or smaller than the corresponding two-beam gap. For a given sign of the deviation parameter, the direction of the fringe bending is found to depend on the sign of the product between the three structure factors involved in the three-beam calculations. This effect may thus be utilized to determine three-phase structure invariants experimentally. In structure work, however, the method may at the present stage of development only be applied in a limited number of cases as relatively large single crystals are needed.

To determine the branches which contribute most strongly to the intensity the calculations of excitation coefficients have proved to be essential. Only the four branches, which correspond to the ones in the two-beam case, are, through such calculations, found to be of importance. These branches contribute to the intensity oscillations in pairs corresponding to the σ and π components. The present calculations therefore show that polarization contributes in the usual way to the fading, but are not essential for the interpretation of the three-beam effects studied.

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Diffraction by Crystals with Planar Domains

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Dedicated to Professor Dr H. Jagodzinski on his 65th birthday

Abstract

A general theory is developed for kinematical scattering by crystals with lamellar domains, having two equal lattice translations **b** and **c** which form a congruent plane of intergrowth. The domains differ in the lattice constant **a**, electron density distribution, and inter-domain distances. The size of the domains is described by arbitrary statistical distribution functions and the scattered intensity is calculated by forming the Patterson function and its Fourier transform. Examples with two types of domains are discussed.

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1. Introduction

A great number of electron microscopic and X-ray investigations deal with crystals with planar faults due

to exsolution processes or phase transitions. In order to understand the fault structure of these crystals it is necessary to calculate the diffuse scattering.

For this problem many theories have been developed (Wilson, 1942; Jagodzinski, 1949*a*,*b*,*c*; Kakinoki & Komura, 1952, 1954*a*,*b*; Cowley, 1976). In these theories it is assumed that the crystal consists of a certain number of different layers. Mathematically, the disordered structure is described by introducing *a priori* probabilities for the occurrence of a certain layer and assuming probabilities for a fault. The second variable describes the probability that, for instance, a layer of type i will be followed by a layer of type j. With either the so-called difference-equations method (Wilson, 1942) or the equivalent matrix method (Hendricks & Teller, 1942) or the direct calculation of the Patterson function (Cowley, 1976) it is possible to calculate the scattered intensity from the probabilities mentioned above.

A different approach to the calculation of the diffraction intensity of a disordered structure is to describe neighbouring layers of type i as one domain of type i. For this method, which was first used by Houston & Park (1970, 1971) to calculate the scattering of antiphase domains, the statistical distribution function of the various domain sizes occurring in the crystal is of great importance. If, for instance, a crystal consists of a sequence of two types of layers, i and j, and if α is the probability that an i layer will be followed by a j layer, then a layer of type i will follow again with a probability of $(1-\alpha)$. The probability that a domain of type i consists of three different layers is $(1-\alpha)^2$ *etc.* Fig. 1(*a*) demonstrates that one probability for a fault leads to an asymmetric domain size distribution relative to the average domain size $1/\alpha$. If we want to describe a symmetric distribution function with probabilities (see Fig 1), it is necessary to introduce several probabilities for a fault, generally up to the layers in the distance of the maximum domain size. Although Jagodzinski (1954) has shown that it is possible to solve even these cases, the mathematical difficulties grow rapidly with increasing domain sizes, owing to the increasing number of probabilities for a fault.

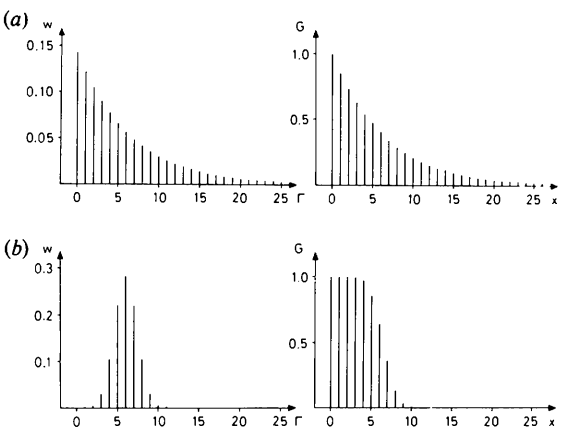

Fig. 1. Distribution function $w(\Gamma)$ of the domain sizes and averaged grating function $G(x)$ (equation $A2$) for two examples.

This problem does not arise when describing the disorder by a domain size distribution, because the number of parameters does not depend on the domain size. Furthermore the influence of different types of distribution functions on the diffuse scattering can be studied easily. In this work we present the calculation of the scattering of a crystal with an arbitrary number of lamellar types of domains. The sequence of the K different domains is cyclic, so that each domain has the same *a priori* probability of occurrence. Most frequently we have to deal with a crystal consisting of two types of domains. But it is possible that at the domain boundaries structures arise which differ from the two types of domains and then we generally have four types of domains. It should be noted that domain boundary structures consisting of only one layer can also be treated as a 'domain'.

2. Theory

A domain D_k of type k consists of a certain number of equal unit cells, characterized by the cell constants $\mathbf{a}_k$, $\mathbf{b}$, $\mathbf{c}$ and the electron density distribution ρ_k of a unit cell. Different domains of the same type k may have different domain sizes $\mathbf{a}_k.\Gamma_k$. The statistical distribution of these sizes is described by the domain size distribution function $w_k(\Gamma)$. Furthermore, it is assumed that the sizes of adjacent domains are uncorrelated, thus the different domain size distribution functions are statistically independent. The domains of different types vary in $\mathbf{a}_k$ and Γ_k, but they have the same lattice constants $\mathbf{b}$ and $\mathbf{c}$ (congruent plane of intergrowth). For an infinite crystal scattered intensity can therefore only be observed on lattice rods perpendicular to $\mathbf{b}$ and $\mathbf{c}$. Along these rods we can detect the disorder scattering which is caused either by differing lattice constants $\mathbf{a}_k$ or electron densities ρ_k or both.

The crystal consists of a cyclic sequence of K different types of domains. The domains of type 1 to K will be combined to a domain group characterized by the index l (Fig 2). A special domain $D_{k,l}$ of type k within the group l is characterized by a grating function $G_{k,l}(\mathbf{r})$, the electron density $\rho_k(\mathbf{r})$, and the domain distance $\mathbf{d}_k$. Since $\mathbf{b}$ and $\mathbf{c}$ are constant, we can omit the lattice sum along these directions and get the following domain function:

$$D_{k,l}(\mathbf{r}) = G_{k,l}(\mathbf{r}) * \rho_k(\mathbf{r}), \tag{1}$$

where

$$G_{k,l}(\mathbf{r}) = \sum_{\nu=0}^{\Gamma_{k,l}} \delta(\mathbf{r} - \nu\mathbf{a}_k).$$

The symbol $*$ stands for the convolution integral,

$$f(\mathbf{r}) * g(\mathbf{r}) = \int f(\mathbf{r}-\mathbf{r}')\, g(\mathbf{r}')\, \mathrm{d}\mathbf{r}'.$$

From (1) the electron density $\rho(\mathbf{r})$ of the entire crystal can be written as

$$\rho(\mathbf{r}) = \sum_k \sum_l D_{k,l}(\mathbf{r}-\mathbf{r}_{k,l}).$$

In order to get the scattered intensity we first form the Patterson function $P(\mathbf{r})$ per domain group.

$$LP(\mathbf{r}) = \rho(\mathbf{r}) * \rho(-\mathbf{r}),$$

where L is the total number of domain groups and

$$f(\mathbf{r}) * g(-\mathbf{r}) = \int f(\mathbf{r}+\mathbf{r}')\, g(\mathbf{r}')\, \mathrm{d}\mathbf{r}'.$$

2.1. *Two types of domains*

Because of the importance of this case we want to calculate its Patterson function first. The electron density is

$$\rho(\mathbf{r}) = D_{1,1}(\mathbf{r}-\mathbf{r}_{1,1}) + D_{2,1}(\mathbf{r}-\mathbf{r}_{2,1}) + D_{1,2}(\mathbf{r}-\mathbf{r}_{1,2}) + D_{2,2}(\mathbf{r}-\mathbf{r}_{2,2}) + \ldots.$$

The Patterson function $P(\mathbf{r})$ contains all pairs of domains. We sum up all pairs that are separated by an equal number m of domain walls and contribute to $P(\mathbf{r})$ for $\mathbf{r} \geq 0$. For $m = 0$ we get

$$\sum_l D_{1,l}(\mathbf{r}) * D_{1,l}(-\mathbf{r}) + \sum_l D_{2,l}(\mathbf{r}) * D_{2,l}(-\mathbf{r}).$$

In this sum always the same type of domain occurs and therefore it can be replaced by the average value

$$\{\langle D_{1,l}(\mathbf{r}) * D_{1,l}(-\mathbf{r})\rangle + \langle D_{2,l}(\mathbf{r}) * D_{2,l}(-\mathbf{r})\rangle\}\, L.$$

Since the electron density $\rho_k(\mathbf{r})$ is independent of the actual domain length we must only average the term with the grating functions and we get, with (1),

$$\begin{aligned}\Phi_k(\mathbf{r}) &= \langle D_{k,l}(\mathbf{r}) * D_{k,l}(-\mathbf{r})\rangle \\ &= \langle G_{k,l}(\mathbf{r}) * G_{k,l}(-\mathbf{r})\rangle * \rho_k(\mathbf{r}) * \rho_k(-\mathbf{r}),\end{aligned} \tag{2}$$

with

$$\langle G_{k,l}(\mathbf{r}) * G_{k,l}(-\mathbf{r})\rangle = \int G_k(\mathbf{r}) * G_k(-\mathbf{r})\, w_k(\Gamma)\, \mathrm{d}\Gamma.$$

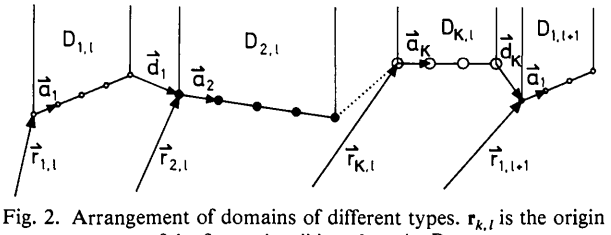

Fig. 2. Arrangement of domains of different types. $\mathbf{r}_{k,l}$ is the origin of the first unit cell in a domain $D_{k,l}$.

For $m = 1$ we have the contributions

$$\begin{aligned}&\langle G_{2,l}(\mathbf{r}-\mathbf{r}_{2,l}) * G_{1,l}(-\mathbf{r}-\mathbf{r}_{1,l})\rangle * \rho_2(\mathbf{r}) * \rho_1(-\mathbf{r}) \\ &\quad + \langle G_{1,l+1}(\mathbf{r}-\mathbf{r}_{1,l+1}) * G_{2,l}(-\mathbf{r}-\mathbf{r}_{2,l})\rangle \\ &\quad * \rho_1(\mathbf{r}) * \rho_2(-\mathbf{r}).\end{aligned}$$

Substituting $\mathbf{r}_{2,l} - \mathbf{r}_{1,l}$ by $\mathbf{a}_1.\Gamma_{1,l} + \mathbf{d}_1$ (see Fig. 2) the grating function of the first term becomes

$$\langle G_{2,l}(\mathbf{r}) * G_{1,l}(-\mathbf{r} + \mathbf{a}_1.\Gamma_{1,l})\rangle * \delta(\mathbf{r}-\mathbf{d}_1).$$

Now the statistically independent quantities are uncoupled and can be averaged separately. With the transformation

$$G_{k,l}(-\mathbf{r} + \mathbf{a}_k.\Gamma_{k,l}) = G_{k,l}(\mathbf{r}), \tag{3}$$

the contributions to the Patterson function become

$$\begin{aligned}\Phi_{2,1}(\mathbf{r}) &= \langle G_{2,l}(\mathbf{r})\rangle * \langle G_{1,l}(\mathbf{r})\rangle * \delta(\mathbf{r}-\mathbf{d}_1) \\ &\quad * \rho_2(\mathbf{r}) * \rho_1(-\mathbf{r})\end{aligned}$$

and (4)

$$\begin{aligned}\Phi_{1,2}(\mathbf{r}) &= \langle G_{1,l+1}(\mathbf{r})\rangle * \langle G_{2,l}(\mathbf{r})\rangle * \delta(\mathbf{r}-\mathbf{d}_2) \\ &\quad * \rho_1(\mathbf{r}) * \rho_2(-\mathbf{r}).\end{aligned}$$

For $m = 2$ we get

$$\begin{aligned}&\langle G_{1,l+1}(\mathbf{r}-\mathbf{r}_{1,l+1}) * G_{1,l}(\mathbf{r}-\mathbf{r}_{1,l})\rangle * \rho_1(\mathbf{r}) * \rho_1(-\mathbf{r}) \\ &\quad + \langle G_{2,l+1}(\mathbf{r}-\mathbf{r}_{2,l+1}) * G_{2,l}(\mathbf{r}-\mathbf{r}_{2,l})\rangle * \rho_2(\mathbf{r}) * \rho_2(-\mathbf{r}).\end{aligned}$$

Replacing $\mathbf{r}_{1,l+1} - \mathbf{r}_{1,l}$ by $\mathbf{a}_1.\Gamma_{1,l} + \mathbf{a}_2.\Gamma_{2,l} + \mathbf{d}_1 + \mathbf{d}_2$ in the first term we can separate again the statistically independent quantities $\Gamma_{1,l}$ and $\Gamma_{2,l}$. With the average domain size

$$B_k(\mathbf{r}) = \langle \delta(\mathbf{r} - \mathbf{a}_k.\Gamma_{k,l})\rangle, \tag{5}$$

we get

$$\begin{aligned}\Phi_{1,1}(\mathbf{r}) &= \langle G_{1,l+1}(\mathbf{r})\rangle * \langle G_{1,l}(\mathbf{r})\rangle * \rho_1(\mathbf{r}) * \rho_1(-\mathbf{r}) * B_2(\mathbf{r}) \\ &\quad * \delta(\mathbf{r}-\mathbf{d}_1-\mathbf{d}_2)\end{aligned}$$

and (6)

$$\begin{aligned}\Phi_{2,2}(\mathbf{r}) &= \langle G_{2,l+1}(\mathbf{r})\rangle * \langle G_{2,l}(\mathbf{r})\rangle * \rho_2(\mathbf{r}) * \rho_2(-\mathbf{r}) * B_1(\mathbf{r}) \\ &\quad * \delta(\mathbf{r}-\mathbf{d}_1-\mathbf{d}_2).\end{aligned}$$

Analyzing the contributions for $m = 3$ we obtain, as in the case of $m = 1$, pairs of different types of domains. But the mean distance of the domain pairs is larger by the average superstructure period of

$$R(\mathbf{r}) = B_1(\mathbf{r}) * B_2(\mathbf{r}) * \delta(\mathbf{r}-\mathbf{d}_1-\mathbf{d}_2). \tag{7}$$

Since the average values of the domain pairs do not depend on l we get with (7) the contribution

$$\{\Phi_{1,2}(\mathbf{r}) + \Phi_{2,1}(\mathbf{r})\} * R(\mathbf{r}).$$

The analogous comparison holds for $m = 4$ and $m = 2$.

From (2), (4), (6) and (7) we can now impose a scheme for the Patterson function for $\mathbf{r} \geq 0$.

$$P(\mathbf{r}) = P_s(\mathbf{r}) + P_d(\mathbf{r}) * \sum_{m \geq 0} R_m(\mathbf{r}), \tag{8}$$

with

$$P_s(\mathbf{r}) = \Phi_1(\mathbf{r}) + \Phi_2(\mathbf{r}),$$

$$P_d(\mathbf{r}) = \Phi_{1,2}(\mathbf{r}) + \Phi_{2,1}(\mathbf{r}) + \Phi_{1,1}(\mathbf{r}) + \Phi_{2,2}(\mathbf{r})$$

and the m-fold convolution

$$R_m(\mathbf{r}) = R(\mathbf{r}) * \ldots * R(\mathbf{r}).$$

$P_s(\mathbf{r})$ corresponds to the average Patterson function of uncorrelated domains, whereas $P_d(\mathbf{r})$ describes the correlation between the domains.

2.2. *General formulation*

The general formulation of the Patterson function per domain group is

$$LP(\mathbf{r}) = \sum_k \sum_{k'} \sum_l \sum_{l'} D_{k',l'}(\mathbf{r} - \mathbf{r}_{k',l'}) * D_{k,l}(-\mathbf{r} - \mathbf{r}_{k,l}).$$

If we substitute l' by $l + m$, for constant k, k' and m, the sum over l involves only pairs of the same type of domain. Therefore it can be replaced by a mean value that is independent of l.

$$LP(\mathbf{r}) = L \sum_k \sum_{k'} \sum_m \Phi_{k,k',m}(\mathbf{r}),$$

where

$$\Phi_{k,k',m}(\mathbf{r}) = \langle D_{k',l+m}\{\mathbf{r} - (\mathbf{r}_{k',l+m} - \mathbf{r}_{k,l})\} * D_{k,l}(-\mathbf{r})\rangle. \tag{9}$$

Now the index m sums over the various domain groups and we want to calculate $\Phi_{k,k',m}(\mathbf{r})$ for various m. Because the Patterson function is centrosymmetric we must calculate only $P(\mathbf{r})$ for $\mathbf{r} \geq 0$ and consequently $\mathbf{r}_{k',l} - \mathbf{r}_{k,l} \geq 0$ and $k' \geq k$ in the case of $m = 0$ (see Fig. 2). For $k = 0$ we get, as in (2),

$$\Phi_{k,k,0}(\mathbf{r}) = \langle G_{k,l}(\mathbf{r}) * G_{k,l}(-\mathbf{r})\rangle * \rho_k(\mathbf{r}) * \rho_k(-\mathbf{r}). \tag{10}$$

If $k' > k$ we introduce the substitution

$$\mathbf{r}_{k',l} - \mathbf{r}_{k,l} = \Gamma_{k,l} \cdot \mathbf{a}_k + \Gamma_{k+1,l} \cdot \mathbf{a}_{k+1} + \ldots + \Gamma_{k'-1,l} \cdot \mathbf{a}_{k'-1} + \mathbf{d}_k + \ldots + \mathbf{d}_{k'-1}.$$

Except for the domain width $\Gamma_{k,l}$ all other widths are statistically independent of the domain functions $D_{k',l}(\mathbf{r})$ and $D_{k,l}(\mathbf{r})$ in (9). From (3) and (5) we obtain for $k' > k$

$$\Phi_{k,k',0}(\mathbf{r}) = \langle G_{k',l}(\mathbf{r})\rangle * \langle G_{k,l}(\mathbf{r})\rangle * B_{k+1}(\mathbf{r}) * \ldots * B_{k'-1}(\mathbf{r}) * \delta(\mathbf{r} - \mathbf{d}_k - \ldots - \mathbf{d}_{k'-1}) * \rho_{k'}(\mathbf{r}) * \rho_k(-\mathbf{r}). \tag{11}$$

This equation contains $k' - k - 1$ widths $B_k(\mathbf{r})$ and $k' - k$ distances $\mathbf{d}_k$.

Investigating the case where $m = 1$ and $k' \leq k$ we get

$$\Phi_{k,k',1}(\mathbf{r}) = \langle G_{k',l+1}(\mathbf{r})\rangle * \langle G_{k,l}(\mathbf{r})\rangle * B_{k+1}(\mathbf{r}) * \ldots * B_K(\mathbf{r}) * B_1(\mathbf{r}) * \ldots * B_{k'-1}(\mathbf{r}) * \delta(\mathbf{r} - \mathbf{d}_k - \ldots - \mathbf{d}_K - \mathbf{d}_1 - \ldots - \mathbf{d}_{k'-1}) * \rho_{k'}(\mathbf{r}) * \rho_k(-\mathbf{r}). \tag{12}$$

Equation (12) contains $K - (k - k') - 1$ widths $B_k(\mathbf{r})$ and $K - (k - k')$ distances $\mathbf{d}_k$. The indices of B_k and $\mathbf{d}_k$ must be used cyclically because $k' \leq k$. If we use the indices of (11) in the same way, (11) and (12) become identical because the average values of the grating functions are independent of the index l. Consequently, we may combine the contributions to $P(\mathbf{r})$ for $m = 0$, $k' > k$ and $m = 1$, $k' \leq k$ as follows:

$$P_d(\mathbf{r}) = \sum_k \sum_{k'} \langle G_{k'}(\mathbf{r})\rangle * \langle G_k(\mathbf{r})\rangle * B_{k+1}(\mathbf{r}) * \ldots * B_{k'-1}(\mathbf{r}) * \delta(\mathbf{r} - \mathbf{d}_k - \ldots - \mathbf{d}_{k'-1}) * \rho_{k'}(\mathbf{r}) * \rho_k(-\mathbf{r}). \tag{13}$$

If $m = 1$ and $k' > k$ the number of widths in the term $\Phi_{k,k',1}$ is $K - (k - k') - 1 \geq K$. The number of K widths $B_k(\mathbf{r})$ and the distances $\mathbf{d}_k$ can be separated and combined to an average superstructural period

$$R(\mathbf{r}) = B_1(\mathbf{r}) * \ldots * B_K(\mathbf{r}) * \delta(\mathbf{r} - \mathbf{d}_1 - \ldots - \mathbf{d}_K), \tag{14}$$

and we get

$$\Phi_{k,k',1}(\mathbf{r}) = \Phi_{k,k',0}(\mathbf{r}) * R(\mathbf{r}).$$

The procedure is similar for $m = 2$, 3, *etc.*, so that we can form the Patterson function as in the case of two types of domains, this time with (10), (13) and (14).

$$P(\mathbf{r}) = P_s(\mathbf{r}) + P_d(\mathbf{r}) * \sum_{m \geq 0} R_m(\mathbf{r}),$$

with

$$P_s(\mathbf{r}) = \sum_k \Phi_{k,k,0}(\mathbf{r}).$$

The Patterson function consists of three terms:

(i) the average Patterson function of the uncorrelated domains $P_s(\mathbf{r})$;

(ii) the average Patterson function of two neighbouring domain groups $P_d(\mathbf{r})$;

(iii) the sum $\sum_{m \geq 0} R_m(\mathbf{r})$ considering the correlation of all domain groups.

After Fourier transforming $P(\mathbf{r})$ we get the scattered intensity. For the transformed figures we use the same symbols as for the Patterson function except for $P(\mathbf{r})$

and $\rho_k(\mathbf{r})$ which become $I(\mathbf{h})$ and $F_k(\mathbf{h})$ after the transform, respectively. The transformed figures are

$$R(\mathbf{h}) = B_1(\mathbf{h})\,B_2(\mathbf{h}) \times \ldots \times B_K(\mathbf{h}) \exp\{2\pi i\mathbf{h}(\mathbf{d}_1 + \ldots + \mathbf{d}_K)\}$$

$$P_s(\mathbf{h}) = \sum_k \langle G_k(\mathbf{h})\, G_k^*(\mathbf{h})\rangle F_k F_k^*$$

$$P_d(\mathbf{h}) = \sum_k \sum_{k'} \langle G_{k'}(\mathbf{h})\rangle \langle G_k(\mathbf{h})\rangle F_{k'} F_k^* B_{k+1}(\mathbf{h}) \times \ldots \times B_{k'-1}(\mathbf{h}) \exp\{2\pi i\mathbf{h}(\mathbf{d}_k + \ldots + \mathbf{d}_{k'-1})\}.$$

$\mathbf{h} = \mathbf{a}^* h + \mathbf{b}^* k + \mathbf{c}^* l$ is a vector within the reciprocal space. Because of the centrosymmetry of the Patterson function the scattered intensity per domain group is

$$I(\mathbf{h}) = P_s(\mathbf{h}) + 2\,\mathrm{Re}\left\{\frac{1}{1 - R(\mathbf{h})} P_d(\mathbf{h})\right\}. \qquad (15)$$

Equation (15) describes the diffuse scattering and the Bragg scattering which occurs when $R(\mathbf{h}) = 1$.

3. Examples

The theory developed above is exact within the framework of the kinematical theory, *i.e.* it contains no approximation. It makes it possible to calculate the scattering of very disordered as well as perfect crystals. The description of the domains by domain distribution functions allows for an investigation of the influence of different distribution functions. They can also be determined directly with electron microscopy and the comparison between calculated and measured intensities may possibly give additional information about the structure of the domain boundaries.

Yet the following will be restricted to examples of two types of domains, because these occur fairly often owing to exsolution processes. Especially in low-symmetric systems, one-dimensional disorder phenomena can be found frequently (Champness & Lorimer, 1975). The Fourier transform of (8) yields

$$I(\mathbf{h}) = \langle G_1(\mathbf{h})\, G_1^*(\mathbf{h})\rangle |F_1|^2 + \langle G_2(\mathbf{h})\, G_2^*(\mathbf{h})\rangle |F_2|^2 + 2\,\mathrm{Re}\left(\frac{1}{1 - R(\mathbf{h})} \{\langle G_2(\mathbf{h})\rangle \langle G_1(\mathbf{h})\rangle F_2 F_1^* \times \exp(2\pi i\mathbf{h}\mathbf{d}_1) + \langle G_1(\mathbf{h})\rangle \langle G_2(\mathbf{h})\rangle F_1 F_2^* \times \exp(2\pi i\mathbf{h}\mathbf{d}_2) + \langle G_1(\mathbf{h})\rangle \langle G_1(\mathbf{h})\rangle |F_1|^2 \times \exp[2\pi i\mathbf{h}(\mathbf{d}_1 + \mathbf{d}_2)]\, B_2(\mathbf{h}) + \langle G_2(\mathbf{h})\rangle \times \langle G_2(\mathbf{h})\rangle |F_2|^2 \exp[2\pi i\mathbf{h}(\mathbf{d}_1 + \mathbf{d}_2)]\, B_1(\mathbf{h})\}\right).$$

Let us first investigate the simple case where $\mathbf{a}_1 = \mathbf{a}_2 = \mathbf{d}_1 = \mathbf{d}_2 = (a,0,0)$. Here the translation vectors of the domains are equal and so the domains differ only in their structure factors. This fault structure can lead to a satellite diagram. The averaged period of the super-

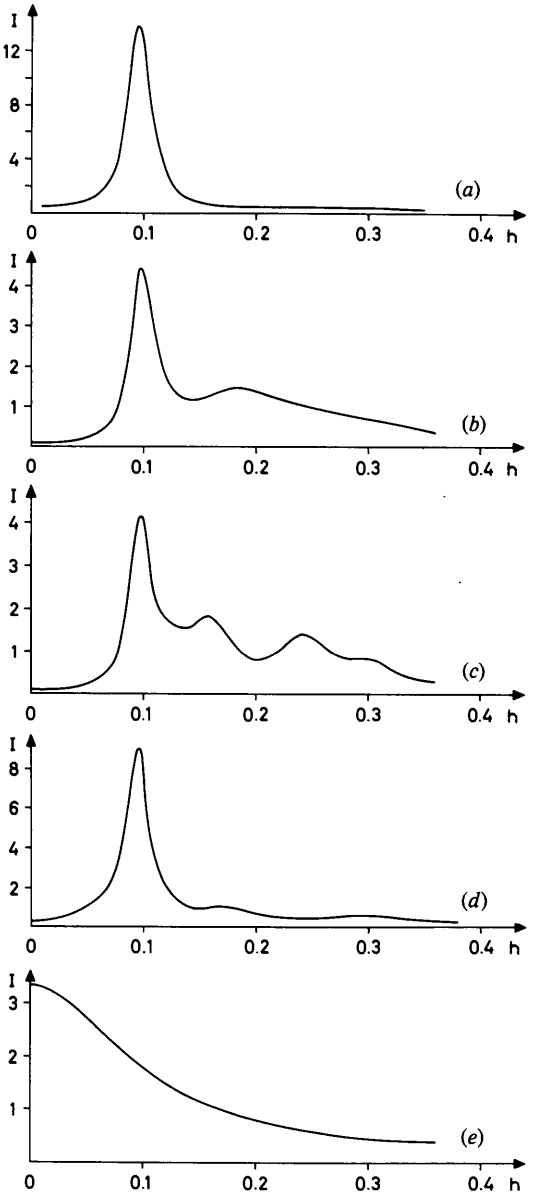

Fig. 3. Examples for diffuse scattering of a crystal with two types of domains with constant average superstructure = $10a$ and various domain distribution functions $v(\Gamma)$ (equation $A1$). (*a*) $v(\Gamma) = C\exp[-(\Gamma - \Gamma_0)^2/\sigma^2]$, where $\Gamma_0 = 5$ and $\sigma = 2$ for both types of domains. C = normalization constant, $F_1 = 1$, $F_2 = 2$. (*b*) The distribution function is Gaussian with $\Gamma_{0,1} = 8$, $\sigma_1 = 3$, $F_1 = 1$, $\Gamma_{0,2} = 2$, $\sigma_2 = 0$, and $F_2 = 2$. (*c*) The distribution function is rectangular $v(\Gamma) = C$ for $(\Gamma_0 - \sigma) \le \Gamma \le (\Gamma_0 + \sigma)$, $= 0$ otherwise. $\Gamma_{0,1} = 8$, $\sigma_1 = 3$, $F_1 = 1$, $\Gamma_{0,2} = 2$, $\sigma_2 = 0$, $F_2 = 2$. (*d*) The distribution function is Gaussian with $\Gamma_{0,1} = 7$, $\sigma_1 = 2$, $F_1 = 1$, $\Gamma_{0,2} = 3$, $\sigma_2 = 2$, $F_2 = 2$. (*e*) The distribution function is an exponential function $v(\Gamma) = C(1 - 1/\Gamma_0)^\Gamma$. Γ_0 stands for the average domain size (see Fig. 2). The special parameters are $\Gamma_{0,1} = 7$, $F_1 = 1$, $\Gamma_{0,2} = 3$, $F_2 = 2$.

structure can then easily be determined by the position of the first-order satellites. Furthermore, we are interested in the individual sizes and structure factors of the two domain types. If we deal with an exact superstructure, *i.e.* no variation of the domain sizes, the parameters mentioned above can be calculated with Korekawa's (1967) satellite theory or by constructing a supercell which equals the superstructure period. If the superstructure is not exact, sharp main reflections and diffuse scattering can be detected. The intensity distribution of the diffuse scattering depends mainly on the average size of the domains and the statistical distribution function $v_k(\Gamma)$ (equation $A1$, Fig. 1). Figs. 3 and 4 show the results of model calculations as a function of the width of the distribution function $v_k(\Gamma)$. Here Lorentzian functions have been fitted to the calculated profiles with the least-squares method. The standard deviation was always much better than for Gaussian functions. It is remarkable that the first-order satellites never have larger half-widths than 0·04**a*** despite the large widths σ_1 of the distribution functions. The integral intensity decreases rapidly with increasing σ_1. This indicates that for a quantitative analysis the integral intensity must be corrected even if the satellite profiles are broadened only slightly.

The line position of the satellites (Fig. 5) depends also on the width σ_1. Thus the line position of the satellites is not in commensurate positions in the

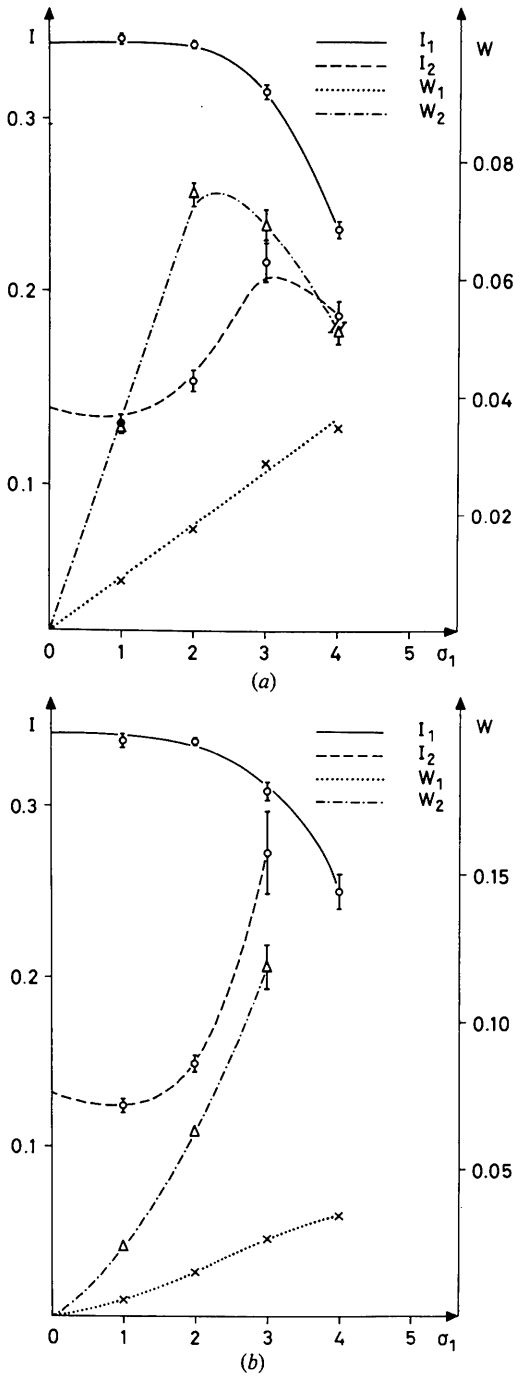

Fig. 4. Profile analysis of satellites of first and second order. Calculations for two types of domains with different distribution functions as a function of the width σ_1. The constant parameters are $\Gamma_{0,1} = 8$, $F_1 = 1$, $\Gamma_{0,2} = 2$, $\sigma_2 = 0$, $F_2 = 2$. (*a*) The integrated intensity I and full half-width W (in units of $1/a$) of the satellites of first and second order for a rectangular distribution function. (*b*) The same as in (*a*), but for Gaussian distribution.

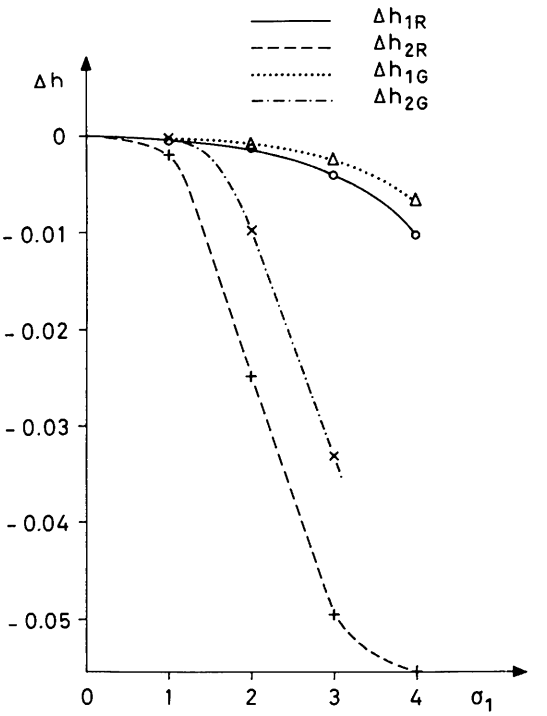

Fig. 5. Line shift of the satellites of first and second order as a function of σ_1 for the cases dealt with in Fig. 4. The quantity Δh is the difference between the position of the satellites in a perfect superstructure and the observed position. There is always a shift in the direction of the main reflection.

reciprocal lattice, although the average superstructure period represents an integral multiple of the lattice translation **a**. This can be explained easily by the fact that the position q_1 of the first-order satellite corresponds to the reciprocal value of the superstructure period P. For the case when we have a distribution over different periods the average line position $\langle q_1 \rangle$ does not correspond to the reciprocal period of $\langle P \rangle$ because $1/\langle P \rangle \neq \langle 1/P \rangle$. That is to say, an incommensurate structure can be simulated by diffraction effects although the structural modulation is locally and statistically commensurate.

The second-order satellites are already very diffuse at small σ_1 values. At the same time, the intensity increases rapidly. This may be explained if the observed intensities for large σ_1 do not represent the second-order scattering of the domains alone. First-order scattering of small domains and second-order scattering of large domains overlap. This explains also the significant line shift. Contrary to a Gaussian distribution, extremely large and small domains are equally probable as average-sized domains in a rectangular distribution and therefore the overlap of different orders has more weight. This explains the larger line shift for this distribution function.

An application to the case of two domains with different lattice constants **a** is shown in Fig. 6. The high-resolution focusing Weissenberg photograph (Peterat, 1981) of a lunar pyroxene demonstrates the well-known exsolution into Ca-poor (Pigeonit, P) and Ca-rich (Augit, A) lamellae (Ross, Bence, Dwoonik, Clark & Papike, 1970). The reflections of the Pigeonit and Augit lamellae are additionally split along the $(2\mathbf{a}^* - \mathbf{c}^*)$ direction ($A1$–$A2$, $P1$–$P2$). From the photometer curve (Fig. 6*c*) five maxima can be distinguished. The diffuse scattering along the $\mathbf{c}^*$ direction has two maxima which can be explained by the scattering of two types of lamellae with non-collinear lattice constants **c** (Peterat, 1981; Jagodzinski & Korekawa, 1972; Dorner & Jagodzinski, 1972). In this case the distribution function of the domains is described by one fault probability and then corresponds to the asymmetric domain size distribution function shown in Fig. 1.

The diffuse scattering along the $(2\mathbf{a}^* - \mathbf{c}^*)$ direction looks more complex. However, it is possible to explain this diffuse scattering by introducing an additional system of only two types of domain. With an asymmetric distribution function a splitting similar to the one along the $\mathbf{c}^*$ direction into $A1$–$A2$ and $P1$–$P2$ would be observed. A symmetric distribution function with small values of σ leads to an almost perfect superstructure. This is the reason for further splitting of the $A1$, $A2$ and $P1$, $P2$ reflections. The parameters of the distribution functions and the structure factors can be determined with a model calculation (Fig. 6*d*). It is also interesting to note that the width of the $A1$ reflections is determined by the width of the distribution function of the $A2$ domains and *vice versa*.

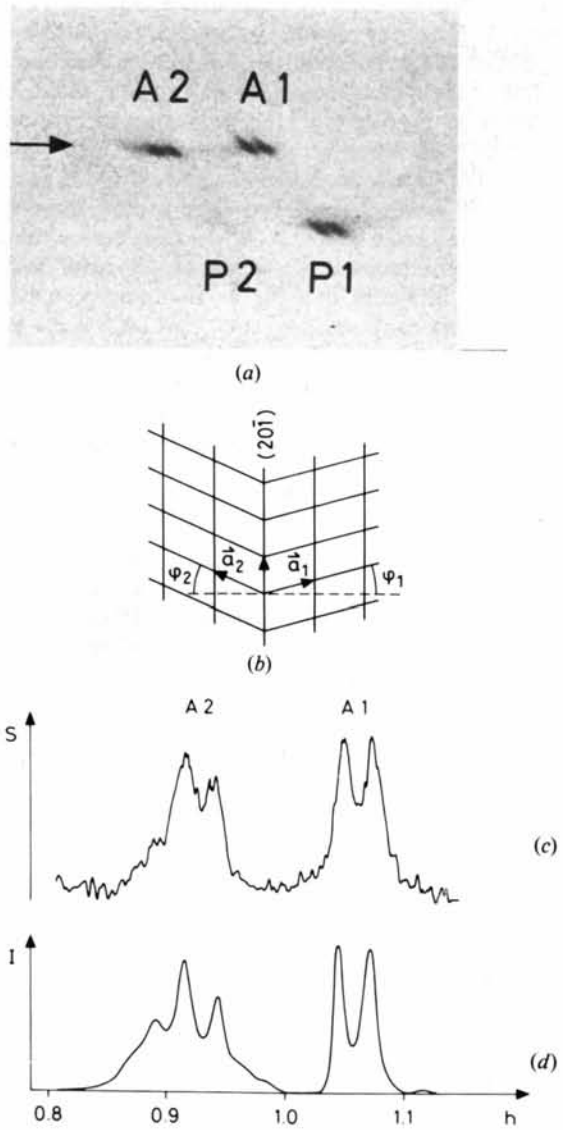

Fig. 6. (*a*) 004 reflection of an ($h0l$) Weissenberg photograph from a pyroxene of a lunar basalt (14053). Cu $K\alpha_1$ radiation, cylindrically bent Ge_{111} monochromator, and focusing technique was applied (Jagodzinski, 1968). (*b*) Two domains with a (20$\bar{1}$) plane of intergrowth and different lattice constants $\mathbf{a}_1$ and $\mathbf{a}_2$. (*c*) Densitometer curve of the $A1$–$A2$ reflections. The arrow in (*a*) indicates the scanning direction. (*d*) Model calculation where $d_{20\bar{1}}$ is equal for the two domains. Rectangular distribution function with $\Gamma_{0,1} = 24$, $\sigma_1 = 5$, $F_1 = 1$, $\varphi_1 = 4{\cdot}57°$, $\Gamma_{0,2} = 11$, $\sigma_2 = 2$, $F_2 = -\sqrt{2} + i\sqrt{2}$, $\varphi_2 = -3{\cdot}43°$.

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APPENDIX

For the calculation of the scattered intensity following equation (8), two averaging processes have to be performed with the computer. We would like to give some valuable transformations for the calculation.

The normalized domain size distribution function is given by

$$w_k(\Gamma) = \sum_{n=0}^{\infty} v_k(\Gamma)\,\delta(\Gamma - n), \qquad (A1)$$

where $v_k(\Gamma)$ is an arbitrary distribution function. With (A 1), the calculation of $\langle G_k(\mathbf{r})\rangle$ becomes

$$\langle G_k(\mathbf{r})\rangle = \int G_k(\mathbf{r})\, w_k(\Gamma)\,\mathrm{d}\Gamma$$
$$= \sum_{\nu=0}^{n} \delta(\mathbf{r} - \nu\mathbf{a}_k) \sum_{n=0}^{\infty} v_k(n).$$

The double sum can be transformed into the following more convenient equation:

$$\langle G_k(\mathbf{r})\rangle = \sum_{\nu=0}^{\infty} \delta(\mathbf{r} - \nu\mathbf{a}_k) \sum_{n=\nu}^{\infty} v_k(n). \qquad (A2)$$

The expression $\langle G_k(\mathbf{r}) * G_k(-\mathbf{r})\rangle$ is calculated similarly.

Following (A 1) we have

$$G_k(\mathbf{r}) * G_k(-\mathbf{r}) = \sum_{\nu=-\Gamma_k}^{\Gamma_k} \delta(\mathbf{r} - \nu\mathbf{a}_k)\,(\Gamma_k + 1 - |\nu|).$$

The averaging leads to

$$\langle G_k(\mathbf{r}) * G_k(-\mathbf{r})\rangle$$
$$= \int G_k(\mathbf{r}) * G_k(-\mathbf{r})\, w_k(\Gamma)\,\mathrm{d}\Gamma$$
$$= \sum_{\nu=-n}^{+n} \delta(\mathbf{r} - \nu\mathbf{a}_k)\,(n + 1 - |\nu|) \sum_{\nu=0}^{\infty} v_k(n).$$

This sum can again be transformed into

$$\langle G_k(\mathbf{r}) * G_k(-\mathbf{r})\rangle$$
$$= \sum_{\nu=-\infty}^{+\infty} \delta(\mathbf{r} - \nu\mathbf{a}_k) \sum_{n=|\nu|}^{\infty} (n + 1 - |\nu|)\, v_k(n).$$

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