

rotation function to attain interpretable results and, thus, provides a significant saving of computation time. Furthermore, an accurate solution can be obtained from a reasonably small number of strong reflections using relatively low-resolution data.

Dr S. Krishnaswamy carried out initial tests on a locked rotation function generated by point-group-specific modifications to the laboratory's standard program. These he showed to be highly successful on HRV14 and Mengo virus and provided the incentive for the study reported here. We are most grateful for and encouraged by his initial work. We thank Robert McKenna and Peter Willingmann for providing the φ X174 diffraction data before publication, Walter Keller and Kathy Smith for the tetragonal CPV data and Sung-Hou Kim for permission to use the monellin data and coordinates. We also thank Helene Prongay and Sharon Wilder for help in the preparation of this manuscript. The work was supported by grants to MGR from the National Science Foundation and the National Institutes of Health.

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Diffraction Theory for Diffuse Scattering by Correlated Microdomains in Materials with Several Atoms per Unit Cell

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Abstract

A general diffraction theory is presented for the diffuse scattering by correlated microdomains within a disordered structure. The theory applies to crystals with several atoms per unit cell and several types of different microdomains. An analytical expression is given for an assumed distribution function of the microdomains within the disordered matrix of the host. Since the analytical Fourier transform of this distribution function is also given, very fast calculation of the diffuse intensity is possible.

Introduction

Quantitative interpretations of diffuse scattering are frequently done on the basis of the Warren short-range-order (SRO) parameters. In the case of simple alloy structures, good agreement of calculated and observed intensities is obtained. Hayakawa & Cohen (1975) presented a generalized solution for structures with several sublattices. Any description by SRO parameters, however, will not yield information on the actual distribution of the defects. Extended defects cannot be described. In particular, if the distri-

bution of the defects within the crystal shows short-range order, a description with SRO parameters will give a misleading picture of the true defect structure.

The theory described in this paper assumes the following model. The host crystal is comprised of one or several atoms per unit cell. Each of these sites may be occupied by one or several different chemical elements and/or vacancies. The distribution of these elements on a specific site is assumed to be random. No displacements from the average structure are assumed to exist within the host crystal. Small domains, called microdomains, are coherently intergrown within the crystal, *i.e.* the lattice of the crystal is undisturbed. The structure of these microdomains may be an ordered superstructure of the host crystal or a completely new structure with the same lattice. An ordered superstructure may include substitutional order as well as displacements from the positions of the average structure. The displacements are assumed to be small enough for an expansion of the exponential function to be valid. Several different types of microdomains may be present and no restrictions apply to the size of the microdomains. The distribution of the microdomains within the host crystal shows short-range order. Fig. 1 shows schematically the distribution of microdomains within the matrix of a crystal.

Hashimoto (1974) developed a theory to describe the diffuse scattering by a distribution of correlated microdomains in binary alloys. The SRO parameters for the single pairs of atoms are replaced by three parameters: a phase factor that describes the type of order of the specific microdomain, a shape function that describes the extent and form of the microdomains and a distribution function that describes the correlation between the microdomains. The definition of the phase factor restricts the theory to binary alloys with one kind of microdomain. No attempt has been made to give an analytical description of the distribution function permitting a direct Fourier transformation.

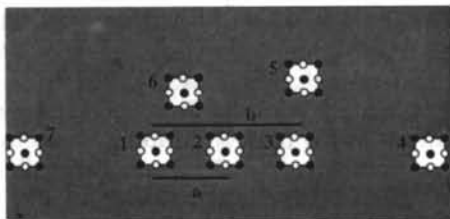


Fig. 1. Schematic distribution of microdomains within a disordered matrix. Modified after Hashimoto (1974). Microdomains 1, 2 and 3 lie correlated within the crystal, while the other microdomains are distributed at random. The distance a is the average separation of the microdomains. The length of coherence, b , is the average length over which the microdomains are distributed at constant spacing.

In this paper the theory by Hashimoto (1974) is extended to general structures. Instead of the phase factor the structure factor of each microdomain is explicitly calculated. This allows for any number of types of microdomain in a structure with several atoms per unit cell. An analytical model for the distribution function is based on the model for a paracrystal as described by Hosemann (1951; Hosemann & Bagchi, 1952). For this distribution function a Fourier transform is presented, which permits direct calculation of the intensity distribution in reciprocal space.

General theory

The amplitude scattered by a crystal with several atoms per unit cell is given by

$$A(\mathbf{h}) = \sum_K \sum_i (\bar{f}_K + \Delta f_{iK}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}_{iK}), \quad (1)$$

where K = index of site in the unit cell, i = index of unit cell of the average structure, \bar{f}_K = average atomic scattering factor of atoms on the site K including the Debye-Waller factor, Δf_{iK} = deviation from the average atomic scattering factor at the site K in unit cell i , \mathbf{h} = scattering vector, \mathbf{r}_{iK} = vector from the origin to the atom i (actual position, *i.e.* displacements from ideal positions are included).

This general expression includes substitutional disorder and displacement disorder.

The intensity is given by the square of the amplitude:

$$I(\mathbf{h}) = \sum_K \sum_i (\bar{f}_K + \Delta f_{iK}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}_{iK}) \times \sum_L \sum_j (\bar{f}_L^* + \Delta f_{jL}^*) \exp(+2\pi i \mathbf{h} \cdot \mathbf{r}_{jL}). \quad (2)$$

The multiplication can be carried out and the terms be sorted according to the various products of f :

$$\begin{aligned} I(\mathbf{h}) &= I_1 + I_2 + I_3 + I_4 \\ &= \sum_K \sum_L \sum_i \sum_j \bar{f}_K \bar{f}_L^* \exp[-2\pi i \mathbf{h} \cdot (\mathbf{r}_{iK} - \mathbf{r}_{jL})] \\ &\quad + \sum_K \sum_L \sum_i \sum_j \bar{f}_K \Delta f_{jL}^* \exp[-2\pi i \mathbf{h} \cdot (\mathbf{r}_{iK} - \mathbf{r}_{jL})] \\ &\quad + \sum_K \sum_L \sum_i \sum_j \Delta f_{iK} \bar{f}_L^* \exp[-2\pi i \mathbf{h} \cdot (\mathbf{r}_{iK} - \mathbf{r}_{jL})] \\ &\quad + \sum_K \sum_L \sum_i \sum_j \Delta f_{iK} \Delta f_{jL}^* \exp[-2\pi i \mathbf{h} \cdot (\mathbf{r}_{iK} - \mathbf{r}_{jL})]. \end{aligned} \quad (3)$$

The first sum I_1 includes the intensity of the Bragg reflections. Since \mathbf{r}_{iK} and \mathbf{r}_{jL} deviate from the ideal positions only for the atoms within the microdomains, I_1 also includes a weak contribution of diffuse scattering due to the displacement from the averaged structure. This contribution, which has been described by Cowley (1981) and Schwartz & Cohen (1977), is neglected in the present approximation. $I_2 + I_3$ average approximately to zero, see Appendix A. The

fourth sum I_4 describes the scattering by short-range order and will be denoted by I_{SRO} . Within this sum it can be distinguished whether an atom is within or outside a microdomain:

$$\begin{aligned}
 I_{\text{SRO}}(\mathbf{h}) &= I_{\text{SRO},1} + I_{\text{SRO},2} + I_{\text{SRO},3} + I_{\text{SRO},4} \\
 &= \sum_K \sum_L \sum_m \sum_n \Delta f_{mK} \Delta f_{nL}^* \\
 &\quad \times \exp[-2\pi i \mathbf{h}(\mathbf{r}_{mK} - \mathbf{r}_{nL})] \\
 &\quad + \sum_K \sum_L \sum_o \sum_n \Delta f_{oK} \Delta f_{nL}^* \\
 &\quad \times \exp[-2\pi i \mathbf{h}(\mathbf{r}_{oK} - \mathbf{r}_{nL})] \\
 &\quad + \sum_K \sum_L \sum_m \sum_p \Delta f_{mK} \Delta f_{pL}^* \\
 &\quad \times \exp[-2\pi i \mathbf{h}(\mathbf{r}_{mK} - \mathbf{r}_{pL})] \\
 &\quad + \sum_K \sum_L \sum_o \sum_p \Delta f_{oK} \Delta f_{pL}^* \\
 &\quad \times \exp[-2\pi i \mathbf{h}(\mathbf{r}_{oK} - \mathbf{r}_{pL})]. \quad (4)
 \end{aligned}$$

The subscripts m and n in (4) describe atoms that do not lie in a microdomain, while the subscripts o and p describe atoms that lie within a microdomain. The first sum is the monotonic Laue scattering due to a random distribution of point defects. The next two sums average to zero since no correlation exists between atoms within a microdomain and outside, see Appendix B. The fourth sum describes the contribution to diffuse scattering by correlated microdomains and will be denoted I_{MD} . At this point, the center of a microdomain is defined to be a lattice point within the microdomain. With this definition, the vector from the origin to the atoms is now separated into a vector from the origin to the center of the microdomain and a vector from the center of the microdomain to the atom: $\mathbf{r}_{oK} = \mathbf{R}_u + \mathbf{r}'_{Kou}$. Thus, $I_{\text{SRO},4}$ in (4) can be split into a sum over all microdomains u, v and a sum over all atoms within the microdomain:

$$\begin{aligned}
 I_{\text{MD}}(\mathbf{h}) &= \sum_u \sum_v \sum_K \sum_L \sum_o \sum_p \Delta f_{Kou} \Delta f_{Lpv}^* \\
 &\quad \times \exp\{-2\pi i \mathbf{h}[(\mathbf{R}_u + \mathbf{r}'_{Kou}) - (\mathbf{R}_v + \mathbf{r}'_{Lpv})]\}. \quad (5)
 \end{aligned}$$

Equation (5) can be written as

$$\begin{aligned}
 I_{\text{MD}}(\mathbf{h}) &= \sum_u \sum_v \left[\sum_K \sum_o \Delta f_{Kou} \exp(-2\pi i \mathbf{h} \mathbf{r}'_{Kou}) \right] \\
 &\quad \times \left[\sum_L \sum_p \Delta f_{Lpv}^* \exp(+2\pi i \mathbf{h} \mathbf{r}'_{Lpv}) \right] \\
 &\quad \times \exp[-2\pi i \mathbf{h}(\mathbf{R}_u - \mathbf{R}_v)]. \quad (6)
 \end{aligned}$$

The terms in square brackets describe the deviation of the structure factor of the microdomains from the structure factor of the average structure and will be abbreviated to F_u or F_v . Thus, (6) describes the diffuse

scattering by a distribution of microdomains:

$$I_{\text{MD}}(\mathbf{h}) = \sum_u \sum_v F_u F_v^* \exp[-2\pi i \mathbf{h}(\mathbf{R}_u - \mathbf{R}_v)]. \quad (7)$$

For all microdomains of one type the structure factor F_u is now denoted by the structure factor of this type F_t . The sums over u and v will then be separated into a sum over all types of microdomains t and s and a sum over all microdomains u' of type t and v' of type s , respectively:

$$I_{\text{MD}}(\mathbf{h}) = \sum_j \sum_s F_t F_s^* \sum_{u'v'} \exp[-2\pi i \mathbf{h}(\mathbf{R}_{u'} - \mathbf{R}_{v'})]. \quad (8)$$

Since the exact distribution of microdomains is unknown, a distribution function must be assumed. With $\mathbf{R}_1 = \mathbf{R}_{u'} - \mathbf{R}_{v'}$, the explicit sum over all microdomains can be replaced by a sum over all vectors \mathbf{R}_1 . The distribution function $P'_{ts}(\mathbf{R}_1)$ is equal to one if \mathbf{R}_1 connects two microdomains of type t and s , and otherwise zero. Thus, the value of $P'_{ts}(\mathbf{R}_1)$ explicitly depends on the length and direction of \mathbf{R}_1 as well as its origin:

$$I_{\text{MD}}(\mathbf{h}) = \sum_t \sum_s F_t F_s^* \sum_l P'_{ts}(\mathbf{R}_l) \exp(-2\pi i \mathbf{h} \mathbf{R}_l). \quad (9)$$

In (9) the sum over l includes all vectors within the crystal between any pair of lattice sites. This sum can be separated into a sum over all free vectors l' of a given direction and length and an inner sum over all the origins j of the vectors of direction and length given by l' :

$$\begin{aligned}
 I_{\text{MD}}(\mathbf{h}) &= \sum_t \sum_s F_t F_s^* \sum_{l'} \left[\sum_j P'_{ts}(\mathbf{R}_{l'j}) \exp(-2\pi i \mathbf{h} \mathbf{R}_{l'j}) \right] \\
 &= \sum_t \sum_s F_t F_s^* \sum_{l'} \left[\sum_j P'_{ts}(\mathbf{R}_{l'j}) \right] \exp(-2\pi i \mathbf{h} \mathbf{R}_{l'}). \quad (10)
 \end{aligned}$$

Since $\mathbf{R}_{l'j}$ is identical for all j , the exponential term in (10) can be taken out of the summation over j . The explicit distribution function $P'_{ts}(\mathbf{R}_{l'j})$ can now be approximated by the average distribution function $P_{ts}(\mathbf{R}_{l'})$ for a given free vector l' . For $\mathbf{R}_{l'} = 0$, the distribution function is equal to δ_{ts} and thus the sum over j equals $N_t \delta_{ts}$ where N_t is the number of microdomains of type t in the crystal. For $\mathbf{R}_{l'} \neq 0$ the sum over j can be calculated exactly for two extreme situations, perfect periodic distribution and random distribution. For a perfect periodic distribution the sum over j is equal to N_t or 0, depending upon whether $\mathbf{R}_{l'}$ is a superlattice vector for the perfect periodic distribution or not. For a random distribution of microdomains the probability of finding another microdomain N of type s is $N_s v / V$, where N_s is the number of microdomains of type s , V the volume of the crystal and v the volume of a microdomain. Thus, the sum over j is equal to $N_t N_s v / V$. For a distribution with short-range order the probabil-

ity to find a microdomain oscillates around $N_s v/V$ and the value of the sum over j oscillates around $N_t N_s v/V$. The oscillation around $N_s v/V$ is described by the average distribution function $P_{is}(\mathbf{R}_l)$, and thus (10) is

$$I_{\text{MD}}(\mathbf{h}) = \sum_t \sum_s F_t F_s^* N_t \left[\delta_{ts} + \sum_{l'(\mathbf{R}_{l'} \neq 0)} P_{is}(\mathbf{R}_{l'}) \times \exp(-2\pi i \mathbf{h} \mathbf{R}_{l'}) \right]. \quad (11)$$

Now the sum over l' describes only all free lattice vectors. In (11) the term

$$\sum_{l'} P_{is}(\mathbf{R}_{l'}) \exp(-2\pi i \mathbf{h} \mathbf{R}_{l'}) \quad (12)$$

describes the Fourier transformation of the distribution function of the microdomains. An analytical solution of this Fourier transformation, which is based on the model of a paracrystal, is given in the next section.

Fourier transformation of the distribution function

Hosemann (1951; Hosemann & Bagchi, 1952) introduced the concept of a paracrystal. In this model the position of the next neighbor of an atom is described by a continuous distribution function $H_1(\mathbf{r})$. The distribution of the second neighbor is given by the self-convolution of $H_1(\mathbf{r})$. The distribution of all neighbors is then given by the geometric sum over all self convolutions:

$$P(\mathbf{r}) = H_0 + \sum_m (\tilde{H}_1^m + \tilde{H}_1^{m-1}). \quad (13)$$

Here \tilde{H}_1^m denotes the m th self convolution of H_1 and H_0 is the δ function. Fig. 2 shows the first two parts of the distribution function for a one-dimensional distribution. Vainshtein (1966) described in detail the Fourier transformation of this distribution function $P(\mathbf{r})$. For a one-dimensional distribution the Fourier

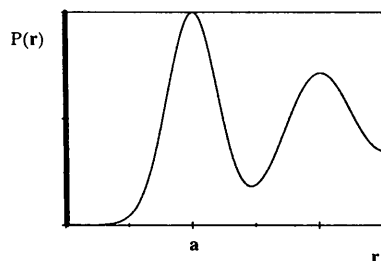


Fig. 2. Distribution function $P(\mathbf{r}) = H_0(\mathbf{r}) + H_1(\mathbf{r}) * H_1(\mathbf{r})$. The distribution of the first and second neighbors. The distribution of the second neighbors is given by the self convolution of the distribution of the first neighbors.

transform is

$$Z(\mathbf{h}) = \frac{1 + |E|^2}{1 - 2|E| \cos(2\pi \mathbf{h} \bar{a}) + |E|^2}, \quad (14)$$

where \bar{a} is the average distance of the first neighbor and $|E|$ the modulus of the Fourier transform of $H_1(\mathbf{r})$. In the simplest approach, the transform of a three-dimensional distribution is given by the product of three components.

To apply this model to the distribution of microdomains in a crystal, it has to be taken into account that the vector connecting two microdomains cannot be of any length. Rather, the vector is limited to lattice vectors. The easiest way to describe this limitation is to multiply (13) with a lattice function. The continuous distribution H_1 is then replaced by a discontinuous distribution:

$$H'_1 = \sum_n H_1 \delta(x - nR_0), \quad (15)$$

where R_0 gives the separation of lattice points, *i.e.* the length of the unit cell. The average distance of the first neighbour, \bar{a} in (14), is $\bar{a} = qR_0$, where q does not need to be an integer number. Fig. 3 shows this discontinuous distribution. The distribution H'_2 of the second neighbor follows from the self convolution of H'_1 :

$$\begin{aligned} H'_2 &= H'_1 * H'_1 \\ &= \left[\sum_n H_1(x) \delta(x - nR_0) \right] * \left[\sum_m H_1(x) \delta(x - mR_0) \right] \\ &= \sum_n \sum_m \int H_1(U) H_1(x - U) \\ &\quad \times \delta(U - nR_0) \delta(x - U - mR_0) dU. \end{aligned} \quad (16)$$

The terms in the integral can be sorted to yield

$$\begin{aligned} H'_2 &= \sum_n \sum_m \int [H_1(U) H_1(x - U) \delta(x - U - mR_0)] \\ &\quad \times \delta(U - nR_0) dU. \end{aligned} \quad (17)$$

Since $\int g(t) \delta(t - b) = g(b)$, the integration of the term in square brackets with the δ function can be

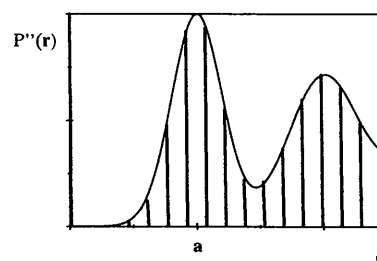


Fig. 3. Discontinuous distribution function $P''(\mathbf{r})$. A one-dimensional lattice has been multiplied with the continuous distribution of Fig. 2. The probability of finding a microdomain on a given site is determined by the height of the distribution function.

evaluated as

$$H'_2 = \sum_n \sum_m H_1(nR_0) H_1(x - nR_0) \delta[x - (n+m)R_0]. \quad (18)$$

The subscript m can be replaced by a subscript k with $k = n + m$ and thus $m = k - n$. The summations over n and m run from minus infinity to plus infinity. Thus, the sum over k runs from minus infinity to plus infinity as well and is independent of n :

$$H'_2 = \sum_n \sum_k H_1(nR_0) H_1(x - nR_0) \delta(x - kR_0). \quad (19)$$

The product of the two sums n and k can be written separately again:

$$H'_2 = \left[\sum_n H_1(nR_0) H_1(x - nR_0) \right] \sum_k \delta(x - kR_0). \quad (20)$$

After multiplying this equation by unity in the form of R_0/R_0 , the sum over n yields a good approximation for an integral:

$$H'_2 = 1/R_0 \left[\sum_n H_1(nR_0) H_1(x - nR_0) R_0 \right] \sum_k \delta(x - kR_0) \\ \approx 1/R_0 \left[\int H_1(U) H_1(x - U) dU \right] \sum_k \delta(x - kR_0). \quad (21)$$

The approximation of the sum over n by the integral is very close if R_0 is small compared to the FWHM of H_1 . This integral is the self convolution of the continuous distribution H_1 :

$$H'_2 = 1/R_0 [H_1 * H_1] \sum_k \delta(x - kR_0). \quad (22)$$

For all further convolutions the same algorithm holds. The complete discontinuous distribution function follows as the sum of the self convolutions of the continuous distribution multiplied by the lattice function:

$$P''(x) = \left[H_0 + \sum_m (1/R_0)^{m-1} (\tilde{H}_1^m + \tilde{H}_{-1}^m) \right] \\ \times \sum_n \delta(x - nR_0) \\ = \left[H_0 + R_0 \sum_m (1/R_0)^m (\tilde{H}_1^m + \tilde{H}_{-1}^m) \right] \\ \times \sum_n \delta(x - nR_0). \quad (23)$$

The Fourier transformation of this equation is the convolution of the correlation function Z and the reciprocal lattice G^* :

$$\mathcal{F}[P''(x)] = Z(h) * G^* \\ = \left[1 + R_0 \sum_m (1/R_0)^m (E_1^m + E_{-1}^{*m}) \right] \\ * \mathcal{F} \left[\sum_n \delta(x - nR_0) \right] \\ = \left[1 + R_0 \frac{E/R_0}{1 - E/R_0} + \frac{E^*/R_0}{1 - E^*/R_0} \right] \\ * \mathcal{F} \left[\sum_n \delta(x - nR_0) \right] \\ = \{ [1 + 2|E| \cos(2\pi x \bar{a}) (R_0 - 1)/R_0 \\ + |E|^2/R_0^2 (1 - 2R_0)] \\ \times [1 - 2|E| \cos(2\pi x \bar{a}) + |E|^2/R_0]^{-1} \} \\ * \mathcal{F} \left[\sum_n \delta(x - nR_0) \right]. \quad (24)$$

Here, \mathcal{F} symbolizes the Fourier transform and E is the Fourier transform of the continuous distribution H_1 . With this result (11) yields

$$I_{MD}(\mathbf{h}) = \sum_t \sum_s F_t F_s^* N_t \{ \delta_{ts} \\ + [Z_{ts}(h_1) Z_{ts}(h_2) Z_{ts}(h_3) * G^*] \}. \quad (25)$$

In (25) the correlation function Z_{ts} is a different function for all pairs of microdomain types t and s based on the distribution function P_{ts} as given by (11) and (12). In most cases these different distribution functions can be approximated by one distribution function Z' that is identical for all pairs of microdomain types t and s . In order to determine whether two microdomain types t and s are correlated at all, a function C_{ts} will be introduced. This function is one if correlation between the types t and s is allowed and otherwise zero. Thus (25) will be

$$I_{MD}(\mathbf{h}) = \sum_t \sum_s F_t F_s^* N_t \{ \delta_{ts} \\ + C_{ts} [Z'(h_1) Z'(h_2) Z'(h_3) * G^*] \}. \quad (26)$$

To allow fast calculation of the diffuse intensity a suitable function H_1 should be chosen. Most straight forward solutions will be obtained by Gaussian or Lorentzian distributions.

Distribution function for Gaussian distribution

The Fourier transform of the distribution in the case of a Gaussian distribution will be derived. The integral over the distribution H_1 must be unity so that a normalized distribution results:

$$H_1(x) = (2\pi)^{-1/2} \sigma^{-1} \exp[-\frac{1}{2}(x - \bar{a})^2/\sigma^2]. \quad (27)$$

The modulus of the Fourier transform is given by

$$|E| = \exp[-2\pi(x^2\sigma^2)]. \quad (28)$$

The m th self convolution of H_1 and its Fourier transform are:

$$\tilde{H}_1^m(x) = (2\pi)^{-1/2} \sigma^{-1} m^{-1/2} \times \exp[-\frac{1}{2}(x - m\bar{a})^2 / \sigma^2 m], \quad (29)$$

$$|E_1^m| = \exp[-2\pi(x^2 \sigma^2) m]. \quad (30)$$

These formulas can easily be expanded for three-dimensional distributions:

$$H_{11}(\mathbf{x}) = (2\pi)^{-3/2} (\sigma_{11} \sigma_{12} \sigma_{13})^{-1} \times \exp\{-\frac{1}{2}[(x_1 - \bar{a}_1)^2 / \sigma_{11}^2 + (x_2 - \bar{a}_2)^2 / \sigma_{12}^2 + (x_3 - \bar{a}_3)^2 / \sigma_{13}^2]\} \quad (31)$$

$$|E_{11}| = \exp[-2\pi^2(x_1^2 \sigma_{11}^2 + x_2^2 \sigma_{12}^2 + x_3^2 \sigma_{13}^2)] \quad (32)$$

Here H_{11} is the x component of the distribution function and E_{11} the x component of the Fourier transform. In analogy to (31) and (32), the functions H_{12} , H_{13} and E_{12} , E_{13} are defined. The matrix σ_{ij} of the displacement coefficients describes the FWHM of the distribution function in three-dimensional space.

Fig. 4 shows the continuous distribution function P for various values of σ . The length of coherence is the length at which the FWHM of the peaks is equal to the separation of the peaks. At this length the distribution function is almost constant. The argument of the exponential function in (29) has to be equal to $\ln(0.5)$ for $(x - ma) = \bar{a}/2$. Thus the critical value of m will be

$$M = -1/[8 \ln(0.5)(\sigma/\bar{a})^2]. \quad (33)$$

The length of coherence is the product of the average distance \bar{a} and M .

Concluding remarks

The theory presented in this paper expands the theory given by Hashimoto (1974) to materials with several atoms per unit cell. Each microdomain is described

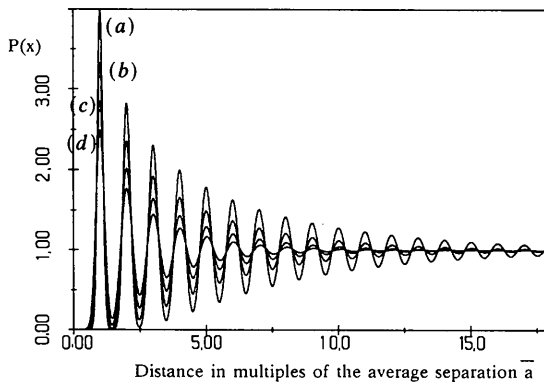


Fig. 4. Distribution function $P(x)$ for a Gaussian distribution. The distribution is shown for different values of σ/a : (a) $\sigma/a = 0.10$, (b) $\sigma/a = 0.12$, (c) $\sigma/a = 0.14$, (d) $\sigma/a = 0.16$.

by the deviation of its structure factor from the structure factor of the average structure. The correlation between the microdomains is described by a discontinuous distribution function. Following Hosemann (1951) and Hosemann & Bagchi (1952) the analytical Fourier transformation of this distribution function is developed. Therefore, fast calculation of intensities is made possible, which allows for quantitative analysis of diffuse scattering by disordered structures.

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APPENDIX A

It is shown here that the two sums $I_2 + I_3$ in (3) average approximately to zero. The two sums are

$$I_{2,3}(\mathbf{h}) = I_2 + I_3 = \sum_K \sum_L \sum_i \sum_j \bar{f}_K \Delta f_{jL}^* \exp[-2\pi i \mathbf{h}(\mathbf{r}_{iK} - \mathbf{r}_{jL})] + \sum_K \sum_L \sum_i \sum_j \Delta f_{iK} \bar{f}_L^* \exp[-2\pi i \mathbf{h}(\mathbf{r}_{iK} - \mathbf{r}_{jL})]. \quad (A1)$$

The vector \mathbf{r}_{iK} can be expressed by the sum of the vector to the ideal position plus a deviation from this position: $\mathbf{r}_{iK} = \mathbf{R}_{iK} + \Delta \mathbf{r}_{iK}$. Thus (A1) can be written as

$$I_{2,3}(\mathbf{h}) = \sum_K \sum_L \sum_i \sum_j \bar{f}_K \Delta f_{jL}^* \times \exp[-2\pi i \mathbf{h}(\mathbf{R}_{iK} - \mathbf{R}_{jL})] \times \exp[-2\pi i \mathbf{h}(\Delta \mathbf{r}_{iK} - \Delta \mathbf{r}_{jL})] + \sum_K \sum_L \sum_i \sum_j \Delta f_{iK} \bar{f}_L^* \times \exp[-2\pi i \mathbf{h}(\mathbf{R}_{iK} - \mathbf{R}_{jL})] \times \exp[-2\pi i \mathbf{h}(\Delta \mathbf{r}_{iK} - \Delta \mathbf{r}_{jL})]. \quad (A2)$$

Both sums can be sorted into groups of atoms for which $\mathbf{R}_{iK} - \mathbf{R}_{jL}$ is identical. Within each of these groups the exponential function is identical and can be taken out of the sums. For the sake of clarity the outer sum over these groups of atoms will be omitted. The inner sum over all atoms of one group is thus

$$I_{2,3}^g(\mathbf{h}) = \sum_K \sum_L \sum_i \sum_j \bar{f}_K \Delta f_{jL}^* \times \exp[-2\pi i \mathbf{h}(\Delta \mathbf{r}_{iK} - \Delta \mathbf{r}_{jL})] + \sum_K \sum_L \sum_i \sum_j \Delta f_{iK} \bar{f}_L^* \times \exp[-2\pi i \mathbf{h}(\Delta \mathbf{r}_{iK} - \Delta \mathbf{r}_{jL})]. \quad (A3)$$

These sums can further be sorted into a group of pairs with both atoms outside any microdomain and a group with at least one atom within a microdomain. Within the first group, all atoms occupy ideal lattice positions. Therefore, the exponential term is unity.

Since the sum over all Δf is zero:

$$\sum_K \sum_i \Delta f_{iK} = \{(m_{AK}m_{BK}) - (m_{BK}m_{AK})\}(f_{AK} - f_{BK}) = 0, \quad (A4)$$

the contribution of this first group is zero. Here m_{AK} and m_{BK} are the relative occupations of the site K by atoms A and B , respectively. The argument of the exponential function is abbreviated to $\varepsilon_{ijKL} = 2\pi\mathbf{h} \times (\Delta\mathbf{r}_{iK} - \Delta\mathbf{r}_{jL})$. Since deviations from ideal lattice positions are small, the exponential function can be expanded into a series up to second-order terms:

$$I'_{2,3} = \sum_K \sum_L \sum_i \sum_j \bar{f}_K \Delta f_{jL}^* (1 - i\varepsilon_{ijKL} - \frac{1}{2}\varepsilon_{ijKL}^2) + \sum_K \sum_L \sum_i \sum_j \Delta f_{iK} \bar{f}_L^* (1 - i\varepsilon_{ijKL} - \frac{1}{2}\varepsilon_{ijKL}^2). \quad (A5)$$

The constant term averages to zero, due to (A4). Since $\varepsilon_{ijKL} = -\varepsilon_{jiLK}$, the linear terms add up to zero, if anomalous dispersion can be neglected. The remaining sum can be grouped into a group with exactly one atom within a microdomain and a group with both atoms within a microdomain. The first group can further be sorted into groups with ε constant. For these groups (A4) holds so that their contribution is zero. The only significant part left is the group with both atoms within a microdomain. For these pairs ε and Δf are correlated. It cannot be expected that their contribution will average to zero. Since ε is small, this quadratic contribution of $I_2 + I_3$ to (3) can be neglected.

APPENDIX B

The terms $I_{\text{SRO},2}$ and $I_{\text{SRO},3}$ in (4) describe the contribution of pairs with one atom inside a microdomain and the other outside a microdomain to the scattered intensity. It will be shown that these contributions average to zero. The two terms are

$$I_{\text{SRO},2,3}(\mathbf{h}) = I_{\text{SRO},2} + I_{\text{SRO},3} = \sum_K \sum_L \sum_o \sum_n \Delta f_{oK} \Delta f_{nL}^* \times \exp[-2\pi i \mathbf{h}(\mathbf{r}_{oK} - \mathbf{r}_{nL})] + \sum_K \sum_L \sum_m \sum_p \Delta f_{mK} \Delta f_{pL}^* \times \exp[-2\pi i \mathbf{h}(\mathbf{r}_{mK} - \mathbf{r}_{pL})]. \quad (B1)$$

The vector \mathbf{r}_{iK} can be expressed by the sum of the vector to the ideal position plus a deviation from this

position: $\mathbf{r}_{iK} = \mathbf{R}_{iK} + \Delta\mathbf{r}_{iK}$. Thus (B1) can be written as

$$I_{\text{SRO},2,3} = \sum_K \sum_L \sum_o \sum_n \Delta f_{oK} \Delta f_{nL}^* \times \exp[-2\pi i \mathbf{h}(\mathbf{R}_{oK} - \mathbf{R}_{nL})] \times \exp[-2\pi i \mathbf{h}(\Delta\mathbf{r}_{oK})] + \sum_K \sum_L \sum_m \sum_p \Delta f_{mK} \Delta f_{pL}^* \times \exp[-2\pi i \mathbf{h}(\mathbf{R}_{mK} - \mathbf{R}_{pL})] \times \exp[-2\pi i \mathbf{h}(-\Delta\mathbf{r}_{pL})]. \quad (B2)$$

Both sums can be sorted into groups of atoms for which $\mathbf{R}_{iK} - \mathbf{R}_{jL}$ is identical. Within each of these groups the exponential function is identical and can be taken out of the sums. For the sake of clarity the outer sum over these groups of atoms will be omitted. The inner sum over all atoms of one group will thus be

$$I'_{\text{SRO},2,3} = \sum_K \sum_L \sum_o \sum_n \Delta f_{oK} \Delta f_{nL}^* \exp[-2\pi i \mathbf{h}(\Delta\mathbf{r}_{oK})] + \sum_K \sum_L \sum_m \sum_p \Delta f_{mK} \Delta f_{pL}^* \exp[-2\pi i \mathbf{h}(-\Delta\mathbf{r}_{pL})]. \quad (B3)$$

The product of the sums over K and L can be written separately:

$$I'_{\text{SRO},2,3} = \sum_K \sum_o \Delta f_{oK} \exp[-2\pi i \mathbf{h}(\Delta\mathbf{r}_{oK})] \sum_L \sum_n \Delta f_{nL}^* + \sum_K \sum_m \Delta f_{mK} \sum_L \sum_p \Delta f_{pL}^* \times \exp[-2\pi i \mathbf{h}(-\Delta\mathbf{r}_{pL})]. \quad (B4)$$

Owing to the definition of Δf , (A4) holds and the sums over L , n and K , m are zero. Therefore, the contribution of $I_{\text{SRO},2}$ and $I_{\text{SRO},3}$ in (4) is zero.

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