Since the kinematical approach involves n differential equations, governing the exchange of power among the n diffracted beams, it is a formidable task to find the exact solution for the differential equations when $n > 3$. The approximation of using a Tayor's-series expansion, up to second order, as a solution has been proposed by Moon & Shull (1964). This approximation is only valid for reflections having low secondary extinction, *i.e.* with $Ql \ll 1$ and $\mu l \ll 1$, where Q, μ, and l are the reflectivity, the linear absorption coefficient and the path lengths of the diffracted beam within the crystal, respectively. However, these two conditions of validity are usually not satisfied simultaneously for highly absorbing crystals in X-ray cases. For these cases, iterative calculation for higher-order terms of the series expansion (Parente & Caticha-Ellis, 1974) together with correct polarization factors (Unangst & Melle, 1975) need to be considered. As pointed out by Chang (1981), it is difficult to find the correct polarization factors to insert in this iterative procedure, as far as the numerical calculation is concerned. To eliminate this difficulty, it seems helpful to reconsider the calculation procedure, based on the dynamical theory for n-beam Borrmann diffraction, in which polarization factors are associated with the corresponding electric susceptibilities and the susceptibilities are involved only once in the fundamental equation of the wavefield (Laue, 1949). In this paper, we assume that the approximate solutions of the differential equations are exponential functions. The kinematical problem is then treated in a way similar to the dynamical problem so that the polarization factors only enter in the beginning of the calculation. The series expansion for reflection power and the iterative procedure can therefore be omitted. For simplicity, only the cases involving infinitely thick crystals are considered. Measurement of the diffracted intensity of multiple reflection of Cu $K\alpha$ radiation from GaAs, InAs and InP plate-like crystals was carried out so as to compare with the theory.

Theoretical consideration

Multiple diffraction occurs when a crystal is so oriented that n sets of atomic planes are simultaneously in position to diffract an incident beam. Interaction among the n diffracted beams, within the crystal, modifies the reflection power of simple Bragg (2-beam) diffraction for each individual beam involved. Following Moon & Shull (1964), the interaction can be described by the following differential equation, involving the change of power in beam i, as it traverses a

* On leave at Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 7000 Stuttgart 80, Federal Republic of Germany, from March 1981 to February 1982.

0567-7394/82/010041-08$01.00

crystal layer of thickness dx at depth x below the upper crystal surface:

$$\pm \frac{dP_i}{dx} = -\frac{P_i}{\gamma_i}\mu + \sum_{j=1}^{n}\left(\frac{Q_{ji}P_j}{\gamma_j} - \frac{Q_{ij}P_i}{\gamma_i}\right) \quad (1)$$

for each i beam, where the summation is taken over all n diffracted beams. The plus and minus signs are for transmission (Laue) and reflection (Bragg) cases, respectively. P_i is the power in beam i and γ_i is the magnitude of the direction cosine of beam i relative to the crystal normal. Q_{ij} is the effective reflectivity, defined as

$$Q_{ij} = \left(\frac{\lambda^3 N^2 |F|^2}{\sin 2\theta}\right)_{ij} p_{ij}(i-j)\, W(\Delta\theta_{ij}), \quad (2)$$

where $W(\Delta\theta_{ij})$, the mosaic distribution function, is usually assumed to be a Gaussian. $\Delta\theta_{ij}$ is the deviation from the Bragg angle of $(i-j)$ reflection, θ_{ij}. λ, N and F are the wavelength of the X-rays used, the number of unit cells per unit volume, and the structure factor, respectively. $P_{ij}(i-j)$ is the polarization factor given by Zachariasen (1965), defined as

$$P_{ij}(i-j) = \tfrac{1}{2}[\cos^2 2\theta_i + \cos^2 2\theta_j + (\cos 2\theta_{l-j} - \cos 2\theta_i \cos 2\theta_j)^2]. \quad (3)$$

We assume that

$$P_i(x) = P_i e^{-\alpha x} \quad (4)$$

for all i is an approximate solution for (1), where α is a linear attenuation coefficient. By substituting (4) into (1), the differential equations form a set of linear equations:

$$(-S_i a_{ii} - \alpha) P_i - S_i \sum_{j\neq i}^{n} a_{ij} P_j = 0 \quad (5)$$

for all i, where

$$a_{ii} = -\left(\mu + \sum_{j\neq i}^{n} Q_{ij}\right)/\gamma_i \quad (6)$$

$$a_{ij} = Q_{ji}/\gamma_j \quad (7)$$

and

$$S_i = \begin{cases} +1 \text{ for transmission} \\ -1 \text{ for reflection.} \end{cases}$$

Equation (5) can be solved as an eigenvalue problem. There are n eigenvalues and eigenvectors. The former describe the attenuation and the latter give the ratio of reflection powers among the n diffracted beams. It is known, in dynamical theory, that the fundamental equations of the wavefield can be written as an eigenvalue equation. The eigenvectors provide the ratios of wavefield amplitudes among the diffracted waves, and the eigenvalues, which determine the resonance failure [*Anpassung* (Ewald, 1917)], define the mode of wave propagation. Correspondingly, in the present kinematical treatment, it would be convenient to define the state of attenuation associated with a given eigenvalue α as a mode of attenuation. For each k mode, there exists a ratio, *i.e.*

$$P_{l_1}(k) : P_{l_2}(k) : \ldots : P_{l_{n_T}}(k) : P_{m_1}(k) : \ldots : P_{m_{n_R}}(k) = C_{l_1}(k) : C_{l_2}(k) : \ldots : P_{l_{n_T}}(k) : C_{m_1}(k) : \ldots : C_{m_{n_R}}(k), \quad (8)$$

and a corresponding α_k, supposing that there are n_T transmissions and n_R reflections. They are labelled with subscripts l and m, respectively. l_1 indicates the direct beam. From (8), the proportionality constant X_k, defined as

$$X_k = \frac{P_l(k)}{C_l(k)} = \frac{P_m(k)}{C_m(k)} = \ldots, \quad (9)$$

can be determined from the boundary conditions at the upper crystal surface ($x = 0$). That is, the total reflected power inside the crystal should be equal to that outside the crystal for each diffracted beam. In other words,

$$\sum_k P_{l_1}(k) = P_{l_1}(x=0) = 1 \quad \text{for the incident beam,}$$

$$\sum_k P_l(k) = 0 \quad \text{for } l = l_2, l_3, \ldots, l_{n_T} \quad (10)$$

and

$$\sum_k P_m(k) = P_m(0) \quad \text{for } m = m_1, \ldots, m_{n_R}.$$

The summation is taken over all the permitted modes of attenuation. $P_m(0)$ is the power of the Bragg reflected beam, m, at $x = 0$. As mentioned by Chang (1979), the number of permitted modes, n_p, of X-ray dynamical diffraction for infinitely thick crystals is equal to $2(n - n_R)$. Similarly, by considering the fact that $|a_{ij}| \ll |a_{ii}|$ and $\mu \gg Q_{ij}$ in high absorption cases of X-rays, it would be very easy to show that the number of permitted modes in a kinematical n-beam diffraction is equal to $n - n_R$. The corresponding α for these permitted modes are positive. The rest of the modes, with negative α, are considered as the attenuation for a back-reflected beam from the lower crystal surface. Since we are dealing with an infinitely thick crystal, the modes associated with the back reflection from the lower surface should not be included in the calculation. By considering this point, (9) and (10) can be combined into the following matrix form:

$$\begin{pmatrix} C_{l_1}(1) & C_{l_1}(2)\ldots C_{l_1}(n_p) & \overbrace{0} & \overbrace{0 \ldots 0} \\ \vdots & & & \\ C_{l_{n_T}}(1) & C_{l_{n_T}}(2)\ldots C_{l_{n_T}}(n_p) & 0 & 0 \ldots 0 \\ C_{m_1}(1) & C_{m_1}(2)\ldots C_{m_1}(n_p) & -1 & 0 \ldots 0 \\ \vdots & & & -1 \\ & & & \qquad -1 \\ C_{m_{n_R}}(1) & C_{m_{n_R}}(2)\ldots C_{m_{n_R}}(n_p) & 0 & 0 \ldots -1 \end{pmatrix} \begin{pmatrix} X_1 \\ X_2 \\ \vdots \\ X_{n_p} \\ P_{m_1}(0) \\ P_{m_2}(0) \\ \vdots \\ P_{m_{n_R}}(0) \end{pmatrix} = \begin{pmatrix} 1 \\ 0 \\ \vdots \\ \\ \vdots \\ 0 \end{pmatrix}. \quad (11)$$

(n_R terms)

The diffracted powers $P_{m_1}(0)$, $P_{m_2}(0)$, ..., $P_{m_{n_R}}(0)$ of the reflections m_1, m_2, ..., m_{n_R} from the upper crystal surface are then obtained. The excitations of modes, defined as

$$\text{Ex}\,(k) = \frac{|X_k|}{\sum_{k=1}^{n_p} |X_k|},$$

are also determined.

Experimental

To verify the theoretical treatment given above, a multiple reflection experiment, similar to the one described by Renninger (1937), was carried out. The experimental set-up previously reported (Chang, 1981) was used. The angular divergence of the incident beam from a Cu target was about 20′ of arc. The specimens were GaAs, InAs and InP single-crystal slabs 10 × 10 mm square with a thickness of 500 μm. The large face was cut normal to [001].

The crystals were first set at the Bragg angles for 006 reflection and then rotated about the [006] reflection vector. When the so-called secondary reflections also satisfied Bragg's law, the interaction among reflections within the crystal modified the diffracted intensity of the primary 006 reflection. The modification on the 006 reflected intensity was recorded with a detector. Indexed 006 multiple diffraction patterns obtained for GaAs, InAs and InP of Cu $K\alpha_1$ and Cu $K\alpha_2$ are shown in Figs. 1, 2, 3 and 4. There, only the indices of secondary reflections are given. The peak intensities of multiple diffraction were measured with a slit of 0·3° at the azimuthal positions with maximum intensities. The total counts for each peak were accumulated up to more than 10^4 so that the error in counting statistics is less than 1%.

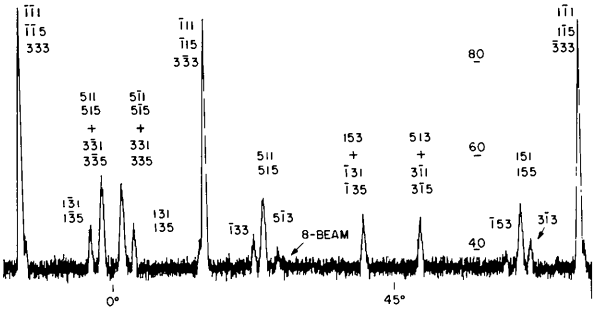

Fig. 1. 006 multiple diffraction pattern for Cu $K\alpha_1$ from GaAs. 45 kV, 30 mA. Full scale = 4 k counts s^{-1}.

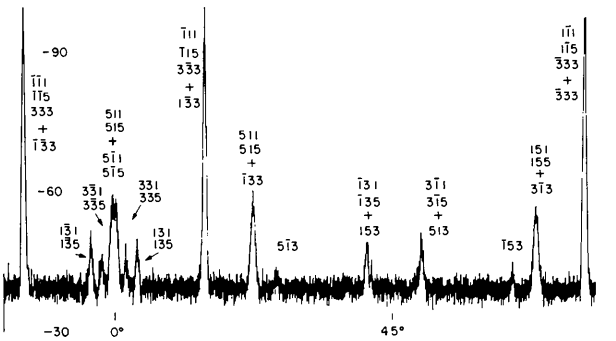

Fig. 2. 006 multiple diffraction pattern for Cu $K\alpha_2$ from GaAs. 45 kV, 30 mA. Full scale = 2 k counts s^{-1}.

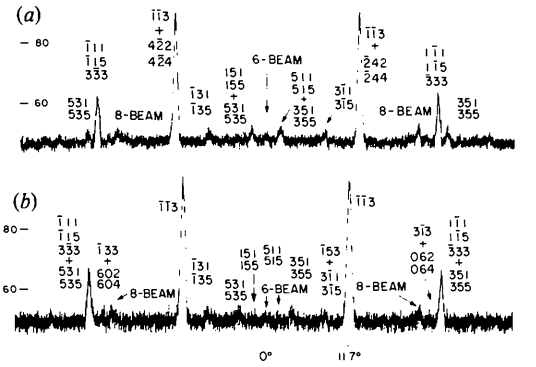

Fig. 3. 006 multiple diffraction patterns for Cu $K\alpha_1$ (*a*) and Cu $K\alpha_2$ (*b*) from InAs. 40 kV, 14 mA. (*a*) Full scale = 4 k counts s^{-1}. (*b*) Full scale = 2 k counts s^{-1}.

Calculations

Based on the above theoretical consideration, a computer program was written for calculating the diffracted intensities of a general *n*-beam diffraction, with $n \geq 2$. Since only the relative peak intensities were measured, the mosaic distribution $\bar{W}$ was set equal to unity so as to facilitate the calculation. Input data include the wavelength, λ, of the radiation used, lattice constant a, linear absorption coefficient μ, Miller indices of the n reflections, and structure factors. The structure factors were calculated based on the atomic factors from *International Tables for X-ray Crystallography* (1974), with temperature and anomalous scattering corrections (Table 1). The Debye parameters are 0·6 Å^2 for both Ga and As (*International Tables for X-ray Crystallography*, 1968) and 0·624 and 0·591 Å^2 for In and P at room temperature (Post, 1974). The values of μ and $1/\mu$ for GaAs, InAs and InP are listed in Table 2.

Because of the geometric relation between the reciprocal-lattice points and the Ewald sphere, secondary reflections with their reciprocal-lattice points lying on the equatorial circle are surface reflections. (The equatorial plane represents the crystal surface.) In the present case, the primary reflection is 006; therefore, secondary reflections *hkl* with $l = 3$ are surface reflections. They are always associated with those *n*-beam cases where n is an odd integer. Since the direction cosine of a surface reflection is zero, (1) has a singularity at $\gamma = 0$. To overcome this difficulty, an approximate solution for the diffracted intensity can be obtained in the following way (Chang, 1981). Surface reflection can be treated as either a Bragg (reflection) with $\gamma < 0$ or a Laue (transmission) with $\gamma > 0$. The approximate value for diffracted intensity can be obtained by interpolating the calculated intensity curve *versus* γ as γ approaches zero. For convenience, the calculations for even- and odd-order multiple diffractions are discussed separately below.

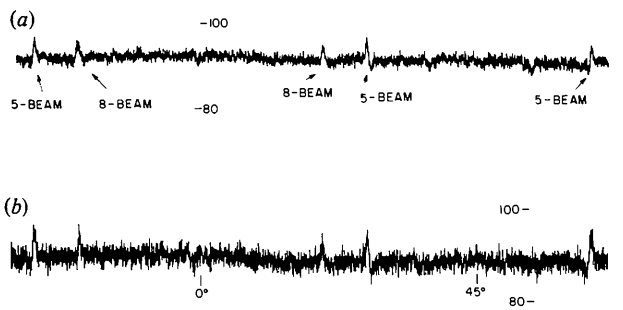

Fig. 4. 006 multiple diffraction patterns for Cu $K\alpha_1$ (*a*) and Cu $K\alpha_2$ (*b*) from InP. 42 kV, 22 mA. (*a*) Full scale = 10 k counts s^{-1}. (*b*) Full scale = 4 k counts s^{-1}.

(i) *Even-order multiple diffractions*

The cases 000 006 and 000 006 $1\bar{3}1$ $1\bar{3}5$ from GaAs and 000 006 $\bar{2}02$ 200 402 $\bar{2}04$ 206 404 from InP for Cu $K\alpha_1$ were chosen to illustrate the calculations for 2-, 4- and 8-beam diffractions, where 006 is a symmetric Bragg reflection. In Table 3, the attenuation coefficients α, the number of permitted modes n_p, the excitation of modes Ex, and the intensities of Bragg reflected beams I_{Bragg} are listed. The positive α's are approximately equal to μ/γ_l, where γ_l is the direction cosine of the corresponding transmitted beam l. The negative α's, approximately equal to μ/γ_m, are associated with the reflected beams. The α's are then listed together with their corresponding reflections in Table 3. It is clear that the small difference between α and μ/γ is due to the term $\sum_{j \neq i}^{n} Q_{ij}$ in (6). The mode associated with the direct beam, 000, is always the most excited mode. For GaAs, the attenuation α of this mode increases from

Table 1. *The structure factors for the reflections involved*

The + and − signs indicate $h + k + l$ are $4n + 1$, and $4n - 1$, respectively.

	|F|		
hkl	GaAs	InAs	InP
111(+)	147·971	209·778	179·572
111(−)	146·975	194·960	172·117
113(+)	114·179	168·289	137·216
113(−)	113·311	154·106	127·600
115(+) / 333(−)	80·865 / 80·251	126·299 / 113·012	105·159 / 98·806
133(+)	95·032	143·882	120·182
133(−)	94·296	130·088	113·780
135(+)	69·863	112·677	93·863
135(−)	69·339	100·019	87·649
335(+)	60·863	101·767	85·026
335(−)	60·406	89·827	79·051
155(+)	53·559	92·728	77·753
155(−)	53·156	81·530	72·076
355(+)	47·529	85·123	71·628
355(−)	47·158	74·662	66·279
002	6·863	60·741	118·863
006 / 244	6·538	34·679	63·256
024	6·538	42·491	81·839
022	174·219	239·243	182·361
004	142·769	200·174	151·332
044	103·526	153·000	116·049
026	89·927	136·989	104·385

Table 2. *The lattice constants a, the absorption coefficients* μ, *and* $1/\mu$ *for* GaAs, InAs *and* InP

		μ (mm^{-1})		$1/\mu$ (μm)	
	a (Å)	Cu $K\alpha_1$	Cu $K\alpha_2$	Cu $K\alpha_1$	Cu $K\alpha_2$
GaAs	5·6539	40·283	40·543	24·82	24·67
InAs	6·0580	101·763	102·375	9·83	9·77
InP	5·8696	98·979	99·568	10·10	10·04

Table 3. *Kinematical calculations for* 2-, 4- *and* 8-*beam cases*

	n	Laue	Bragg	α(mm^{-1})	n_p	Ex (%)	I_{Bragg}
GaAs	2	000		49·276	1	100·00	
			006	−49·276			0·27888 × 10^{-6}
GaAs	4	000		49·290		99·95	
		1$\bar{3}$1		73·942	2	0·05	
			006	−49·281			0·28211 × 10^{-6}
			1$\bar{3}$5	−73·933			0·24240 × 10^{-4}
InP	8	000		125·797		57·39	
		2$\bar{0}$2		377·297	4	0·01	
		200		125·751		42·60	
		402		377·236		0·2 × 10^{-2}	
			006	−125·729			0·78176 × 10^{-6}
			2$\bar{0}$4	−377·225			0·69310 × 10^{-5}
			206	−125·750			0·24106 × 10^{-4}
			404	−377·223			0·12261 × 10^{-4}

49·2755 to 49·2902 mm^{-1} as n changes from 2 to 4. The corresponding excitation decreases from 100 to 99·05%. The excitation of the mode with $\alpha = 73·942$ mm^{-1} is only about 0·05%. The 006 reflected intensity also changes from 0·2789 × 10^{-6} to 0·2821 × 10^{-6}.

In the 4-beam case, the ratio $I_{006}/I_{1\bar{3}5}$, equal to 1·16 × 10^{-2}, is approximately equal to $|F_{006}|^2/|F_{1\bar{3}5}|^2$, whose value is 0·88 × 10^{-2}. This means that the interaction among the 4 diffracted beams has modified the reflected intensities away from their 2-beam characteristics, *i.e.* $I \propto |F|^2$. For the 8-beam case from InP, two modes are most effectively excited. The excitations for these two modes are 57·39% for $\alpha = 125·797$ mm^{-1} and 42·60% for $\alpha = 125·751$ mm^{-1}. It seems that those modes with α very close to μ/γ_{000} play a more important role in the attenuation than those with α very different from μ/γ_{000}. In other words, the interaction between 000 and 200 reflections dominates the diffraction process. Also, because of this interaction, the reflected intensities are no longer proportional to $|F|^2$. For example, $I_{006} > I_{2\bar{0}4}$ and $I_{206} > I_{404}$, although $|F_{006}| < |F_{2\bar{0}4}|$ and $|F_{206}| < |F_{404}|$.

(ii) *Odd-order multiple diffractions*

000 006 3$\bar{1}$3 and 000 006 1$\bar{1}$1 1$\bar{1}$5 $\bar{3}$33 for Cu $K\alpha_1$ from GaAs are discussed here. 3$\bar{1}$3 and $\bar{3}$33 are surface reflections. As mentioned above, a surface reflection can be treated as either a Bragg reflection, Bragg–Bragg type, or a Laue transmission, Bragg–Laue type, with $|\gamma| \to 0$. In Table 4, the calculated reflected intensities, α and Ex are listed for the direction cosines γ_{surf} of the surface reflection equal to 0·4 and 0·6 for illustration. For the 3-beam Bragg–Laue-type diffraction in GaAs, there are two permitted modes, *i.e.* α = 49·281 and 67·148 mm^{-1}. The excitation of the mode (49·281 mm^{-1}) associated with 000 reflection increases while that of the mode (67·148 mm^{-1}) associated with 3$\bar{1}$3 reflection decreases as γ increases. This agrees with the physical situation that the closer to the crystal surface normal the 3$\bar{1}$3 diffracted beam is, the greater the excitation of the mode associated with it. For 3-beam Bragg–Bragg type, there is only one mode being excited. The magnitude of α and the 006 reflected intensity have the same values as those for the Bragg–Laue case. The 3$\bar{1}$3 reflected intensity decreases as γ decreases, since the transfer of power from the incident beam to the surface reflected beam for smaller γ is more difficult than for larger γ. Similar results are also obtained for the 5-beam case.

Results and discussions

Because the incident beam possessed an angular divergence about 0·3°, overlapping of several multiple diffraction peaks cannot be avoided. Those overlapped peaks, shown in Figs. 1, 2, 3 and 4, were indexed with

Table 4. *Kinematical calculations for* 3- *and* 5-*beam cases for* GaAs

Type	γ_{surf}	n	Laue	Bragg	α(mm^{-1})	n_p	Ex (%)	I_{Bragg}
BL	0·6	3	000		49·2810	2	99·97	
			3$\bar{1}$3		67·148		08·03	
				006	−49·277			0·27967 × 10^{-6}
BL	0·4	3	000		49·281	2	99·99	
			3$\bar{1}$3		100·721		0·01	
				006	−49·277			0·27980 × 10^{-6}
BB	0·6	3	000		49·281	1	100·00	
				006	−49·277			0·27967 × 10^{-6}
				3$\bar{1}$3	−67·148			0·47970 × 10^{-4}
BB	0·4	3	000		49·281	1	100·00	
				006	−49·279			0·27980 × 10^{-6}
				3$\bar{1}$3	−100·721			0·37233 × 10^{-4}
BL	0·6	5	000		49·329	3	97·79	
			1$\bar{1}$1		74·005		0·19	
			$\bar{3}$33		67·144		0·02	
				006	−49·302			0·30930 × 10^{-6}
				1$\bar{1}$5	−73·971			0·28885 × 10^{-4}
BL	0·4	5	000		49·329	3	99·80	
			1$\bar{1}$1		74·005		0·19	
			$\bar{3}$13		100·716		0·01	
				006	−49·302			0·30933 × 10^{-6}
				1$\bar{1}$5	−73·971			0·28885 × 10^{-4}
BB	0·6	5	000		49·329	2	99·81	
			1$\bar{1}$1		74·005		0·19	
				006	−49·302			0·30930 × 10^{-6}
				1$\bar{1}$5	−73·971			0·28885 × 10^{-4}
				$\bar{3}$33	−67·144			0·30070 × 10^{-4}
BB	0·4	5	000		49·329	2	99·81	
			1$\bar{1}$1		74·005		0·19	
				006	−49·302			0·30933 × 10^{-6}
				1$\bar{1}$5	−73·971			0·28885 × 10^{-4}
				$\bar{3}$33	−100·716			0·23342 × 10^{-4}

two or more sets of Miller indices. Those peaks labelled as 5-, 6- and 8-beam are 000 006 $1\bar{1}1$ $1\bar{1}5$ $\bar{3}33$, 000 006 $\bar{2}22$ $\bar{2}24$ $2\bar{2}2$ $2\bar{2}4$, and 000 006 $\bar{2}02$ 200 402 $\bar{2}04$ 206 404 reflections. Only the peak intensities of well-resolved multiple diffraction peaks were measured and subjected to the calculation.

By comparing the diffraction patterns of GaAs, InAs and InP, it is interesting to note that, because of the difference in lattice constant, the $1\bar{1}3$ reflection does not exist for GaAs and InP while it has a notable intensity for InAs. Also, the GaAs diffraction patterns show more peaks than those for InAs. The InP patterns have only two diffraction peaks above the 006 reflection background. This is because the 006 background for GaAs is lower than for InAs and much lower than for InP, since $F_{006}(\text{GaAs}) < F_{006}(\text{InAs}) < F_{006}(\text{InP})$ (Table 1). Clearly, some diffraction peaks, especially for InP, were smeared out by the high background.

In Tables 5, 6 and 7, the measured and calculated peak intensities for those well resolved multiple diffraction peaks are given for GaAs, InAs and InP for Cu $K\alpha_1$ and Cu $K\alpha_2$ radiation. ΔI and t_{eff} are the difference in intensity between the multiple diffraction peak and the 006 background, and the effective thickness by which the sample diffracts the incident beam. The latter were obtained with the program for calculating the intensity of multiple diffractions involving multilayers (Chang, 1981). The t_{eff}'s have a value of about $3/\mu$. (See Table 2 for $1/\mu$.) They increase when the multiple diffraction involves more transmission reflections and less Bragg and surface reflections, since transmission reflections tend to decrease absorption through Borrmann effects and Bragg reflections tend to shorten the length of beam penetration. The stronger the transmission is, the lower the absorption. And the weaker the Bragg reflection is, the longer the penetration path. In view of the values of the t_{eff}'s in Tables 5, 6 and 7 and the structure factors in Table 1, the above explanation seems reasonable.

As the diffracted intensity is concerned, I/I^* showed a good qualitative and fair quantitative agreement between the experimental and calculated results for the cases involving weak reflections. However, the calculated $\Delta I/I$ is about one or two orders of magnitude smaller than the observed values. This is because $\Delta I/I$ is very sensitive to small variations in diffracted intensity when I is very close to $I_{n=2}$. (Better agreement can be obtained if one normalizes $\Delta I/I$ in the same way as I/I^*.) The lack of quantitative agreement can directly be understood from (1) and (5). For high-absorption cases, μ is much greater than Q_{ij}. For instance, the μ's are about three orders of magnitude larger than Q_{ij} for GaAs, InAs and InP crystals with respect to Cu $K\alpha$ radiation. Taking this into account, the equations imply that (i) α must be almost equal to μ/γ and (ii) the participation of secondary reflections

Table 5. *The effective thicknesses and the calculated and observed* 006 *reflected intensities of multiple diffractions for* Cu $K\alpha_1$ *and* Cu $K\alpha_2$ *from* GaAs

† indicates that the intensity of this reflection is used as unity for normalization.

		I_{006}		I/I^*		$\Delta I/I_{n=2}$		
n	Reflection	Obs. (counts/60 s)	Calc. (10^{-6})	Obs.	Calc.	Obs.	Calc.	t_{eff} (μm)
	Cu $K\alpha_1$							
2		27545	0·27888	0·928	0·999	0	0	67·95
3	$3\bar{1}3$	32432	0·27980	1·093	1·002	0·177	0·003	61·28
3	$\bar{1}53$†	29677	0·27914	1·00	1·00	0·077	0·001	65·70
4	151 155	38795	0·28284	1·307	1·013	0·408	0·014	67·54
4	$1\bar{3}1$ $1\bar{3}5$	33996	0·28211	1·146	1·011	0·234	0·012	67·09
4	131 135	33424	1·28221	1·126	1·011	0·213	0·012	67·05
5	$1\bar{1}1$ $1\bar{1}5$ $\bar{3}33$	69366	0·30934	2·337	1·108	1·518	0·109	60·11
	Cu $K\alpha_2$							
2		15720	0·15171	0·864	0·994	0	0	67·81
3	$\bar{1}53$	16633	0·15186	0·914	0·995	0·058	0·001	65·41
4	$3\bar{1}1$ $3\bar{1}5$	19173	0·15347	1·054	1·005	0·220	0·012	67·02
4	131 135	19157	0·15353	1·053	1·006	0·219	0·012	66·97
4	331† 335	18189	0·15267	1·00	1·00	0·157	0·006	67·97
4	$3\bar{3}1$ $3\bar{3}5$	18380	0·15270	1·011	1·000	0·169	0·007	67·95

Table 6. *The effective thicknesses and the calculated and observed* 006 *reflected intensities of multiple diffraction for* Cu $K\alpha_1$ *and* Cu $K\alpha_2$ *from* InAs

† indicates that the intensity of this reflection is used as unity for normalization.

		I_{006}		I/I^*		$\Delta I/I_{n=2}$		
n	Reflection	Obs. (counts/60 s)	Calc. (10^{-5})	Obs.	Calc.	Obs.	Calc.	t_{eff} (μm)
	Cu $K\alpha_1$							
2		38582	0·19652	0·956	0·999	0	0	26·79
4	351 355	40562	0·19663	0·995	1·001	0·051	0·0005	27·53
4	$3\bar{1}1$† $3\bar{1}5$	40366	0·19659	1·00	1·00	0·046	0·0003	27·52
5	$1\bar{1}1$ $1\bar{1}5$ $\bar{3}33$	51802	0·19733	1·265	1·004	0·324	0·0041	26·86
8	$\bar{2}02$ 200 402 $\bar{2}04$ 206 404	41476	0·19661	1·027	1·000	0·075	0·0004	20·90
	Cu $K\alpha_2$							
2		19812	0·10657	0·954	0·999	0	0	26·78
3	$\bar{1}\bar{1}3$	38470	0·10676	1·853	1·002	0·942	0·0018	24·93
4	$3\bar{1}1$† $3\bar{1}5$	20759	0·10660	1·00	1·00	0·048	0·0003	27·55
4	351 355	20921	0·10663	1·008	1·000	0·056	0·0006	27·55
8	$\bar{2}02$ 200 402 $\bar{2}04$ 206 404	20816	0·10661	1·003	1·000	0·051	0·0004	20·91

Table 7. *The effective thicknesses and the calculated and observed* 006 *reflected intensities of multiple diffraction for* Cu $K\alpha_1$ *and* Cu $K\alpha_2$ *from* InP

† indicates that the intensity of this reflection is used as unity for normalization.

		I_{006}		I/I^*		$\Delta I/I_{n=2}$		
n	Reflection	Obs. (counts/100 s)	Calc. (10^{-5})	Obs.	Calc.	Obs.	Calc.	t_{eff} (μm)
	Cu $K\alpha_1$							
2		945692	0·78157	0·968	0·999	0	0	31·99
5	$1\bar{1}1$† $1\bar{1}5$ $\bar{3}33$	977426	0·78184	1·00	1·00	0·034	0·0004	38·51
8	$\bar{2}02$ 200 402 $\bar{2}04$ 206 404	967906	0·78176	0·990	1·000	0·024	0·0003	23·78
	Cu $K\alpha_2$							
2		474647	0·42448	0·983	0·999	0	0	31·98
5	$1\bar{1}1$† $1\bar{1}5$ $\bar{3}33$	488303	0·42463	1·00	1·00	0·029	0·0004	39·01
8	$\bar{2}02$ 200 402 $\bar{2}04$ 206 404	484207	0·42458	0·992	1·000	0·020	0·0002	23·76

will only give a third-order modification on the diffraction power of the primary reflection for these particular cases. These points have clearly been demonstrated in Tables 3 and 4. As diffraction in real crystals is concerned, the dynamical effects always accompany the diffraction process. The dynamical interaction among diffracted beams usually gives the 2-beam reflected intensity a zeroth-order modification for strong reflections and a first- or second-order modification for medium or weak reflections. It is therefore not surprising that the calculated $\Delta I/I$ are much smaller than the observed ones. The big difference between the observed and calculated I/I^* of $\bar{1}\bar{1}3$ from InAs for Cu $K\alpha_2$ is possibly due to the large Lorentz factor for $\bar{1}\bar{1}3$, which was not considered in the calculation.

For low-absorption thin crystals, back reflections from the lower crystal surface are as important as the forward diffraction from the upper surface. Those modes with negative α therefore need to be included in the calculation. There are n permitted modes for n-beam diffractions. If the reflectivities, Q_{ij}, are of the same order of magnitude as that of the linear absorption coefficient μ, the reflected intensity of the primary beam is expected to be modified a great deal and all modes involved will be considerably excited. The extinction would then play a more important role in diffraction for this case than for high-absorption cases.

Conclusion

From the above treatment, we conclude that the use of exponential functions for the calculation of the reflection powers leads us to treat the intensity problem of n-beam kinematical diffraction as an eigenvalue problem. The excitation of modes and reflected intensities can be calculated in a way similar to an n-beam Borrmann diffraction problem. It is therefore possible to know how the attenuation varies among the n diffracted beams, especially for cases involving many strong reflections. This would be impossible for the method that uses a series expansion as an approximate solution for the reflection powers.

It is also clear that the difference between our treatment and the dynamical one lies in the fact that, as usual, the former considers the reflection power, or intensity, and the latter the wavefield. Hence, in our treatment, the modes are associated with the attenuation of the intensities and in the dynamical cases they are associated with wave propagation.

Moreover, in our treatment, the effect of crystal thickness on the diffracted intensities has been considered. Direct comparison of reflected intensities can be made for various crystals as long as the experimental conditions remain the same.

References

CATICHA-ELLIS, S. (1969). *Acta Cryst.* A**25**, 666–673.

CHANG, S. L. (1979). *Acta Cryst.* A**35**, 543–547.

CHANG, S. L. (1981). *Acta Cryst.* A**37**, 876–889.

EWALD, P. P. (1917). *Ann. Phys. (Leipzig)*, **54**, 519–556, 557–597.

International Tables for X-ray Crystallography (1968). Vol. III, p. 241. Birmingham: Kynoch Press.

International Tables for X-ray Crystallography (1974). Vol. IV, p. 80, Birmingham: Kynoch Press.

LAUE, M. VON (1949). *Acta Cryst.* **2**, 106–113.

MOON, R. M. & SHULL, C. G. (1964). *Acta Cryst.* **17**, 805–812.

PARENTE, C. B. R. & CATICHA-ELLIS, S. (1974). *Jpn. J. Appl. Phys.* **13**, 1501–1505.

POST, B. (1974). *Anomalous Scattering*, edited by S. RAMASESHAN & S. C. ABRAHAMS, p. 85. Copenhagen: Munksgaard.

PRAGER, P. R. (1971). *Acta Cryst.* A**27**, 563–569.

RENNINGER, M. (1937). *Z. Phys.* **106**, 141–176.

UNANGST, D. & MELLE, W. (1975). *Acta Cryst.* A**31**, 234–235.

ZACHARIASEN, W. H. (1965). *Acta Cryst.* **18**, 705–710.