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Substitutional Disorder Diffraction and Domain Size

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Abstract

The relation between the diffraction and the distribution of the linear dimensions of the ordered domains in substitutionally disordered polycrystalline material is elaborated. It is shown that, contrary to some theoretical expectances and experimental efforts, the fundamental reflections do not broaden (negligible change of cell dimensions with ordering being assumed). The forbidden reflections, on the contrary, are shown to be suitable, through their broadening, for the investigation of this disorder. The integral breadth is calculated for three important types of size distribution. The possibility of obtaining the average domain size from the integral breadth is analyzed in relation to these distributions. The variance may provide a second parameter for the size distribution, thus allowing selection among different plausible types. Therefore its relation to the size distribution is also elaborated.

1. Introduction

The relation between polycrystalline diffraction line profiles and the size distribution of the ordered domains in substitutionally disordered crystals is elaborated in this paper. Most of the interesting cases are cubic. Thus our derivations refer explicitly to cubic crystals.

2. Domain-size distribution and related functions

First, we shall derive relations, not to be found in the literature, between the size distribution and certain other probability functions. These will allow us to express the diffracted intensity in a compact form.

Let us take a set of N_o symmetrically equivalent positions in the unit cell. When occupied by more than one type of atom, the latter may be arranged in different manners in the different unit cells of the same crystal. The disorder thus arising is called *substitutional disorder*. A group of neighbouring unit cells with the same arrangement is called a *domain*. The arrangement in neighbouring domains is, of course, different.

In what follows, the occurrence of the different types of domains in a crystal will be taken as of equal probability. Furthermore, the change of the cell dimensions with the ordering will be assumed to be negligible. Substitutional disorder complying with these plausible assumptions will be referred to as 'our' type of disorder.

Let N be the number of different possible arrangements, *i.e.* of different domain types. When the equivalent positions are occupied, for example by two types of atoms so that there are N'_o of one and $N_o - N'_o$ of the other, then evidently

$$N = \binom{N_o}{N'_o}. \tag{2.1}$$

Let us denote by M a linear dimension of a domain, parallel to the Bragg vector $\mathbf{H}$ of the reflection under consideration, taken in the unit of the reticular distance D, parallel to $\mathbf{H}$. (In cubic lattices, there exists for every Bragg vector a family of reticular lines parallel to it.) Thus M is a dimensionless number! If $\mathbf{b}_1$, $\mathbf{b}_2$, $\mathbf{b}_3$ are the basic vectors of the reciprocal lattice, and h, k, l the (whole-number) indices of the reflection so that

$$\mathbf{H} = h\mathbf{b}_1 + k\mathbf{b}_2 + l\mathbf{b}_3, \tag{2.2}$$

then

$$D = \frac{h^2 + k^2 + l^2}{n^2}\, d, \tag{2.3}$$

as may easily be seen. n is the common divisor of h, k, l, *i.e.* the order of the reflection, and d the 'true' interplanar spacing of the lattice planes perpendicular to $\mathbf{H}$.

M may vary in one domain along its cross section perpendicular to $\mathbf{H}$, and it may take different sets of values for the different domains even of the same crystal, as the domains are not, in general, of the same shape and size. Let $q(M)$ be the relative number of occurrences, referred to unit range $\mathrm{d}M = 1$, of M in one crystal. It will be called *size distribution.*

Let us call Λ the reciprocal of the average domain size, *i.e. of the average value of M.* Thus

$$1/\Lambda = \int_0^\infty Mq(M)\,\mathrm{d}M. \tag{2.4}$$

As M, Λ is dimensionless.

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For the derivation of the relation between $q(M)$ and diffracted intensity, it is convenient to make use of the probability function $P(m)$, meaning the probability that, taking the arrangement at a randomly selected point, the same arrangement will be found at a distance m from it. m is taken in the same units as M.

$P(m)$ may be expressed as the sum of products of two factors. One is the probability that, *if* there are ν changes of arrangement along the segment m, the arrangement at a distance m will be the same as at 0. The other is the probability that the number of these changes will be ν. The product of these two factors, summed over ν, evidently gives $P(m)$. The reasoning is tedious but elementary, and leads to

$$P(m) = \int_m^\infty p(m_1)\,\mathrm{d}m_1 + \frac{1}{N}\sum_{\nu=2}^{\infty}\left\{\left[\frac{(-1)^\nu}{(N-1)^{\nu-1}} + 1\right] \times \int p(m_1)\,q(m_2)\ldots q(m_{\nu+1})\,\mathrm{d}m_1\,\mathrm{d}m_2\ldots\mathrm{d}m_{\nu+1}\right\}. \tag{2.5}$$

Here

$$p(m_1) = A\int_{m_1}^{\infty} q(M)\,\mathrm{d}M, \tag{2.6}$$

and the integrals are to be taken

$$\text{over}\left\{\begin{array}{l} m_1 \\ m_2 \\ \vdots \\ m_\nu \end{array}\right\} \text{ from 0 to } \left\{\begin{array}{l} m \\ m-m_1 \\ \\ m-(m_1+m_2+\ldots+m_{\nu-1}) \end{array}\right\},$$

and over $m_{\nu+1}$ from $[m-(m_1+m_2+\ldots+m_\nu)]$ to ∞.

3. Polycrystalline disorder diffraction

In what follows, the crystals will be assumed to be large enough not to cause particle-size broadening. The assumption of cubic crystals is maintained. A common function $q(M)$ is assumed to describe the size distribution parallel to $\mathbf{H}$, for all crystals of the polycrystalline material. This assumption is discussed in §7.1.

In the cubic case, it is possible to choose for every reflection such *new* basic lattice vectors $\mathbf{a}_1$, $\mathbf{a}_2$, $\mathbf{a}_3$, that $\mathbf{a}_1$ is parallel to $\mathbf{H}$ and has length D, and that $(\mathbf{a}_1\,\mathbf{a}_2\,\mathbf{a}_3) = a^3$, where a is the edge-length of the original cubic unit cell. This choice makes the calculation very simple, as now two of the new basic reciprocal vectors ($\mathbf{b}_2$ and $\mathbf{b}_3$) will be perpendicular to $\mathbf{H}$.

In this manner, one arrives at the expression of the diffracted intensity:

$$I(s) = \sum_m J(m)\cos(2\pi ms) - K(m)\sin(2\pi ms) \tag{3.1}$$

(see, for example, Szabó, 1975*a*), where

$$s = \left(\frac{2\sin\theta}{\lambda} - \frac{n}{d}\right)D; \tag{3.2}$$

θ is half of the angle of scattering; λ is the wavelength; $J(m)$ and $K(m)$ are the real and imaginary parts of the average of $F_{j_1j_2j_3}F^*_{j'_1j_2j_3}$ for a given value of

$$m = j'_1 - j_1; \tag{3.3}$$

the j's are the whole-number coordinates of the lattice points; F is the structure amplitude of the unit cell designated by its indices; and the asterisk means complex conjugate. The summation over m, here and in the following, is to be understood to mean from $-\infty$ to ∞.

$J(m) + iK(m)$ can easily be expressed by $P(m)$, taking all possible values of the product of structure amplitudes with their respective probabilities, and summing them. The probability for $F_{j'_1j_2j_3}$ being the same as $F_{j_1j_2j_3}$ is $P(m)$; and for $F_{j'_1j_2j_3}$ being a certain one among those which are different from $F_{j_1j_2j_3}$ the probability is $[1-P(m)]/(N-1)$, as can easily be seen. The result is

$$K(m) = 0, \tag{3.4}$$

$$J(m) = \frac{NP(m)-1}{N-1}\langle|F|^2\rangle + [1-P(m)]\frac{N}{N-1}|\langle F\rangle|^2. \tag{3.5}$$

Here

$$\langle F\rangle = \frac{1}{N}\sum_{n=1}^{N} F_n, \tag{3.6}$$

$$\langle|F|^2\rangle = \frac{1}{N}\sum_{n=1}^{N}|F_n|^2, \tag{3.7}$$

the index n designating the different possible arrangements in the unit cell.

Substituting (3.4) and (3.5) into (3.1),

$$I(s) = \sum_m\left\{\frac{NP(m)-1}{N-1}\langle|F|^2\rangle + [1-P(m)]\frac{N}{N-1}|\langle F\rangle|^2\right\}\cos(2\pi ms). \tag{3.8}$$

To be explicit, let us take here the case of substitution in only one set of equivalent positions. The generalization to substitution in more than one set is trivial (Szabó, 1975*b*).

The structure amplitude of a unit cell with the nth type of arrangement may be written as the sum of two terms: the first referring to the atoms of the substituted set, and the second to all the other atoms of the unit cell:

$$\mathbf{F}_n = \sum_t f_{n,t} \exp(-2\pi i \mathbf{H}\mathbf{r}_t) + C. \qquad (3.9)$$

The cardinal numbers t designate the different positions in the set; $f_{n,t}$ is the scattering amplitude of the atom in the tth position of the nth type of cell; and $\mathbf{r}_t$ is its position vector. The sum over t is to be taken from 1 to N_0. C is the contribution of the atoms which do not belong to the set. It does not have the index n, as it does not change with the change of arrangement in the substituted set.

The average of F_n over n is

$$\langle F\rangle = \left\langle \sum_t f_{n,t} \exp(-2\pi i \mathbf{H}\mathbf{r}_t)\right\rangle + C$$

$$= \langle f\rangle \sum_t \exp(-2\pi i \mathbf{H}\mathbf{r}_t) + C. \qquad (3.10)$$

Here we could write $\langle f\rangle$ instead of $\langle f_n\rangle_t$ because, on the basis of our assumptions, the latter evidently becomes the same for every t, and the averaging over n makes the index n superfluous.

Forbidden reflections are those for which $\sum_t \exp(-2\pi i \mathbf{H}\mathbf{r}_t) = 0$ and $C = 0$, *i.e.* which in the absence of substitution would not occur. The other reflections are called *fundamental*, as they occur in the presence as well as in the absence of substitution. Thus from (3.10),

$$\langle F\rangle \begin{Bmatrix} =0 \\ \neq 0 \end{Bmatrix} \text{ for the } \begin{Bmatrix} \text{forbidden} \\ \text{fundamental} \end{Bmatrix} \text{ reflections.} \qquad (3.11)$$

The displacement of the centroid of the reflection:

$$\langle s\rangle = \int_{-1/2}^{1/2} sI(s)\,\mathrm{d}s \bigg/ \int_{-1/2}^{1/2} I(s)\,\mathrm{d}s = 0 \qquad (3.12)$$

for the fundamental as well as for the forbidden reflections. In other words, no centroid displacement occurs with our type of disorder, as already stated in a less general type of treatment (Szabó, 1975*a*).

4. Integral breadth

The integral breadth β_s is the quotient of the integrated intensity and the maximum intensity. From (3.8), taking into account that $P(0) = 1$, the integrated intensity is

$$\int_{-1/2}^{1/2} I(s)\,\mathrm{d}s = \langle |F|^2\rangle; \qquad (4.1)$$

and

$$I_{\max} = I(0) = \langle |F|^2\rangle \sum_m \frac{NP(m)-1}{N-1}$$

$$+ |\langle F\rangle|^2 \frac{N}{N-1} \sum_m [1-P(m)]. \qquad (4.2)$$

Thus

$$\beta_s = \frac{(N-1)\langle |F|^2\rangle}{\langle |F|^2\rangle \sum [NP(m)-1] + |\langle F\rangle|^2 N \sum [1-P(m)]}. \qquad (4.3)$$

The integral breadth in the scale of the scattering angle 2θ, may be obtained (in radians) from β_s on grounds of (3.2):

$$\beta_{2\theta} = \frac{\lambda}{D\cos\theta_B}\,\beta_s. \qquad (4.4)$$

Here θ_B is the angle of reflection or Bragg angle.

From (4.3), because of (3.11),

$$\beta_s\,(\text{forbidden reflections}) = \frac{N-1}{\sum_m [NP(m)-1]}. \qquad (4.5)$$

In the case of the fundamental reflections, the second term in the denominator of (4.3) becomes infinite, and therefore

$$\beta_s\,(\text{fundamental reflections}) = 0. \qquad (4.6)$$

(4.5) and (4.6) lead to the conclusion, arrived at already in the approximate treatment of Szabó (1975*a*,*b*), that in the case of our type of disorder, broadening may occur only in the forbidden reflections, whereas the fundamental reflections remain sharp (as to the variance, see §6).

(4.5) shows the dependence of β_s on $P(m)$. The latter is determined by the domain-size distribution $q(M)$ through (2.5) and (2.6). In what follows, broadening will always refer to the forbidden reflections, unless stated otherwise.

5. Integral breadth for three important size distributions

Wilson (1958) quotes two domain-size distributions given by L. Landau and I. M. Lifschitz. We shall write them in our notation and units. One corresponds to the growth model of domain formation:

$$q(M) = \Lambda \exp(-\Lambda M), \qquad (5.1a)$$

and the other to the nucleation model:

$$q(M) = (2\Lambda)^2 M \exp(-2\Lambda m). \tag{5.1b}$$

In addition, there might be some theoretical interest in the model of a constant domain size:

$$q(M) = \delta(M - 1/\Lambda), \tag{5.1c}$$

where δ is the Dirac delta function. Layer-type domain patterns can have such distributions for all the Bragg vectors at the same time, although with different values of Λ for the different Bragg vectors.

$P(m)$ will now be calculated for these three distributions, using (2.6) and (2.5). Thus for case (a),

$$\begin{aligned} P(m) &= \exp(-\Lambda m)\left\{1 + \frac{1}{N}\sum_{\nu=2}^{\infty}\left[\frac{(-1)^{\nu}}{(N-1)^{\nu-1}} + 1\right]\frac{(\Lambda m)^{\nu}}{\nu!}\right\} \\ &= \frac{1}{N} + \frac{N-1}{N}\exp\left(-\frac{N}{N-1}\Lambda m\right). \end{aligned} \tag{5.2a}$$

For case (b),

$$\begin{aligned} P(m) &= \exp(-2\Lambda m)\left\{(1 + \Lambda m) + \frac{1}{N}\sum_{\nu=2}^{\infty}\left[\frac{(-1)^{\nu}}{(N-1)^{\nu-1}} + 1\right]\right. \\ &\quad \left.\times\left[\frac{1}{2}\frac{(2\Lambda m)^{2\nu-1}}{(2\nu-1)!} + \frac{(2\Lambda m)^{2\nu}}{(2\nu)!} + \frac{1}{2}\frac{(2\Lambda m)^{2\nu+1}}{(2\nu+1)!}\right]\right\} \\ &= \exp(-2\Lambda m)\left\{\frac{N-1}{N}\cos\frac{2\Lambda m}{(N-1)^{\frac{1}{2}}}\right. \\ &\quad \left. + \frac{(N-1)^2 - (N-1)}{2N(N-1)^{\frac{1}{2}}}\sin\frac{2\Lambda m}{(N-1)^{\frac{1}{2}}} + \frac{1}{N}\right\}. \end{aligned} \tag{5.2b}$$

For case (c), we obtain for $P(m)$ a continuous function which is linear in the intervals between the points $m = 0, 1/\Lambda, 2/\Lambda, \ldots, k/\Lambda, \ldots$. In the first interval, the derivative of $P(m)$ is negative, and changes sign and diminishes in absolute value from interval to interval. Designating $P(m)$ in the interval $(k-1)/\Lambda \leq m \leq k/\Lambda$ by $P_k(m)$, one obtains

$$\begin{aligned} P_k(m) &= \frac{1}{N} + (-1)^k\frac{1}{N(N-1)^{k-1}} \\ &\quad + (-1)^{k-1}\frac{k - \Lambda m}{(N-1)^{k-1}}. \end{aligned} \tag{5.2c}$$

In order to obtain the expression of β_s for these three cases, we have to substitute the expressions of $P(m)$ from (5.2a,b,c), one by one, into (4.5). Substituting (5.2a),

$$\beta_s = \frac{1 - \exp\left(-\frac{N}{N-1}\Lambda\right)}{1 + \exp\left(-\frac{N}{N-1}\Lambda\right)}. \tag{5.3a}$$

Substituting (5.2b),

$$\beta_s = \frac{\exp(2\Lambda) + \exp(-2\Lambda) - 2\cos\frac{2\Lambda}{(N-1)^{\frac{1}{2}}}}{\exp(2\Lambda) - \exp(-2\Lambda) + \frac{N-2}{(N-1)^{\frac{1}{2}}}\sin\frac{2\Lambda}{(N-1)^{\frac{1}{2}}}} \tag{5.3b}$$

The expression resulting from (5.2c) cannot be given in closed form, but may be evaluated by computer methods.

It is interesting to note that Wilson (1943) used for Cu_3Au the expression (5.2a), with $N = 4$, and in spite of this his integral breadth formula (11) differs from our (5.3a). Namely, expressing his quantity δ by our Λ, using our unit of length D, and transforming from the scale of 2θ to that of s, his equation (11) becomes

$$\beta_s = \tfrac{2}{3}\Lambda, \tag{5.3a)$'$}$$

which is only an approximation to our (5.3a), for $\Lambda \ll 1$. The origin of this difference is an approximation procedure used by Wilson.

In order to calculate Λ from an experimental value of β_s, one would have to know the type of the distribution $q(M)$ and use the corresponding formula of β_s. Let us make the plausible assumption that $q(M)$ is of the general type given by Lifschitz (see Wilson, 1958), *i.e.* that it has some intermediate form between the limiting forms (5.1a) and (5.1b). Then Λ obtained from the given β_s will be of some intermediate value between those calculated from (5.3a) and (5.3b): Λ_a and Λ_b respectively. Let us now compare Λ_a and Λ_b.

In the limit of large domains, *i.e.* $\beta_s \ll 1$, (5.3a) and (5.3b) may be approximated as

$$\beta_s = \frac{N}{2(N-1)}\Lambda_a, \tag{5.4a}$$

and

$$\beta_s = \frac{2N}{3N-4}\Lambda_b \tag{5.4b}$$

respectively. From these two equations,

$$\Lambda_a/\Lambda_b = \begin{Bmatrix} 3/2 \\ 4/3 \end{Bmatrix} \text{ for } N = \begin{Bmatrix} 4 \\ \infty \end{Bmatrix}. \tag{5.5}$$

For the comparison of Λ_a and Λ_b in the opposite limit of small domains, *i.e.* of large β_s, let us take a concrete numerical example: $\beta_s = 0{\cdot}25$. This corresponds to extremely small domains!

For this value of β_s,

$$\Lambda_a/\Lambda_b = \begin{Bmatrix} 0{\cdot}383/0{\cdot}25 \simeq 1{\cdot}5 \\ 0{\cdot}511/0{\cdot}39 \simeq 1{\cdot}3 \end{Bmatrix} \text{ for } N = \begin{Bmatrix} 4 \\ \infty \end{Bmatrix}, \qquad (5.6)$$

i.e. virtually the same as in the other limit.

Thus when we are entitled to assume the Lifschitz type of distribution, without knowing its exact form, it may be sufficient to calculate Λ_a, and remember that this can differ from the correct value of Λ at most by a factor 1·3 to 1·5, depending on N, for $N \geq 4$. This procedure may be carried out for different Bragg vectors (different reflections), thus giving Λ in this approximation for different crystallographic directions.

6. The moments of *I*(*s*)

For the selection of the correct function $q(M)$ among different possible forms, for example among the different forms of the Lifschitz type, one parameter is not sufficient. For instance, the choice between the forms (5.1*a*) and (5.1*b*) would require the determination of at least two parameters.

β_s alone gives only one parameter. More information could be gained only by a more detailed consideration of the intensity distribution $I(s)$: 'line profile analysis'. In principle, the different moments of $I(s)$ can serve for this. Let us now analyze the information they can provide.

The 0th and the 1st moment do not give anything for $q(M)$. Tacitly they have already been implied before dealing with $q(M)$. The 0th moment is the *integrated intensity*. Its dependence on *hkl* is taken to check the crystal structure. The 1st moment, divided by the 0th, is the *centroid displacement*. The vanishing of this proves that $K(m) = 0$, *i.e.* that the type of disorder is in harmony with our assumptions. Namely, in case of $K(m) \neq 0$, (3.12) would not hold.

The 2nd moment, divided by the 0th, is the *variance*. This gives one parameter, as does the integral breadth. Determining both, we can obtain two parameters for $q(M)$. We would like to suggest the carrying out of such determinations, in view of the important new knowledge which could be gained in this manner.

The variance,

$$W_s = \int_{-1/2}^{1/2} s^2 I(s)\, \mathrm{d}s \Big/ \int_{-1/2}^{1/2} I(s)\, \mathrm{d}s, \qquad (6.1)$$

may be expressed by $P(m)$. From (6.1), (3.8), (3.11), and (4.1), the variance for the *forbidden* reflections is

$$W_s\,(\text{forbidden reflections}) = \tfrac{1}{12} + \frac{1}{\pi^2(N-1)} \times \sum_{m=1}^{\infty} [NP(m) - 1] \frac{(-1)^m}{m^2}. \qquad (6.2)$$

Note that the summation over m is to be carried out from 1 to ∞, unlike in (3.8). The even parity of $P(m)$ has been exploited in this.

We have still to see whether W_s is 0 for the *fundamental* reflections, as is β_s. From (6.1), (3.8), and (4.1),

$$W_s = \tfrac{1}{12} + \frac{1}{\pi^2(N-1)} \sum_{m=1}^{\infty} \left\{ [NP(m) - 1] + [1 - P(m)]\, N \frac{|\langle F \rangle|^2}{\langle |F|^2 \rangle} \right\} \frac{(-1)^m}{m^2}. \qquad (6.3)$$

For many fundamental reflections, for example for all those of Cu_3Au, and for part of those of substituted spinels, $|\langle F \rangle|^2 = \langle |F|^2 \rangle$. In such cases, from (6.3),

$$W_s\,(\text{fundamental reflections}) = \tfrac{1}{12} + \frac{1}{\pi^2} \sum_{m=1}^{\infty} \frac{(-1)^m}{m^2} = 0. \qquad (6.4)$$

For those fundamental reflections for which $|\langle F \rangle|^2 \neq \langle |F|^2 \rangle$, W_s will not be strictly 0, but will still be very small, as brief consideration will show. Thus it cannot be obtained with any acceptable precision from the variance of the experimental line profile, in which the term due to the so-called instrumental profile overwhelmingly predominates in this case over the term due to the disorder.

Thus disorder may be investigated through the determination of the variance of forbidden, but not of fundamental, reflections. This statement corroborates the same conclusion of Szabó (1975*a*), arrived at in a less general treatment. In opposition to this, Grimes (1968) asserted the broadening of certain fundamental reflections, and suggested the investigation of disorder through the variance of the latter. Based on this, Grimes, Hilleard, Waters & Yerkess (1968) performed such determinations. Both the theoretical assertion of Grimes as well as the experimental work of Grimes *et al.* have been refuted by Szabó (1975*a*,*b*) in a less general type of treatment. This refutation has now been corroborated.

Moments higher than the second would, of course, give further information on $q(M)$. It is easy to show that, similarly to the integral breadth and to the variance, for the higher moments, too, only the forbidden reflections may come into consideration. However, their determination with the necessary precision would be extremely difficult. It seems very improbable to be able, at the present, to make practical use of them.

7. On some assumptions used in our derivations

7.1. *Crystals with different domain patterns*

In the particle-size problem, Laue (1926) assumed a common crystal shape and size in the polycrystalline sample. The same stringent assumption was implied *tacitly* by Bouman & Wolff (1942), and by Stokes & Wilson (1942, 1944). The averages occurring there do not refer to different crystals but to the distribution of a linear dimension within their common shape and size.

The effect of a distribution of shapes and sizes was taken into account by Waller (1939). But there the *tacit* assumption was made that crystals belonging to every one shape and size occur uniformly distributed over all the spatial orientations, which is still a rather stringent assumption.

The assumption which is implicit in our formulae is that the distribution $q(M)$ be the same for every crystal of the sample. This condition is not so stringent as the former. Namely, it does not require every crystal to have the same domain pattern. For crystals large enough, as they have been supposed to be, *i.e.* containing a great number of domains, its fulfilment is evidently possible and probable even for very different domain patterns in the different crystals.

7.2. *Fluctuating cell composition*

We have still to consider the frequent case when the numbers of the different atoms in the same set of equivalent positions are not the same in every unit cell, as assumed in our calculations, but vary randomly between some limits. The number N_0' defined in §2 then means an average and its value will be a fractional number. This problem was discussed by Szabó (1975*b*). The linear interpolation suggested there evidently holds also for the present calculations

8. Conclusions and summary

Our formulae and considerations may form the basis for the investigation of the distribution of the sizes of ordered domains in substitutionally disordered crystals. The experimental determination of this distribution would give an insight into the formation of these domains, into the relative importance of nucleation and growth. Until now, no adequate diffraction theory existed for this.

The relation between polycrystalline diffracted intensity profile and domain-size distribution has been derived, and the centroid displacement, the integral breadth, and the variance calculated.

(1) The centroid displacement has been shown to be 0 for *all* reflections.

(2) The integral breadth of the forbidden reflections has been shown to be a suitable parameter for the domain-size distribution, in contrast to the fundamental reflections, the integral breadth of which always vanishes.

(3) The variance of the forbidden, but not of the fundamental, reflections, determined in addition to the integral breadth, may give a second parameter for the domain-size distribution. It is urged that such measurements be carried out, at least in some simple cases, in view of the important knowledge to be gained.

(4) Detailed integral breadth calculations have been carried out for three models of the size distribution. An estimate has been given for the error in the determination of Λ if the distribution is of the Lifschitz type but its exact form is not known.

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