Correlative Microdomain Model for Short-Range-Ordered Alloy Structures. II. Applications to Special Cases

By S. Hashimoto

The Research Institute for Iron, Steel and Other Metals, Tohoku University, Sendai, Japan

Abstract

A general theory of the diffuse scattering which has previously been developed on the basis of a microdomain model [Hashimoto (1974). *Acta Cryst.* A**30**, 792–798] is applied to the three special cases in which the microdomains have the ordered structures denoted by $L1_2$, $L1_0$ and $L1_1$. The practical intensity formula is presented in each case, which can be used to obtain, from the observed X-ray intensity distribution, the statistical nature of the microdomains; the average size, the size distribution, the number density and the spatial interdomain correlation.

1. Introduction

In the disordered state of many alloys, the diffuse scattering of X-rays and electrons is often enhanced around the directions of the superlattice reflections expected in the ordered state. In some cases, for example in Cu–Au alloys, fine structure is noticeable in these enhanced diffuse spots. These observations suggest that there exists an ordering of constituent atoms even in the disordered alloys.

For interpreting these observations, the author developed a diffraction theory based on a microdomain model in the previous paper* (Hashimoto, 1974). The model assumes that relatively well-ordered microdomains are dispersed in the disordered matrix. This is in contrast with the homogeneous model, in which every part of the specimen is assumed to be characterized by a pair correlation of alloyed atoms. Inhomogeneous models similar to the present one were proposed as a realistic model by several investigators, as mentioned in I, and recent electron microscopic observations (Sinclair & Thomas, 1975; Tanaka, Ohshima, Harada & Mihama, 1979) support the model.

The main purpose of the present paper is to derive the formulae of the diffraction intensity for a few specific ordered structures of the microdomains. Also

---

* Hereafter, the paper is referred to as I.

presented are the practical procedures to analyze the observed intensity of diffuse scattering and to obtain information on the statistical nature of the microdomains. The types of ordered structures with which we are concerned here are $L1_2(\mathrm{Cu_3Au})$, $L1_0(\mathrm{CuAu})$ and $L1_1(\mathrm{CuPt})$.

2. Basic equations

The intensities of the diffuse scattering are given in the forms (Hashimoto, 1974)

$$I_{\mathrm{sro}}(\mathbf{q}) = I_{\mathrm{LM}}(\mathbf{q}) + I_{\mathrm{MD}}(\mathbf{q}), \tag{1a}$$

$$I_{\mathrm{LM}}(\mathbf{q}) = n_r\, m_A\, m_B |\, f_A - f_B|^2, \tag{1b}$$

$$I_{\mathrm{MD}}(\mathbf{q}) = I_{\mathrm{MD}}^0(\mathbf{q}) + I_{\mathrm{MD}}^i(\mathbf{q}), \tag{1c}$$

$$I_{\mathrm{MD}}^0(\mathbf{q}) = \frac{m_A\, m_B}{v_0^2} |\, f_A - f_B|^2 \sum_{\mathbf{G}_l} \sum_t \sum_{t'} N_t\, \varepsilon_t^2(\mathbf{q}_l)\, \gamma_{\mathbf{G}_l}^{(tt')}, \tag{1d}$$

$$I_{\mathrm{MD}}^i(\mathbf{q}) = \frac{m_A\, m_B}{v_0^2} |\, f_A - f_B|^2 \sum_{\mathbf{G}_l} \sum_t \sum_{t'} N_t\, \varepsilon_t(\mathbf{q}_l)\, \varepsilon_{t'}(\mathbf{q}_l)$$
$$\times \gamma_{\mathbf{G}_l}^{(tt')}\, p_{tt'}(\mathbf{q}_l), \tag{1e}$$

where

$$\mathbf{q}_l \equiv \mathbf{q} - \mathbf{G}_l, \tag{2}$$

and

$$p_{tt'}(\mathbf{q}_l) = \int P_{tt'}(\mathbf{X}) \exp\left(2\pi i \mathbf{q}_l \cdot \mathbf{X}\right) d\mathbf{X}. \tag{3}$$

The notation employed is the same as in I. The intensities $I_{\mathrm{LM}}(\mathbf{q})$ and $I_{\mathrm{MD}}(\mathbf{q})$ are those due to the disordered matrix and the ordered microdomains, respectively. $I_{\mathrm{MD}}^0(\mathbf{q})$ is the intensity expected if the correlations between the microdomains are neglected and $I_{\mathrm{MD}}^i(\mathbf{q})$ is the term due to the interdomain correlations. The addition of I_{MD}^i is an improvement in the present treatment.

3. Intensity equations for special cases

Before going into details, we indicate how the vector $\mathbf{G}_i$ in $(1d)$ and $(1e)$ is defined from the experimental data.

If the microdomain structure of the short-range-ordered alloy is such that all the atoms are located on the sites of an average lattice, the distribution of diffuse scattering intensity is centrosymmetric and its center is at the position of the superlattice reflection. In this simplest case, the vector $\mathbf{G}_i$ can be defined by the center of the distribution.

In real alloys, however, the constituent atoms have different sizes so that they are displaced differently from the atomic positions of the average lattice, and the intensity distribution is non-symmetric. Borie & Sparks (1964, 1971; Sparks & Borie, 1965) analyzed the effects of atomic size on the intensity distribution and gave a method to subtract the effects from the observed distribution. We assume here that the intensity distribution has already been corrected for the effects by the Borie & Sparks' method and define the vector $\mathbf{G}_i$ by the center of the diffuse maximum.

In the following subsections, we shall derive the intensity equations appropriate for each of the cases in which the microdomains have the $L1_2$, $L1_0$ and $L1_1$ structures.

(a) $L1_2$ internal structure

The microdomains with the $L1_2$ structure have four types of order (see Fig. 5 of I), which are mutually in an antiphase relation. There is no need to differentiate between these four types of microdomains in their average shape, and suffix t of $\varepsilon_t(\mathbf{q})$ ($t = 1$–4), the form factor, can be omitted.

The phase factor $\gamma_{\mathbf{G}_i}^{(tt')}$ between the tth and t'th types of order can be expressed in terms of the so-called step shift vector $\boldsymbol{\xi}_{tt'}$ as

$$\gamma_{\mathbf{G}_i}^{(tt')} = (\tfrac{1}{3}) \exp(2\pi i \mathbf{G}_i . \boldsymbol{\xi}_{tt'}), \qquad (4)$$

according to (I-10) and (I-12). It is easily shown that the vector $\boldsymbol{\xi}_{tt'}$ is identical to one of the following four vectors;

$$\boldsymbol{\xi}_{11} = 0, \quad \boldsymbol{\xi}_{12} = (\tfrac{1}{2}) a[110],$$

$$\boldsymbol{\xi}_{13} = (\tfrac{1}{2}) a[011] \quad \text{and} \quad \boldsymbol{\xi}_{14} = (\tfrac{1}{2}) a[101]. \qquad (5)$$

Similarly, the interdomain correlation functions $P_{tt'}(\mathbf{X})$ or their Fourier transforms $p_{tt'}(\mathbf{q})$ can be reduced to four independent ones, denoted by $P_{1t}(\mathbf{X})$ or $p_{1t}(\mathbf{q})$, respectively.

Thus, from $(1a)$–$(1e)$, the intensities divided by a common factor $nm_A m_B |f_A - f_B|^2$ are given by

$$\alpha(\mathbf{q}) = \alpha^r + \alpha^0(\mathbf{q}) + \alpha^i(\mathbf{q}), \qquad (6a)$$

$$\alpha^r = n_r/n, \qquad (6b)$$

$$\alpha^0(\mathbf{q}) = \frac{N_d}{nv_0^2} \sum_{\mathbf{G}_i} \varepsilon^2(\mathbf{q}_i)\, \gamma_{\mathbf{G}_i}^{(11)}, \qquad (6c)$$

$$\alpha^i(\mathbf{q}) = \frac{N_d}{nv_0^2} \sum_{\mathbf{G}_i} \varepsilon^2(\mathbf{q}_i) \sum_{t=1}^{4} \gamma_{\mathbf{G}_i}^{(1t)}\, p_{1t}(\mathbf{q}_i), \qquad (6d)$$

where $\alpha(\mathbf{q})$, α^r, etc. are expressed in the Laue monotonic units and N_t in (1) is replaced by $N_d/4$, N_d being the total number of the microdomains.

If there is no appreciable overlapping of $\varepsilon(\mathbf{q}_i)$'s belonging to different superlattice reflections, we have, in the vicinity of $\mathbf{G}_i$,

$$\alpha^0(\mathbf{q}_i) = \frac{N_d}{3nv_0^2} \varepsilon^2(\mathbf{q}_i), \qquad (7a)$$

$$\alpha^i(\mathbf{q}_i) = \frac{N_d}{nv_0^2} \varepsilon^2(\mathbf{q}_i) \sum_{t=1}^{4} \gamma_{\mathbf{G}_i}^{(1t)}\, p_{1t}(\mathbf{q}_i), \qquad (7b)$$

where $\alpha^0(\mathbf{q}_i)$ and $\alpha^i(\mathbf{q}_i)$ are defined as a function of the vector deviation from $\mathbf{G}_i$.

It is convenient to define a reduced intensity

$$D(\mathbf{q}_i) \equiv \alpha^i(\mathbf{q}_i)/\alpha^0(\mathbf{q}_i). \qquad (8)$$

Then, from $(7a)$ and $(7b)$, it follows that

$$D(\mathbf{q}_i) = 3 \sum_{t=1}^{4} \gamma_{\mathbf{G}_i}^{(1t)}\, p_{1t}(\mathbf{q}_i) \qquad (9)$$

and its Fourier transform is given by

$$D_{\mathbf{G}_i}(\mathbf{X}) = 3 \sum_{t=1}^{4} \gamma_{\mathbf{G}_i}^{(1t)}\, P_{1t}(\mathbf{X}), \qquad (9')$$

for any interdomain vector $\mathbf{X}$.

For the $L1_2$ structure, there are three independent superlattice reflections, e.g. 100, 010 and 001. Thus, $(9')$ can be written in an explicit form:

$$D_{100}(\mathbf{X}) = P_{11}(\mathbf{X}) - P_{12}(\mathbf{X}) + P_{13}(\mathbf{X}) - P_{14}(\mathbf{X}), \qquad (10a)$$

$$D_{010}(\mathbf{X}) = P_{11}(\mathbf{X}) - P_{12}(\mathbf{X}) - P_{13}(\mathbf{X}) + P_{14}(\mathbf{X}), \qquad (10b)$$

$$D_{001}(\mathbf{X}) = P_{11}(\mathbf{X}) + P_{12}(\mathbf{X}) - P_{13}(\mathbf{X}) - P_{14}(\mathbf{X}), \qquad (10c)$$

with the use of $\gamma_{\mathbf{G}_i}^{(1t)}$ given in Table 1 of I. By solving these equations, any difference between the interdomain correlation functions $P_{1t}(\mathbf{X})$ can be obtained.

(b) $L1_0$ internal structure

As illustrated in Fig. 1, six types of order are possible for the microdomain structure. They can be put into three groups. The constituents of each group have a tetragonal structure with the c axis in common and are in an antiphase relation with one another. Then one can

see an identity $\varepsilon_t(\mathbf{q}_i) = \varepsilon_{t+3}(\mathbf{q}_i)$ ($t = 1$–3). The phase factor $\gamma_{\mathbf{G}_i}^{(tt')}$ vanishes, as shown in Table 1, unless the order types t and t' belong to the same group. From this property of the phase factor, it can be seen that a modulation of the diffuse intensity around any particular $\mathbf{G}_i$ is due to the correlation between the microdomains of the same group.

The intensities can be written as

$$\alpha(\mathbf{q}) = \alpha^r + \alpha^0(\mathbf{q}) + \alpha^i(\mathbf{q}), \qquad (11a)$$

$$\alpha^r = n_r/n, \qquad (11b)$$

$$\alpha^0(\mathbf{q}) = \frac{1}{nv_0^2} \sum_{\mathbf{G}_i} \sum_{t=1}^{3} N_t' \, \varepsilon_t^2(\mathbf{q}_i) \, \gamma_{\mathbf{G}_i}^{(tt)}, \qquad (11c)$$

Table 1. $\gamma_{\mathbf{G}_i}^{(tt')}$ for the case of the $L1_0$ ordered structure

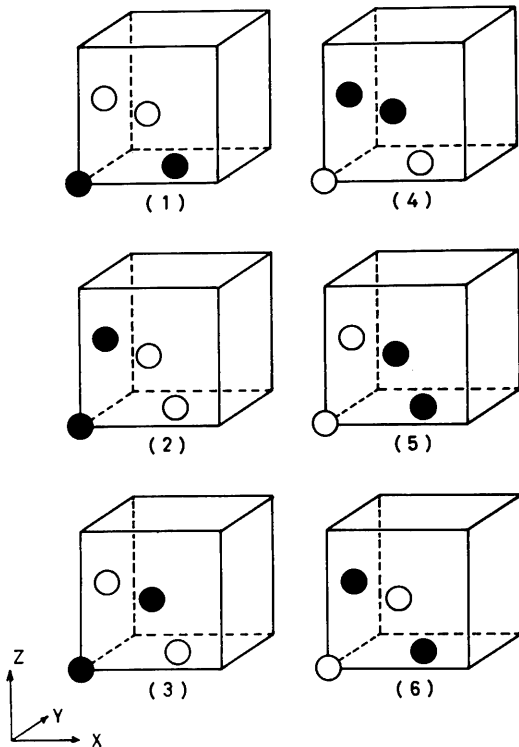|		$\gamma_{\mathbf{G}_i}^{(tt')}$		
t	t'	$\mathbf{G}_{001}$	$\mathbf{G}_{100}$	$\mathbf{G}_{010}$
1	1	1	0	0
	4	−1	0	0
2	2	0	1	0
	5	0	−1	0
3	3	0	0	1
	6	0	0	−1

Fig. 1. Six types of ordered lattice of the $L1_0$ structure. Open and solid circles indicate different kinds of atoms. These are in three groups, which consist of (1) and (4), (2) and (5), and (3) and (6).

$$\alpha^i(\mathbf{q}) = \frac{1}{nv_0^2} \sum_{\mathbf{G}_i} \sum_{t=1}^{3} N_t' \, \varepsilon_t^2(\mathbf{q}_i) \sum_{t'=t,t+3} \gamma_{\mathbf{G}_i}^{(tt')} \, p_{tt'}(\mathbf{q}_i),$$

$$(11d)$$

where $N_t' = N_t + N_{t+3}$. In the case where the overlapping of diffuse scattering belonging to different $\mathbf{G}_i$'s can be neglected, (11c) and (11d) are simplified to

$$\alpha^0(\mathbf{q}_i) = \frac{N_t'}{nv_0^2} \, \varepsilon_t^2(\mathbf{q}_i), \qquad (12a)$$

$$\alpha^i(\mathbf{q}_i) = \frac{N_t'}{nv_0^2} \, \varepsilon_t^2(\mathbf{q}_i) \, \{ p_{tt}(\mathbf{q}_i) - p_{t,t+3}(\mathbf{q}_i) \}. \qquad (12b)$$

Following the arguments described in subsection (a), we have here

$$D_{\mathbf{G}_i}(\mathbf{X}) = P_{tt}(\mathbf{X}) - P_{t,t+3}(\mathbf{X}). \qquad (13)$$

Thus, from the reduced intensity, we can obtain the difference between the spatial correlations of the microdomains belonging to the same group.

(c) $L1_1$ internal structure

In this case, there are eight types of microdomain structures, as illustrated in Fig. 2, which can be grouped
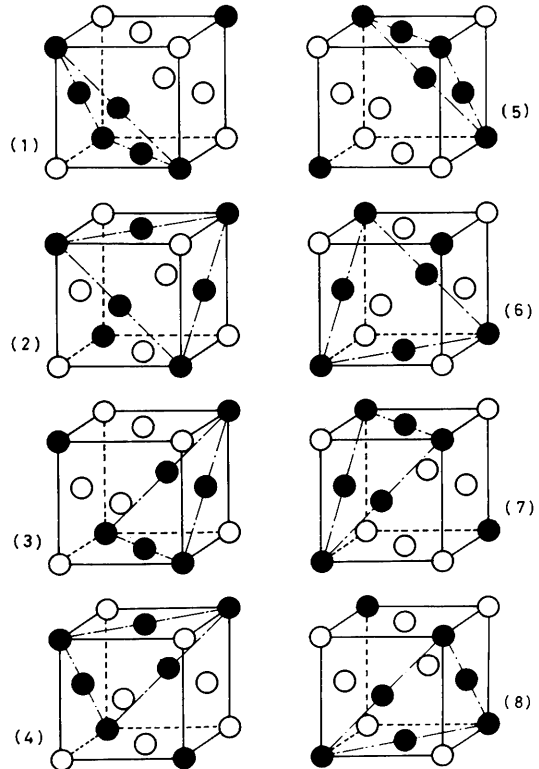

Fig. 2. Eight types of ordered lattice of the $L1_1$ structure. Open and solid circles indicate different kinds of atoms. These are in four groups, which consist of (1) and (5), (2) and (6), (3) and (7), and (4) and (8).

into four, each with the rhombohedral axis in common. The phase factors $\gamma_{G_i}^{(tt')}$ are given in Table 2. The correlation between the microdomains of different groups is shown not to modulate the diffuse intensity, and the intensity formulae are derived as

$$\alpha_0(\mathbf{q}_i) = \frac{N_t'}{nv_0^2}\,\varepsilon_t^2(\mathbf{q}_i), \qquad (14a)$$

$$\alpha^i(\mathbf{q}_i) = \frac{N_t'}{nv_0^2}\,\varepsilon_t^2(\mathbf{q}_i)\{p_{tt}(\mathbf{q}_i) - p_{t,t+4}(\mathbf{q}_i)\}, \qquad (14b)$$

N_t' being the total number of the microdomains of the tth and $(t + 4)$th types. We obtain from (14a) and (14b)

$$D_{G_i}(\mathbf{X}) = P_{tt}(\mathbf{X}) - P_{t,t+4}(\mathbf{X}). \qquad (15)$$

This is similar to (13) obtained in the case of $L1_0$.

4. Separation of $\alpha^0(\mathbf{q}_i)$ and $\alpha^i(\mathbf{q}_i)$ from the diffuse scattering

In order to obtain the interdomain correlation with the formulae derived above, it is necessary for $\alpha^0(\mathbf{q}_i)$ and

Table 2. $\gamma_{G_i}^{(tt')}$ *for the case of the* $L1_1$ *ordered structure*

		$\gamma_{G_i}^{(tt')}$			
t	t'	$G_{\frac{1}{2}\frac{1}{2}\frac{1}{2}}$	$G_{\frac{1}{2}\bar{\frac{1}{2}}\bar{\frac{1}{2}}}$	$G_{\bar{\frac{1}{2}}\frac{1}{2}\bar{\frac{1}{2}}}$	$G_{\bar{\frac{1}{2}}\bar{\frac{1}{2}}\frac{1}{2}}$
1	1	1	0	0	0
	5	−1	0	0	0
2	2	0	1	0	0
	6	0	−1	0	0
3	3	0	0	1	0
	7	0	0	−1	0
4	4	0	0	0	1
	8	0	0	0	−1

$\alpha^i(\mathbf{q}_i)$ to be evaluated separately from observed intensities. Let $\alpha^m(\mathbf{q}_i) \equiv \alpha^0(\mathbf{q}_i) + \alpha^i(\mathbf{q}_i)$. It is obtained by subtracting a constant part α^r from the total intensity $\alpha(\mathbf{q})$, as shown in Fig. 3.

We shall define a function $\alpha_{G_i}^m(\mathbf{R})$ as

$$\alpha_{G_i}^m(\mathbf{R}) = v_0 \int\limits_{(\text{around } G_i)} \alpha^m(\mathbf{q}_i)\exp(-2\pi i\mathbf{q}_i.\mathbf{R})\,d\mathbf{q}_i, \qquad (16)$$

where the integration is carried out over a zone defined by the planes which are perpendicular bisectors of the vectors joining the point G_i to its neighboring super-lattice reflection positions. On the boundaries of the zone, $\alpha^m(\mathbf{q}_i)$ takes negligibly small values. Our experiences with several alloys have shown that $\alpha_{G_i}^m(\mathbf{R})$ takes the form schematically shown in Fig. 4. Appearance of a negative part in $\alpha_{G_i}^m(\mathbf{R})$, indicated by A, is due to the fine structure of the intensity distribution in the vicinity of G_i (Fig. 3), and suggests the existence of the spatial correlation between the microdomains in an antiphase relation with each other. In the region of small $\mathbf{R}$ indicated by B in Fig. 4, however, $\alpha_{G_i}^m(\mathbf{R})$ is not much affected by the interdomain correlation and, therefore, it is not unreasonable to assume that its form is determined only by $\alpha_{G_i}^0(\mathbf{R})$, the Fourier transform of $\alpha^0(\mathbf{q}_i)$. Since this part of the intensity arises from the independent scattering of X-rays by the microdomains, $\alpha_{G_i}^0(\mathbf{R})$ must in general monotonically decrease to zero with increasing $\mathbf{R}$. With the form observed in region B in Fig. 4, it is possible to find a smooth function by fitting to $\alpha_{G_i}^m(\mathbf{R})$ in this region and extrapolating it into the region of large $\mathbf{R}$ (dotted line in Fig. 4). $\alpha^i(\mathbf{q}_i)$ is given by the difference between $\alpha^m(\mathbf{q}_i)$ and the Fourier transform of the function evaluated as $\alpha_{G_i}^0(\mathbf{R})$. Thus, one can obtain $\alpha^0(\mathbf{q}_i)$ and $\alpha^i(\mathbf{q}_i)$ separately from the observed intensity $\alpha(\mathbf{q})$ to a reasonable approximation.
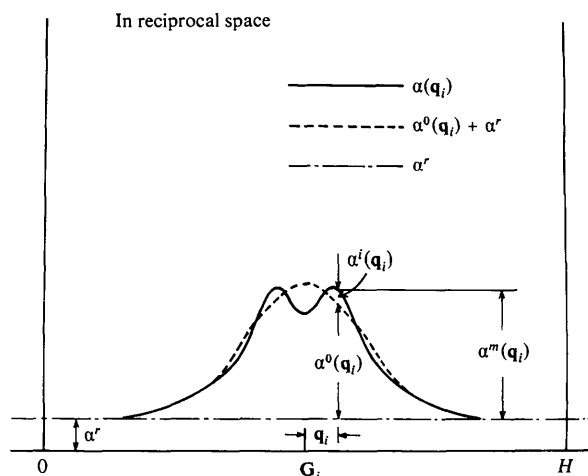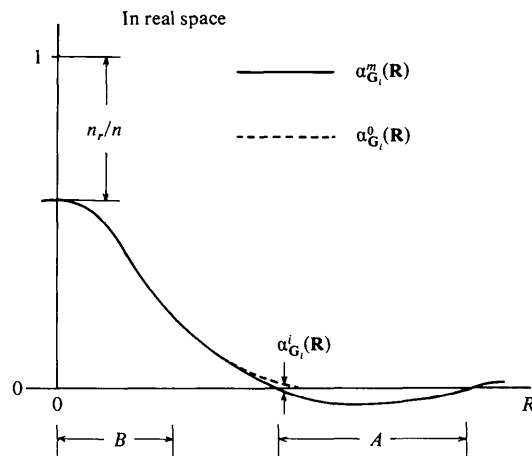

Fig. 3. Relationship between α^r, $\alpha^0(\mathbf{q}_i)$, $\alpha^i(\mathbf{q}_i)$ and $\alpha^m(\mathbf{q}_i)$ in one dimension.

Fig. 4. Relationship between $\alpha_{G_i}^0(\mathbf{R})$, $\alpha_{G_i}^i(\mathbf{R})$ and $\alpha_{G_i}^m(\mathbf{R})$.

5. Average size and number density of the microdomain

The square of the form factor $\varepsilon_t(\mathbf{q})$ defined in I can be written down explicitly as

$$\varepsilon_t^2(\mathbf{q}) = \int\int E_t(\mathbf{r}) E_t(\mathbf{r} + \mathbf{R}) \exp(2\pi i\mathbf{q}.\mathbf{R}) \, d\mathbf{r} \, d\mathbf{R}$$

$$= (v_0^2/N_t) \sum_u \sum_{\mathbf{R}_i} \sum_{\mathbf{R}_l} E_u(\mathbf{R}_i) E_u(\mathbf{R}_i + \mathbf{R}_l)$$

$$\times \exp(2\pi i\mathbf{q}.\mathbf{R}_i)$$

$$= (1/N_t) \sum_u{}' |\varepsilon_u(\mathbf{q})|^2, \tag{17}$$

where $\varepsilon_u(\mathbf{q})$ is the Fourier transform of the shape function of the individual domain $E_u(\mathbf{R}_i)$, and the prime indicates the limited sum taken only over the microdomains of tth type. We will derive the statistical parameters for the microdomain structure using the property of $E_u(\mathbf{R}_i)$ given in (I-6).

Integration of $\alpha^0(\mathbf{q}_i)$ given by $(7a)$, $(12a)$ and $(14a)$ with respect to $\mathbf{q}_i$ leads to

$$v_0 \int_{\text{(around } G_i)} \alpha^0(\mathbf{q}_i) \, d\mathbf{q}_i = (N_t'/n)\left\{\frac{1}{N_t} \sum_u{}' \sum_{\mathbf{R}_i} E_u^2(\mathbf{R}_i)\right\}$$

$$= N_t'\langle n\rangle_t/n, \tag{18}$$

through (17). N_t' must be replaced by $N_d/3$ in the case of $L1_2$. $\langle n\rangle_t$ is the average number of atoms contained in a single microdomain of the tth type. Equation (18) gives a ratio of the total number of atoms in the microdomains of the tth [tth and $(t + 3)$th for $L1_0$, and tth and $(t + 4)$th for $L1_1$] type and the total number of atoms in the alloy crystal. Another useful relation is

$$\alpha^0(\mathbf{q}_i = 0) = (N_t'/n) \sum_u{}' \left\{\sum_{\mathbf{R}_i} E_u(\mathbf{R}_i) \sum_{\mathbf{R}_j} E_u(\mathbf{R}_j)\right\}$$

$$= N_t'\langle n^2\rangle_t/n. \tag{19}$$

Further, we consider an integral

$$v_0 \int \{\alpha^0(\mathbf{q}_i)\}^{1/2} \, d\mathbf{q}_i = (N_t'/n)^{1/2} \int \varepsilon_t(\mathbf{q}_i) \, d\mathbf{q}_i. \tag{20}$$

This integral cannot be evaluated directly. Let $\mathbf{R}_u$ indicate a central position of the domain labeled with u, and let $E_u(\mathbf{R})$ be Fourier transformed taking $\mathbf{R}_u$ as an origin as

$$e_u(\mathbf{q}) = \int E_u(\mathbf{R}) \exp(-2\pi i\mathbf{q}.\mathbf{R} - \mathbf{R}_u) \, d\mathbf{R}. \tag{21}$$

We then define an average function $e_t(\mathbf{q})$:

$$e_t(\mathbf{q}) = (1/N_t) \sum_u{}' e_u(\mathbf{q}). \tag{22}$$

Perfect ordering in the central region of the microdomains requires

$$\int e_t(\mathbf{q}) \, d\mathbf{q} = (1/N_t) \sum_u{}' E_u(\mathbf{R}_u) = 1. \tag{23}$$

We introduce a factor $\eta(\mathbf{q})$ as

$$\varepsilon_t(\mathbf{q}) = \eta(\mathbf{q}) \, e_t(\mathbf{q}). \tag{24}$$

For $\mathbf{q} = 0$,

$$\eta(0) = \frac{\varepsilon_t(0)}{e_t(0)} = \left(\frac{\langle n^2\rangle_t}{\langle n\rangle_t^2}\right)^{1/2}, \tag{25}$$

since $e_t(0) = \langle n\rangle_t$. If we neglect the $\mathbf{q}$ dependence of $\eta(\mathbf{q})$ and replace it in (24) by $\eta(0)$, we can readily evaluate the integral in (20) and obtain

$$v_0 \int \{\alpha^0(\mathbf{q}_i)\}^{1/2} \, d\mathbf{q}_i = (N_t'/n)^{1/2}(\langle n^2\rangle_t/\langle n\rangle_t^2)^{1/2}. \tag{26}$$

We can then solve (18), (19) and (26) for the average number of atoms $\langle n\rangle_t$ in a single microdomain, its mean square $\langle n^2\rangle_t$ and the number density of the domains N_t'/n.

A quantity

$$\sigma^2 = \langle n^2\rangle_t - \langle n\rangle_t^2 \tag{27}$$

gives a measure of the broadness of the atom number distribution and hence the domain size distribution around the average.

Finally, the form factor $\varepsilon_t(\mathbf{q})$ can be derived from $(7a)$, $(12a)$ and $(14a)$ and, therefore, its Fourier transform $E_t(\mathbf{R})$ also. $E_t(\mathbf{R})$ is generally a smooth function in real space, which is a kind of average shape function for the microdomain.

6. Discussion

The inhomogeneous models of the short-range-ordered alloys already proposed by several investigators were not developed quantitatively in the sense that the observed intensity distribution and the statistical parameters of the microdomain structure were directly connected. Along with the parameters, such as the average domain size and the size distribution, we introduced in this series of papers an interdomain correlation, which gives rise to the fine structures on the diffuse maxima. The present author (Hashimoto & Iwasaki, 1979) reported microdomain model analyses for CuPt and CuAu alloys, in which the present model was applied successfully.

In some short-range-ordered alloys, we found σ to be extraordinarily larger than $\langle n\rangle_t$ calculated in the way described in § 5. We shall discuss this kind of abnormal, contradictory result.

σ^2 defined in § 5 is nothing but

$$\sigma^2 = (n/N_t')\left\{a^0(\mathbf{q}_i = 0) - v_0 \int a^0(\mathbf{q}_i)\,d\mathbf{q}_i\right\}$$

$$= \left(\frac{1}{v_0}\int E_t(\mathbf{R})\,d\mathbf{R}\right)^2 - \left(\frac{1}{v_0}\int E_t^2(\mathbf{R})\,d\mathbf{R}\right)^2. \quad (28)$$

From the physical meaning of σ^2, it should not be large compared with $\langle n \rangle_t^2$ in (27) and hence the second term on the right-hand side of (28). Throughout the arguments developed above, it was assumed that the shape function of the individual domain $E_u(\mathbf{R}_i)$ had the form of a step function, but if there exists an inhomogeneous ordering within a domain, σ^2 will have a relatively large value. For the sake of convenience, we consider the shape of the microdomains to be identical and use $S_t(\mathbf{R})$, called the 'ordering function', instead of $E_t(\mathbf{R})$ in (28). The relationship between these two functions is illustrated in Fig. 5, along with $S_t^2(\mathbf{R})$. From this figure it can readily be seen that the integrals of $S_t(\mathbf{R})$ and $S_t^2(\mathbf{R})$ in (28) lead to different values. Thus, σ^2 reveals here a measure of the lowering of the degree of order at the periphery of the domain.

The non-uniform ordering within the domain gives the following relations instead of (18), (19) and (26),

$$v_0 \int a^0(\mathbf{q}_i)\,d\mathbf{q}_i = (N_t'/n)\left\{\frac{1}{v_0}\int S_t^2(\mathbf{R})\,d\mathbf{R}\right\}, \quad (29)$$

$$a^0(\mathbf{q}_i = 0) = (N_t'/n)\left\{\frac{1}{v_0}\int S_t(\mathbf{R})\,d\mathbf{R}\right\}^2 \quad (30)$$

and

$$v_0 \int \{a^0(\mathbf{q}_i)\}^{1/2}\,d\mathbf{q}_i = (N_t'/n)^{1/2}\,S_t(0). \quad (31)$$

Here, we can put $S_t(\mathbf{R}) = 1$ at $\mathbf{R} = 0$, assuming that the degree of order at the center of the domain is perfect.*

---

* Even in the case that the order is not perfect, it is possible by putting $S_t(0) = 1$ to judge whether the ordering is uniform or not.
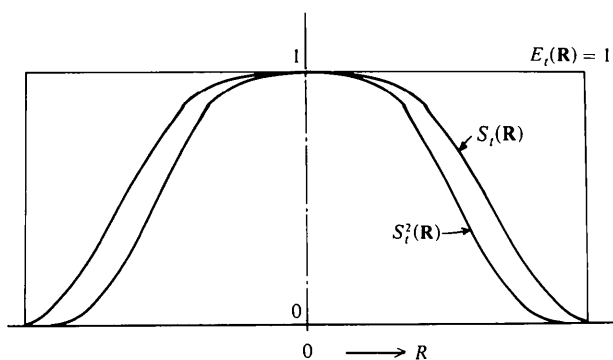

Fig. 5. Illustration of the ordering function $S_t(\mathbf{R})$ and its square. $E_t(\mathbf{R})$ is here taken to be unity within a domain.

Then, (31) gives $(N_t'/n)^{1/2}$, a square root of the number density of the microdomains. $S_t(\mathbf{R})$, associated with the domain shape, can be written from (I-13) and (7a) etc. as

$$S_t(\mathbf{R}) = (n/N_t')\,v_0 \int \{a^0(\mathbf{q}_i)\}^{1/2}\exp(2\pi i\mathbf{q}_i.\,\mathbf{R})\,d\mathbf{q}_i. \quad (32)$$

From the correspondence of (29) to (18), an 'effective number' of atoms in a domain is given as

$$\langle n \rangle_t = (1/v_0)\int S_t^2(\mathbf{R})\,d\mathbf{R}. \quad (33)$$

The interdomain correlation function $P_{tt'}(\mathbf{X})$ can be evaluated in the same way as in § 3.

In § 3, it was seen that only the differences between the interdomain correlation functions $P_{tt'}(\mathbf{X})$ are known. In the case of the $L1_2$ type, for example, if we introduce a quantity $D_0(\mathbf{X}) = \sum_t P_{1t}(\mathbf{X})$, a total pair correlation function of the domains, the partial correlation functions $P_{1t}(\mathbf{X})$ are given by

$$P_{11}(\mathbf{X}) = (\tfrac{1}{4})\{D_0(\mathbf{X}) + D_{100}(\mathbf{X}) + D_{010}(\mathbf{X}) + D_{001}(\mathbf{X})\},$$

$$P_{12}(\mathbf{X}) = (\tfrac{1}{4})\{D_0(\mathbf{X}) - D_{100}(\mathbf{X}) - D_{010}(\mathbf{X}) + D_{001}(\mathbf{X})\},$$

$$P_{13}(\mathbf{X}) = (\tfrac{1}{4})\{D_0(\mathbf{X}) + D_{100}(\mathbf{X}) - D_{010}(\mathbf{X}) - D_{001}(\mathbf{X})\},$$

$$P_{14}(\mathbf{X}) = (\tfrac{1}{4})\{D_0(\mathbf{X}) - D_{100}(\mathbf{X}) + D_{010}(\mathbf{X}) - D_{001}(\mathbf{X})\}. \quad (34)$$

The $P_{1t}(\mathbf{X})$'s can be calculated from these equations, provided that $D_0(\mathbf{X})$ is obtained in an alternative way. The same is true for the $L1_0$ and $L1_1$ types.

References

BORIE, B. & SPARKS, C. J. (1964). Acta Cryst. 17, 827–835.

BORIE, B. & SPARKS, C. J. (1971). Acta Cryst. A27, 198–201.

HASHIMOTO, S. (1974). Acta Cryst. A30, 792–798.

HASHIMOTO, S. & IWASAKI, H. (1979). AIP Conference Proceedings No. 53, pp. 283–285. New York: American Institute of Physics.

SINCLAIR, R. & THOMAS, G. (1975). J. Appl. Cryst. 8, 206–210.

SPARKS, C. J. & BORIE, B. (1965). Local Atomic Arrangements Studied by X-ray Diffraction, edited by J. B. COHEN & J. E. HILLIARD, pp. 5–46. New York: Gordon & Breach.

TANAKA, N., OHSHIMA, K., HARADA, J. & MIHAMA, K. (1979). AIP Conference Proceedings No. 53, pp. 292–294. New York: American Institute of Physics.