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Analytical Theory of Diffuse Scattering from Distributions of Non-overlapping Structures

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Abstract

The diffuse scattering for X-rays or neutrons is derived analytically for equilibrium or non-equilibrium nanoscale structures distributed according to any set of suitable distribution functions. A disordered lattice gas model has been used basically for one dimension but for objects that may have higher dimensions. The results for multilayers, phase separation, domain patterns, surface roughness and 'hut' clusters are discussed and explicit formulae are given.

1. Introduction

With the continued development of high-flux sources for neutrons and X-rays, as well as the He-scattering technique, it has become possible to study the diffuse scattering resulting from nano- or mesoscale objects. Considerable literature already exists on the subject, developed in various fields. A comprehensive review can be found in the work by Jagodzinski & Frey (1992) and further developments by Pflanz & Moritz (1992) and Fullerton, Schuller, Vanderstraeten & Bruynseraede (1992). The latter papers derive expressions for numerical solutions. In this paper, we focus on deriving analytical expressions for the diffuse scattering from disordered structures.

The phenomena of interest on the nano- or mesoscale may be the results of equilibrium properties but are more often related to non-equilibrium states of matter and occur in several different fields of physics. Some of the first problems considered were the stacking faults found as one of the most important defects in metals at low temperatures and in martensitic polytype materials (Landau, 1937; Lifshits, 1937; Hendricks & Teller, 1942; Jagodzinski, 1949; Berliner & Werner, 1986; Berliner & Gooding, 1994). It occurs in pattern formation under epitaxic growth of surfaces where terraced islands or 'huts' may start to be formed (Mo, Savage, Swartzentruber & Lagally, 1990; Zeppenfeld, Krzyowski, Romainczyk, Comsa & Lagally, 1994). A presently very active field of research concerns self-organization (Kern, Niehus, Schatz, Zeppenfeld, Goerge & Comsa, 1991; Zeppenfeld, Krzyowski, Romainczyk, Comsa & Lagally, 1994), which is an equilibrium result of elastic relaxation (Shchukin, Borovkov, Ledentsov & Bimberg, 1995; Shchukin, Borovkov, Ledentsov & Kop'ev, 1995, 1996) in which almost-ordered nanoscale structures spontaneously form on a crystal surface. Other problems of relevance are found in artificially grown multilayer systems with some randomness or distribution in the layer thickness (Fullerton, Schuller, Vanderstraeten & Bruynseraede, 1992). Examples are also found in the phase-separation problem in binary alloys (Fratzl, Lebowitz, Penrose & Amar, 1991) or antiphase structures such as the high-temperature superconductors $YBa_2Cu_3O_{6+x}$ (YBCO) (Poulsen, Andersen, Andersen, Bohr & Mouritsen, 1990; Khachaturyan & Morris, 1990; Aligia, 1993) and in Ising magnets. Related problems occur in biophysics (Zhang, Suter & Nagle, 1994) and even in computer-simulation physics (Lindgård, 1994). It is important to go beyond, for example, the often-used simple Warren approximation (Warren, 1941, 1990) in describing scattering from variously sized objects in order to obtain their statistical size distributions. In a recent paper (Fiig, Andersen, Berlin & Lindgård, 1995) (note that in Table I therein, κ only refers to the half-width, not the FWHM), the diffuse scattering was discussed in terms of distribution probabilities of sizes for the various scattering objects, while neglecting the distribution of the space between the objects. We shall extend this work considerably by first considering the distributions of all the space-filling objects. Second, we consider a large number of pertinent structural problems. For these, we derive the explicit scattering cross sections, taking into account the 'excluded-volume' effect exactly. The theory is a further development of the original work by Uimin (1994) in order to describe the microstructure of YBCO. In order to emphazise the universal aspects of the problems, we present the general theory rather than discussing a particular phenomenon. We shall restrict ourselves to considering the scattering aspects, without going into the physics of why the actual structures occur or are in equilibrium or not.

The microstructures mentioned above are often observed using real-space methods like optical, electronic or scanning tunneling microscope techniques. With these, one obtains a direct picture of structures, whereas the statistical properties are more difficult to assess.

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Scattering methods are in this respect complementary, giving a statistical average of the structures. It is the purpose of this paper to develop the theoretical background for determining statistical properties and distribution functions. We also include a consideration of the internal structure of the particles, which gives rise to characteristic Bragg peaks. For small nanometre-sized objects, this is important. For scattering from larger-scale objects, one is generally not interested in this aspect and therefore only uses the small-angle scattering technique studying the scattering close to the (0, 0, 0) 'Bragg' peak. This result can also be obtained from our theory by simple expansion in the wave vector $\mathbf{q}$.

We shall only consider the diffuse scattering as arising from breaking a perfect order of a lattice gas system. The general scattering theory was introduced by Fiig *et al.* (1995) in some detail and will not be repeated here. The exact results for the cross section will be given for the linear chain and the possible extrapolation of the results to higher dimensions will also be discussed. The 'excluded-volume' effects in higher dimensions is a long-standing problem in statistical mechanics (Reiss, Frisch & Lebowitz, 1959), still vigorously discussed (Torquato, 1995; Pflanz & Moritz, 1992: Fullerton *et al.*, 1992). Analysis of diffuse scattering experimentally makes it clear that any other imperfection, such as lattice deformations and strains, also give rise to diffuse scattering. These will not be discussed here. The seminal work on such 'real' crystal problems is the book by Krivoglaz (1969); a recent discussion may be found in §4 of Jagodzinski & Frey (1992).

The paper is organized as follows. First, an introduction of the probabilistic framework is given. In the following section, several examples are explicitly worked out for various problems of current interest. Concluding remarks complete the paper.

2. Theory for the structure factor

In this section, we shall derive the scattering cross section basically for a lattice gas system with defects. However, in fact, we consider a more general model that describes the scattering of a system of any number of slabs of different lattices with different lattice constants and atoms with different scattering lengths. Their thicknesses are described by any suitable distribution function, for example, a Poisson distribution or an exponential distribution. In a recent paper by Fiig *et al.* (1995), the line shape was calculated and discussed for such distributions of matter separated by arbitrary distributions of empty space, *cf.* Uimin (1994). Since the calculation of the cross sections derived in the present paper is similar, we do not present the equivalent numerical calculations here (also because this is more difficult to do generally with more than one distribution function). By further allowing for a profile in the scattering lengths, the theory can be used for reproducing at least certain aspects of scattering from higher-dimensional objects subject to the 'excluded-volume' constraint.

2.1. *Probabilistic prerequisite*

First, let us consider a simple example of a one-dimensional chain with lattice spacing a (for simplicity we set $a = 1$ in this section). On this chain, we distribute scattering particles in different length fragments of two structures: δ_1, where every site is occupied, and δ_2, where every second site is occupied. In addition, we allow fragments, where there are no scattering particles at the sites. These fragments can be of any length larger than 2 and are restricted by a particle at both ends. For computational convenience, we define 'empty' fragments, δ_0, which include the particle at the left end. This allows us to consider sequences of structured fragments in which any δ_0 fragment can be followed by another δ_0 fragment. The separating particle could be interpreted as an element of the δ_1 structure but including such a particle into a definition of the δ_0 fragment is a suitable computational trick. An example including these definitions is shown in Fig. 1. A realization of this situation is found in YBCO, where δ_1 would correspond to the projection of a domain of the ortho-I structure and δ_2 to the projection of an aligned ortho-II structure. Similar examples can be realized as a sequence of multilayers. We shall thus use the same convention for the fragments of all three types. On any fragment, the leftmost site is occupied by a particle. This is the last particle with the left fragment periodicity but it is counted as the first site of the rightmost fragment. The length of the δ_1 fragment, say ℓ, coincides with the number of particles within that fragment. With the same number of particles, the length of the δ_2 fragment will be equal to 2ℓ. For both cases, $\ell \geq 1$. However, for the 'empty' fragments we have $\ell \geq 3$ because an 'empty' fragment with $\ell = 1$ (2) must be classified as δ_1 (δ_2). Let $\mathcal{N}$ be the total number of 1D lattice sites. If $N_\delta(\ell)$ is the total number of fragments of length ℓ of the δth kind ($\delta = 0, 1, 2$ for 'empty', δ_1 and δ_2 fragments, respectively), the 'probabilities' are defined as follows:

$$\begin{aligned} D_0(\ell) &= N_0(\ell)/\mathcal{N}, \\ D_1(\ell) &= N_1(\ell)/\mathcal{N}, \\ D_2(2\ell) &= N_2(2\ell)/\mathcal{N}. \end{aligned} \tag{1}$$

The sum of 'probabilities' is not equal to 1. The true probabilities, w_0, w_1 and w_2, for finding a fragment belonging to one of the three possible types of fragments are

$$\begin{aligned} w_0 &= \sum_{\ell \geq 3} D_0(\ell)/\mathcal{D}, \\ w_1 &= \sum_{\ell \geq 1} D_1(\ell)/\mathcal{D}, \\ w_2 &= \sum_{\ell \geq 1} D_2(2\ell)/\mathcal{D}, \end{aligned} \tag{2}$$

where

$$\mathcal{D} = \sum_{\ell \geq 3} D_0(\ell) + \sum_{\ell \geq 1} D_1(\ell) + \sum_{\ell \geq 1} D_2(2\ell). \quad (3)$$

The D_δ 'probabilities' satisfy a total length constraint, and the number of particles constraint, yielding

$$\begin{aligned} \sum_{\ell \geq 3} \ell D_0(\ell) + \sum_{\ell \geq 1} \ell D_1(\ell) + \sum_{\ell \geq 1} 2\ell D_2(2\ell) &= 1, \\ \sum_{\ell \geq 3} D_0(\ell) + \sum_{\ell \geq 1} \ell D_1(\ell) + \sum_{\ell \geq 1} \ell D_2(2\ell) &= c, \end{aligned} \quad (4)$$

where c is the concentration of particles. We shall further consider that correlations exist not only within separate fragments, as expressed by the 'probability' set $\{D_\delta\}$, but also between the nearest-neighbor fragments. The reason for including such correlations of higher rank is that a fragment of, say, the δ_2 type can be followed either by a δ_1 fragment or an 'empty' fragment, but not by another δ_2 fragment. On the other hand, an 'empty' fragment is allowed to be followed by any kind of configuration. Clearly, if the 'empty' fragments are excluded, the situation becomes deterministic, which means that the δ_1 and δ_2 fragments strictly alternate. We will be starting §3 with that particular case.

Let us now generalize the above by considering any number of different types of structure. Equations (1)–(4), considered as definitions, are straightforwardly generalized by using an arbitrarily large set of D's: $D_\alpha(\ell_\alpha)$ represents the 'probability' of finding a fragment of a structure type α with length ℓ_α, *i.e.*

$$D_\alpha(\ell_\alpha) = N_\alpha(\ell_\alpha)/\mathcal{N}.$$

Then the analog of (2) and (3) is

$$\mathcal{D}_\alpha = \sum_\ell D_\alpha(\ell_\alpha), \quad \mathcal{D} = \sum_\alpha \mathcal{D}_\alpha, \quad w_\alpha = \mathcal{D}_\alpha/\mathcal{D}. \quad (5)$$

Let us introduce the double-fragment 'probabilities'

$$D_{\alpha\beta}(\ell_\alpha, \ell_\beta) = N_{\alpha\beta}(\ell_\alpha, \ell_\beta)/\mathcal{N}, \quad (6)$$

where $N_{\alpha\beta}(\ell_\alpha, \ell_\beta)$ is the total number of $\alpha\beta$ fragments composed of the α and β types of lengths ℓ_α and ℓ_β, respectively. Then let us define the decoupling scheme for the 'probabilities' of higher ranks,

$$D_{\alpha\beta}(\ell_\alpha, \ell_\beta) = D_\alpha(\ell_\alpha) W_{\alpha\beta} D_\beta(\ell_\beta). \quad (7)$$

This amounts to assuming a random sequence of possible type fragments (only subject to a probability constraint) while neglecting any correlation between the length of the adjacent fragments. Summing over all neighbor-fragment possibilities gives the identities

$$D_\alpha(\ell_\alpha) = \sum_\beta \sum_{\ell_\beta} D_{\alpha\beta}(\ell_\alpha, \ell_\beta) = \sum_\beta \sum_{\ell_\beta} D_{\beta\alpha}(\ell_\beta, \ell_\alpha).$$

Then it is clear from (7) that the elements $W_{\alpha\beta}$ must satisfy the equation

$$\sum_\beta W_{\alpha\beta} \mathcal{D}_\beta = \sum_\beta \mathcal{D}_\beta W_{\beta\alpha} = 1. \quad (8)$$

The $W_{\alpha\beta}$ elements play a role of 'transition probabilities', *i.e.* they are matrix elements of an unnormalized transfer matrix. The transition probabilities satisfy the symmetry properties $W_{\alpha\beta} = W_{\beta\alpha}$ if the system does not exhibit special chiral properties. Within this scheme, the 'probability' of any rank can be decoupled:

$$\begin{aligned} &D_{\alpha_1\alpha_2\ldots\alpha_n}(\ell_1, \ell_2, \ldots, \ell_n) \\ &\quad = D_{\alpha_1\alpha_2\ldots\alpha_{n-1}}(\ell_1, \ell_2, \ldots, \ell_{n-1}) W_{\alpha_{n-1}\alpha_n} D_{\alpha_n}(\ell_n) \\ &\quad = D_{\alpha_1}(\ell_1) W_{\alpha_1\alpha_2} D_{\alpha_2}(\ell_2) \ldots W_{\alpha_{n-1}\alpha_n} D_{\alpha_n}(\ell_n), \end{aligned} \quad (9)$$

with the same constraints as (8) imposed on the set of elements $W_{\alpha\beta}$. We remark that the transfer-matrix method was used very early in the theory of diffuse scattering, *cf.* Jagodzinski & Frey (1992). It has recently been used in a similar way for the domain distribution problem by Pflanz & Moritz (1992). We believe that the present formulation, in a transparent way, elucidates the approximation used in both theories, *i.e.* the Markov process.

For the further derivation of the structure factor, we need a definition of averaging over various realizations for a physical quantity $\mathcal{F} = F_{\alpha_1}(\ell_1) F_{\alpha_2}(\ell_2) \ldots F_{\alpha_n}(\ell_n)$. It can be done as follows:

$$\begin{aligned} \langle \mathcal{F} \rangle = &\sum_{\{\alpha\}} \sum_{\{\ell\}} D_{\alpha_1\alpha_2\ldots\alpha_n}(\ell_1, \ell_2, \ldots, \ell_n) \\ &\times F_{\alpha_1}(\ell_1) F_{\alpha_2}(\ell_2) \ldots F_{\alpha_n}(\ell_n) \\ &\times \left[\sum_{\{\alpha\}} \sum_{\{\ell\}} D_{\alpha_1\alpha_2\ldots\alpha_n}(\ell_1, \ell_2, \ldots, \ell_n) \right]^{-1}. \end{aligned} \quad (10)$$

For $n = 1$, (10) takes the form

$$\langle \mathcal{F} \rangle = \sum_\alpha w_\alpha \overline{F_\alpha}, \quad (11)$$

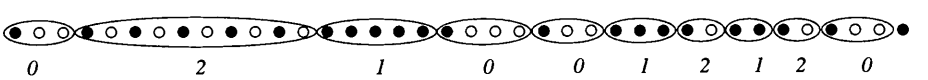

Fig. 1. An example of of two different co-existing structures labeled δ_1 and δ_2 and in addition δ_0 representing an 'empty structure'. The particles (filled circles) are arranged in fragments and counted according to the convention explained in text.

where

$$\overline{F_\alpha} = \sum_\ell D_\alpha(\ell) F_\alpha(\ell)/\mathcal{D}_\alpha \tag{12}$$

is the ensemble average over a given distribution of the αth fragments. Equation (10) can be rewritten for any $n \geq 1$ in the following way:

$$\langle \mathcal{F} \rangle = \mathcal{D}^{n-1} \sum_{\alpha_1 \ldots \alpha_n} w_{\alpha_1} \overline{F_{\alpha_1}} W_{\alpha_1\alpha_2} w_{\alpha_2} \overline{F_{\alpha_2}} \times \ldots \times W_{\alpha_{n-1}\alpha_n} w_{\alpha_n} \overline{F_{\alpha_n}}. \tag{13}$$

Let us explicitly discuss the 'transfer' matrix for the introductory example. When 'empty' fragments are absent, $W_{11} = W_{22} = 0$ because of the alternation, which also causes the number of fragments to be equal, $\mathcal{D}_1 = \mathcal{D}_2$; so, according to (8), one obtains for the off-diagonal elements $W_{12} = W_{21} = 1/\mathcal{D}_1$, *i.e.*

$$\mathbf{W} = \begin{pmatrix} W_{11} & W_{12} \\ W_{21} & W_{22} \end{pmatrix} = \frac{2}{\mathcal{D}} \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \tag{14}$$

The case where we consider 'empty' fragments and one of the $\alpha = \alpha_1$ or α_2 type structures is slightly different because $W_{00} \neq 0$. We find from (8)

$$\mathbf{W} = \begin{pmatrix} W_{00} & W_{0\alpha} \\ W_{\alpha 0} & W_{\alpha\alpha} \end{pmatrix} = \frac{1}{\mathcal{D} w_0} \begin{pmatrix} 1 - w_\alpha/w_0 & 1 \\ 1 & 0 \end{pmatrix}. \tag{15}$$

Finally, we present the form of the 3×3 matrix that describes the 'transition probabilities' when all three kinds of fragments are allowed. Note that there are only three independent constraints imposed by (8) on the four formally independent $W_{\alpha\beta}$'s; this gives rise to one free parameter (w) in the matrix

$$\mathbf{W} = \frac{1}{\mathcal{D}_0} \begin{pmatrix} 1 - (w_1 + w_2)/w_0 + 2w w_1 w_2/w_0^2 & 1 - w w_2/w_0 & 1 - w w_1/w_0 \\ 1 - w w_2/w_0 & 0 & w \\ 1 - w w_1/w_0 & w & 0 \end{pmatrix}. \tag{16}$$

If we do not allow for segments with sparsely distributed single particles, here represented by 'empty' fragments, we have $W_{00} = 0$ and $w = \frac{1}{2}(w_0/w_1 + w_0/w_2 - w_0^2/w_1 w_2)$; then $\mathbf{W}$ is fully determined by the constraints (8).

2.2. *Structure factor: general derivation*

In this section, we shall first derive the general expression for the diffuse part of the structure factor $\mathcal{S}(q)$ for a linear chain consisting of different length fragments of different materials, which may have different lattice constants and different scattering lengths. Subsequently, we consider possible generalizations of the results to higher spatial dimensions.

Let α represent the different kinds of locally ordered fragments. p is the total number of kinds; in our example above, $p = 3$. Next, let $\alpha_1, \alpha_2, \ldots, \alpha_k$ be a realized sequence of a large number, k, of these fragments occurring with the probability $D_{\alpha_1\alpha_2\alpha_3\ldots\alpha_k}(\ell_1, \ell_2, \ell_3 \ldots, \ell_k)$, which will be transformed according to (9). Let the lattice constant specific to an α-type fragment be denoted a_α. The scattering amplitude of the wave number q is the Fourier transform applied to the particles in the sequence

$$\begin{aligned} \mathcal{A}(q,k) &= b_\alpha\{1 + \exp(iqa_\alpha) + \ldots + \exp[iqa_\alpha(\ell_1 - 1)]\} \\ &\quad + b_\beta \exp(iqa_\alpha \ell_1)\{1 + \exp(iqa_\beta) + \ldots \\ &\quad + \exp[iqa_\beta(\ell_2 - 1)]\} + b_\gamma \exp[iq(a_\alpha \ell_1 + a_\beta \ell_2)] \\ &\quad \times \{1 + \exp(iqa_\gamma) + \ldots + \exp[iqa_\gamma(\ell_3 - 1)]\} \\ &\quad + \ldots . \end{aligned} \tag{17}$$

This can be partially summed over the fragments of length $a_\alpha \ell_1$, $a_\beta \ell_2$ *etc.* Symbolically, the Fourier component can thus be written for the realization of k various fragments

$$\mathcal{A}(q,k) = F(1) + \Gamma(1)F(2) + \Gamma(1)\Gamma(2)F(3) + \ldots + \Gamma(1)\Gamma(2)\ldots\Gamma(k-1)F(k). \tag{18}$$

Here, $F(j)$ is the scattering amplitude of the jth fragment (α type) and $\Gamma(j)$ the separation phase factor, *i.e.*

$$F_\alpha(j) = \sum_{r=0}^{\ell_j - 1} b_\alpha \exp(iqa_\alpha r), \quad \Gamma_\alpha(j) = \exp(iqa_\alpha \ell_j),$$

where b_α is the specific scattering length. We notice that $\Gamma(j)\Gamma(j)^* = 1$. By introducing a specific a_α, we can describe the situation with arbitrary lattice constants for the αth fragments. [Note that the 'empty' fragments with one particle to the left considered in §2 represent a special case. They can be treated as separate unit-length α-type structures but then the matrices may become very large. It is simpler to include them as a general special 0 type with arbitrary length r for which we must use $F(j) = 1$ and $\Gamma(j) = \exp(iqr_j)$. The distribution function for the length of the fragments is continuous $D_0(r)$, excluding (prime) any explicitly involved lattice constant a_α: $\int_0^{\infty\prime} D_0(r)\,\mathrm{d}r = \mathcal{D}_0$.]

For the particular example discussed in §2, the a_α's were supposed to be integers 1, 2 and the scattering lengths equal to 1. This gives the expressions for scattering amplitudes for fragments of length ℓ and 2ℓ:

$$\begin{aligned} F_0(\ell) &= 1, \\ F_1(\ell) &= [1 - \exp(iq\ell)]/[1 - \exp(iq)], \\ F_2(2\ell) &= [1 - \exp(iq2\ell)]/[1 - \exp(i2q)]. \end{aligned} \tag{19}$$

The series $\mathcal{A}(q,k)$ depends not only on q but also on the specific realization of the α's. However, upon averaging, it becomes a convergent sum for large values of k and therefore independent of k when considering a large number of fragments. The structure factor is on the other hand in most cases of interest independent of a concrete realization of fragments in the chain. Thus it must be averaged over possible realizations. Let us start by calculating a quantity $\mathcal{K}(q,k)$, which becomes $\mathcal{S}(q)$ for $k \to \infty$.

$$\mathcal{K}(q,k) = \langle \mathcal{A}(q,k)\mathcal{A}^*(q,k)\rangle, \tag{20}$$

where $\langle\,\rangle$ indicates such an average over all possible configurations with k fragments according to (10). We note that the leading term in $\mathcal{K}(q,k)$ is *linear* in k. Further, we notice that $\mathcal{K}(q,k)$ can be written as

$$\begin{aligned}\mathcal{K}(q,k) &= \langle [F(1) + \Gamma(1)F(2) + \ldots \\ &\quad + \Gamma(1)\ldots\Gamma(k-1)F(k)] \times [\text{c.c.}]\rangle \\ &= \mathcal{K}(q,k-1) + \langle F(1)F^*(1)\rangle \\ &\quad + \{\langle F^*(1)[\Gamma(1)F(2) + \ldots \\ &\quad + \Gamma(1)\ldots\Gamma(k-1)F(k)]\rangle + \text{c.c.}\}.\end{aligned}$$

To leading order in k, we then come to the expression

$$\begin{aligned}\mathcal{K}(q,k) = k\{&\langle F(1)F^*(1)\rangle + (\langle F^*(1)[\Gamma(1)F(2) + \ldots \\ &+ \Gamma(1)\ldots\Gamma(k-1)F(k)]\rangle + \text{c.c.})\},\end{aligned} \tag{21}$$

where the first term represents the scattering from the independent fragments

$$\langle F(1)F^*(1)\rangle = \sum_{\alpha=1}^{p} w_\alpha \overline{F_\alpha F_\alpha^*}. \tag{22}$$

The second is an interference term, which is explicitly given by

$$\begin{aligned}&\langle F^*(1)[\Gamma(1)F(2) + \ldots + \Gamma(1)\ldots\Gamma(k-1)F(k)]\rangle \\ &= \mathcal{D}\sum_{\alpha_1} w_{\alpha_1}\overline{F_{\alpha_1}^*\Gamma_{\alpha_1}}\sum_{\alpha_2} W_{\alpha_1\alpha_2} w_{\alpha_2}\overline{F_{\alpha_2}} + \ldots \\ &\quad + \mathcal{D}^{k-1}\sum_{\alpha_1} w_{\alpha_1}\overline{F_{\alpha_1}^*\Gamma_{\alpha_1}}\sum_{\alpha_2} W_{\alpha_1\alpha_2} w_{\alpha_2}\overline{\Gamma_{\alpha_2}}\ldots \\ &\quad \times \sum_{\alpha_{k-1}} W_{\alpha_{k-2}\alpha_{k-1}} w_{\alpha_{k-1}}\overline{\Gamma_{\alpha_{k-1}}}\sum_{\alpha_k} W_{\alpha_{k-1}\alpha_k} w_{\alpha_k}\overline{F_{\alpha_k}}.\end{aligned} \tag{23}$$

The averaging in (22)–(23) is performed in accordance with definitions (10)–(13). Any term on the right-hand side of (23) can be generally written as $\langle L|\mathbf{M}|R\rangle$, where $\langle L|$ and $|R\rangle$ are p-component bra and ket vectors, respectively, and $\mathbf{M}$ is a $p \times p$ matrix. These vectors have the $(\alpha = 1, 2, \ldots, p)$ components

$$\begin{aligned}\langle L|_\alpha &= w_\alpha \overline{F_\alpha^*\Gamma_\alpha}, \\ |R\rangle_\alpha &= w_\alpha \overline{F_\alpha}.\end{aligned} \tag{24}$$

In the particular case of a one-dimensional set of fragments characterized by a_α and b_α, (22) and (24) can be transformed using

$$\begin{aligned}\overline{F_\alpha F_\alpha^*} &= \tfrac{1}{2}b_\alpha^2[(1 - \overline{\Gamma_\alpha}) + \text{c.c.}]/[1 - \cos(qa_\alpha)] \\ &= b_\alpha^2 \sum_\ell [D_\alpha(\ell)/\mathcal{D}_\alpha]\sin^2(a_\alpha \ell q/2)/\sin^2(a_\alpha q/2), \\ \overline{F_\alpha^*\Gamma_\alpha} &= b_\alpha(\overline{\Gamma_\alpha} - 1)/[1 - \exp(-iqa_\alpha)], \\ \overline{F_\alpha} &= b_\alpha(1 - \overline{\Gamma_\alpha})/[1 - \exp(iqa_\alpha)].\end{aligned} \tag{25}$$

The first term has been reformulated to the more familiar form for scattering from independent fragments. The matrix $\mathbf{M}$ can be presented in the form of a series that can be explicitly summed in the case of $k \to \infty$:

$$\mathbf{M} = \mathcal{D}(\mathbf{1} + \mathbf{S} + \mathbf{S}^2 + \ldots + \mathbf{S}^{k-2})\mathbf{W} = \mathcal{D}(\mathbf{1} - \mathbf{S})^{-1}\mathbf{W}. \tag{26}$$

According to (23), the elements of the matrix $\mathbf{S}$ are

$$S_{\alpha\beta} = \mathcal{D}W_{\alpha\beta}w_\beta\overline{\Gamma_\beta}. \tag{27}$$

Finally, collecting all the terms on the right-hand side of (21), we obtain the compact expression for the structure factor:

$$\mathcal{S}(q) = (\mathcal{N}/\langle\ell\rangle)\{\langle F^*(1)F(1)\rangle + (\langle L|\mathbf{M}|R\rangle + \text{c.c.})\}. \tag{28}$$

We have used the fact that in sufficiently long systems the total number of fragments, k, can be defined as $\mathcal{N}/\langle\ell\rangle$, where $\langle\ell\rangle$ is the average length of a fragment. According to (11) and (12), $\langle\ell\rangle = \sum_\alpha w_\alpha \overline{\ell_\alpha}$ and $\overline{\ell_\alpha} = \sum_\ell \ell D_\alpha/\mathcal{D}_\alpha$, so $\langle\ell\rangle = \sum_{\alpha,\ell} w_\alpha \ell D_\alpha(\ell)/\mathcal{D}$. The diffuse part of $\mathcal{S}(q)$ scales as the total number of sites $\mathcal{N}$ of the system while the Bragg peak intensity would scale as $\mathcal{N}^2$. We do not discuss the Bragg scattering contribution in this paper.

Pflanz & Moritz (1992) have derived a similar form to (28) for the diffuse scattering in what appears to be the same approximation. However, they did not succeed in finding the closed analytical form and left the result in terms of a recursive problem.

2.3. *Structure factor: general result*

In the previous section, we derived the important general formula for the diffuse scattering from a one-dimensional non-overlapping distribution of differently scattering fragments:

$$\mathcal{S}(q) \propto \langle F^*(1)F(1)\rangle + (\langle L|\mathbf{M}|R\rangle + \text{c.c.}). \tag{29}$$

The first term represents the independent scattering from the various fragments and the last term is the interference term. The involved quantities are defined in the previous

section. The interference term can be very important in some cases, as will be discussed below and illustrated by examples in the following section.

Let us discuss extensions of the above result to dimensions higher than one. The scattering amplitude $\mathcal{A}(q,k)$ of (17) is of a similar form, except that $qa_\alpha\ell$ is replaced by a scalar product $\mathbf{q}\cdot\mathbf{r}_\ell$ and the sum is over all $\mathbf{r}_\ell$ in the α-type region Ω_α. However, for $\mathbf{q}$ in a given direction, say $\mathbf{q}=(0,0,q)$, the perpendicular phase factors are unity at all sites $r_\ell^z = a_\alpha\ell$. The effect of this can be summed to give a total scattering length for each site $b_\alpha(\ell) = \sum_\Omega^\perp b_\alpha$, where Ω represents the plane perpendicular to z and $\perp$ indicates sum over x and/or y for 3D/2D systems. The results are therefore directly applicable for the configuration of infinite stripes or infinite layers of an arbitrary number of different structures in 2D and 3D, respectively. If we consider, for example, a single chain (say along the z direction) of higher-dimensional non-overlapping objects Ω's (squares, discs *etc.* or cubes, spheres *etc.*), the only modification needed in (18) is $F_\alpha(j) = \sum_{r=0}^{\ell_j-1} b_\alpha^j(r)\exp(iqa_\alpha r)$. The shape function $b_\alpha^{\Omega_j} = b_\alpha^j(r)$ can also arise as a result of a projection of an imperfect structure owing to defects (vacancies) – for example, the boundary regions of multilayer systems. The sum can be considered as an infinite sum over a product of a shape function and an infinite sequence of phase factors. The shape function is zero outside the projection on z of Ω and equal to $b_\alpha^j(r)$ within. The Fourier transform can either be represented by the folding of the individual Fourier transforms: the 'form factor' $\hat{b}_\alpha^\Omega(q)$ and the δ functions for the α-type structure. It is also simply the discrete Fourier transform

$$F(j) = \sum_{r=-\infty}^{\infty} b_\alpha^{\Omega_j}(r)\exp(iqa_\alpha r). \tag{30}$$

It thus influences only the independent scattering term (22) and the bra and ket vectors (24). This result takes into account exactly the excluded-volume problem in one direction. There is no excluded-volume problem in having a similar but different parallel chain at a sufficient perpendicular distance so that no overlap occurs between the objects on the two chains. There can be any number of such chains. However, the projections onto the z direction of the shape functions can now overlap – yet the results (29) and (30) are still applicable and exact. We note in passing that Campbell's theorem (Champeney, 1973) states that, for a completely random overlap of the projected elementary shape functions, the interference terms vanish, leaving only the first, independent, scattering term in (28). For very densely packed objects with excluded-volume restrictions in all directions, *i.e.* between the chains [for example, the dense hard-sphere problem (Torquato, 1995; Reiss, Frisch & Lebowitz, 1959)], additional considerations must be taken into account. The present results do not include two- or three-dimensional correlations.

3. Structure factor: examples

Now we shall apply the general formulae derived in the previous section to get the analytical expressions available for interpretation of pertinent experimental situations. We shall treat structures with two alternating substructures, structures separated by empty regions (or orthogonal antiphases in *e.g.* YBCO) and alternating structures with an intermediate separating region of a third type. With these examples as guidelines, it should be easy to derive expressions for other cases of relevance for a particular experiment. Further, we derive the cross section for higher-dimensional objects (ideal), such as hut clusters on a surface, for a rough surface and for a terraced surface.

3.1. *Structure factor for alternating phases, multilayers etc.*

The case of alternating structures is relevant for multilayers of two structures with different lattice constants $a_\alpha = a_1$ and $a_\beta = a_2$ and scattering lengths b_1, b_2. This is also a rather realistic situation in YBCO if the oxygen content is sufficiently high ($x \sim 0.7$–0.8), in which slabs of ortho-I and ortho-II structures alternate. In this case, we have $a_\alpha = a$ and $a_\beta = 2a$ and $b_1 = b_2$. The transfer matrix $\mathbf{W}$ is given by (14). A detailed derivation, which is given in Appendix *A*, finally lead us to

$$\mathcal{S}(q) \propto \tfrac{1}{4}C(q)\Big\{[(1-\overline{\Gamma_1})(1-\overline{\Gamma_2})]/(1-\overline{\Gamma_1}\,\overline{\Gamma_2}) + \text{c.c.}\Big\}, \tag{31}$$

$$\begin{aligned} C(q) &= b_1^2/[1-\cos(qa_1)] + b_2^2/[1-\cos(qa_2)] \\ &\quad - b_1b_2\{1-\cos(qa_1)-\cos(qa_2)+\cos[q(a_2-a_1)]\} \\ &\quad \times \{[1-\cos(qa_1)][1-\cos(qa_2)]\}^{-1}, \end{aligned} \tag{32}$$

where $C(q)$ reflects the internal structure of the two kinds of fragments, while the curley brackets in (31) reflect the effect of their distribution, which is our main concern here. The distributions in widths enter *via*

$$\overline{\Gamma_\alpha} = \sum_\ell D_\alpha(\ell)\exp(iqa_\alpha\ell)\Big/\sum_\ell D_\alpha(\ell). \tag{33}$$

For the special case considered as the introductory example, $a_2 = 2a_1$, one simply gets that $C(q) = b^2/[1-\cos(qa_2)]$ and the dependence on the lattice constant a_1 of the intervening structure disappears.

Further, if we let $\alpha = 1$ represent 'empty' fragments (indicated by the subscript 0), it will be more convenient to incorporate the leftmost particle of an 'empty' fragment into their $\alpha = 2$ counterpart as its rightmost particle. Although the length of an 'empty' fragment was defined above as $\ell \geq 3$ [*cf.* (3)], we could generalize this particular case by allowing $\ell = 1$ (but $\ell \neq 2$). Owing to the rearrangement of particles between the blocks compared with the case considered before, we should

use in this case

$$F_2 = [1 - \Gamma_2 \exp(iqa_2)]/[1 - \exp(iqa_2)], \quad F_0 = 0.$$

The only change with respect to (31)–(32) is a formal substitution:

$$\overline{\Gamma_1} \to \widetilde{\Gamma_0} = \overline{\Gamma_0} \exp(-iqa_2), \quad \overline{\Gamma_2} \to \widetilde{\Gamma_2} = \overline{\Gamma_2} \exp(iqa_2) \tag{34}$$

and $b_1 \to b_0 = 0$. This corresponds to a particle added to the structure fragment, making the 'empty' fragment really empty. This reveals a full (albeit hidden!) symmetry around $x = \frac{1}{2}$ in YBCO: added empty or occupied chains to the ortho-II structure give the same diffuse scattering, except for the overall intensity, of course. We remark that the 'empty' phase might also be the alternatively oriented ortho-II phase. Equations (31) and (32) represent a generalization of the result derived by Fratzl *et al.* (1991).

We now generalize a bit further and go beyond strict alternation, *i.e.* we allow for the 'empty' fragments to be adjacent to each other. This corresponds to the appearance of isolated chains in YBCO in a region with ortho-II ordered domains, which would be realistic for $x \ll \frac{1}{2}$. The transfer matrix is then more complicated and given by (15):

$$\mathbf{W} = \frac{1}{\mathcal{D}w_0} \begin{pmatrix} p & 1 \\ 1 & 0 \end{pmatrix},$$

where $p = 1 - w_2/w_0$ relates to the probability of finding the isolated particles (chains). A straightforward but somewhat lengthy calculation yields

$$\begin{aligned} \mathcal{S}(q) \propto C(q)(w_0/2)\{&[(1 - \widetilde{\Gamma_0})(1 - \widetilde{\Gamma_2}')] \\ &\times (1 - \widetilde{\Gamma_0}\,\widetilde{\Gamma_2}')^{-1} + \text{c.c.}\}, \\ C(q) = b^2/[1 - \cos(qa_2)]. \end{aligned} \tag{35}$$

The result is similar in form to that given by (31) after the following replacements: $\overline{\Gamma_1} \to \widetilde{\Gamma_0} = \Gamma_0 \exp(-iqa_2)$, $\overline{\Gamma_2} \to \widetilde{\Gamma_2} = [p + \overline{\Gamma_2}(1 - p)] \exp(iqa_2)$. We remark that the case of alternating single-periodic and 'empty' fragments can be simply obtained from the example considered above by the change $2 \to 1$. The limiting case, where we assume a random (uniform) distribution of the length of the 'empty' fragments [$\mathcal{D}_0(\ell) = \text{constant}$] was considered separately by Uimin (1994) and the line shape was calculated in detail by Fiig *et al.* (1995) for several important distribution functions $D_\alpha(\ell)$.

Finally, we mention the case that is relevant for alternating multilayers of two different structures, $\alpha = 1$ and 2, where the interface '0' may be modified from the structures of both. This case could also be relevant for the phase-separation problems in the presence of surfactants. Thus, we consider the sequence $1 \cdot 0 \cdot 2 \cdot 0 \cdot 1 \cdot 0 \cdot 2 \cdot 0 \cdot 1 \cdot 0 \cdot 2 \cdot 0 \ldots$. Let us denote the lattice constants and scattering lengths by a_0, a_1, a_2 and b_0, b_1, b_2, respectively. Using the same procedure, we find after considerable algebra (the intermediate formulae that lead to the equation below are given in Appendix *B*)

$$\begin{aligned} \mathcal{S}(q) \propto \Gamma^{-1}\{&C_0(q)(1 - \overline{\Gamma_0})[2 - \overline{\Gamma_1} - \overline{\Gamma_2}] \\ &+ C_1(q)(1 - \overline{\Gamma_1})[1 - \overline{\Gamma_0}(1 + \overline{\Gamma_2})/2] \\ &+ C_2(q)(1 - \overline{\Gamma_2})[1 - \overline{\Gamma_0}(1 + \overline{\Gamma_1})/2] \\ &- C_{01}(q)(1 - \overline{\Gamma_0})(1 - \overline{\Gamma_1}) \\ &- C_{02}(q)(1 - \overline{\Gamma_0})(1 - \overline{\Gamma_2}) \\ &- C_{12}(q)\overline{\Gamma_0}(1 - \overline{\Gamma_1})(1 - \overline{\Gamma_2})/2\} + \text{c.c.}, \end{aligned} \tag{36}$$

where $\Gamma = 2 - \overline{\Gamma_0}(\overline{\Gamma_1} + \overline{\Gamma_2})$ and

$$\begin{aligned} C_\alpha(q) &= \tfrac{1}{2}b_\alpha^2/[1 - \cos(qa_\alpha)], \\ C_{\alpha\beta}(q) &= b_\alpha b_\beta\{1 - \cos(qa_\alpha) - \cos(qa_\beta) \\ &\quad + \cos[q(a_\alpha - a_\beta)]\} \\ &\quad \times \{2[1 - \cos(qa_\alpha)][1 - \cos(qa_\beta)]\}^{-1}. \end{aligned}$$

Equation (36) is a generalization of (31)–(32) for the case when a subsystem '0' is forced to separate subsystems '1' and '2'. In (36) as in (31)–(32), we can identify scattering terms arising from the pure phases ($\propto b_\alpha^2$), which are modified from the independent scattering terms, as well as interference terms. Equation (36) represents a generalization of the formula developed by Fullerton *et al.* (1992), where they consider a particular case of Gaussian interface layers and a finite stack of layers. Further, their formula must be calculated by a numerical solution. They give an excellent discussion of a number of features of relevance for the multilayer scattering. These considerations can be used for the present case, such as the discussed strain effects of the interface layers, which would modify the interface scattering amplitude F_0. Our formula is valid in the limit of very many layers; if one wants the result for a small number of layers using our method, one can go back to (20).

Performing an expansion in q of $\mathcal{S}(q)$ of (31)–(36) in terms of the moments of the distribution functions, $\overline{\ell_\alpha^n} = \sum_\ell \ell^n D_\alpha(\ell)/\mathcal{D}_\alpha$, it can be shown that $\mathcal{S}(q)$ varies with q as $A + Bq^2$ for q approaching zero (or any other Bragg point for the structure). For narrow distributions around some mean length, $\overline{\ell_\alpha}$, the coefficient B may be positive but usually it is negative and the line shape is close to a Lorentzian or a Gaussian one for sufficiently small q. The line shapes were discussed in detail by Fiig *et al.* (1995). In general, $\mathcal{S}(q)$ is finite for $q = 0$ and positive, *i.e.* $A > 0$. Additional correlations between the lengths in the cluster distributions are needed to make $\mathcal{S}(q = 0)$ vanishing, as discussed by Fratzl *et al.* (1991).

3.2. *Structure factor for hut clusters*

Recently, the growth of clusters of one metal on the surface of another metal has been observed to occur as small elongated 'pyramidal' clusters (Mo *et al.*, 1990; Kern *et al.*, 1991), so-called huts, which are well oriented according to the surface structure. They have a distribution of sizes but all have the same slope of facets. This is a situation that can easily be described by our general formalism and represents an example of a case of variable scattering length, as discussed around (30). Let $\alpha = 1$ represent the huts with lattice constant a_1 and scattering length b_1, and $\beta = 2$ the intervening substrate structure with lattice constant a_2 and scattering length b_2. We disregard any deformation of the huts from the perfect structures, although this is known to occur and probably is instrumental for their stability (Shchukin, Borovkov, Ledentsov & Bimberg, 1995; Shchukin, Borovkov, Ledentsov & Kop'ev, 1995, 1996). Two cases will be distinguished: in the first case, the top-layer substrate atoms are considered as non-equivalent, and we assume that these contribute to the scattering amplitude situated between the huts, whereas the structure of those under the huts do not contribute (as a model). In the second case, the substrate is considered as perfect, not contributing to the diffuse scattering at all, and the space between the huts can then be regarded as empty, albeit of integer length in units of the lattice spacing a_2. When scattering in the *surface* plane and $\mathbf{q}$ perpendicular to the long dimension of the huts is considered, the problem can be described by our one-dimensional theory. The only modification according to (30) is then in the scattering length for the individual $\alpha = 1$ type fragments. The hut clusters are supposed to be built of terraces of unit step a_1 and of width νa_1, providing a uniformly terraced slope. An example for $\nu = 1$ is shown in Fig. 2, where two cases, L even and odd, can be distinguished. For L odd ($L = 2\ell + 1$, one atom is allowed on the top), it is easy to find

$$F(2\ell+1) = b_1\{1 + \exp[i(L+1)qa_1] - 2\exp(iqa_1L/2)\exp(iqa_1/2)\} \times [1 - \exp(iqa_1)]^{-2},$$

whereas, for $L = 2\ell + 2$ (two atoms on the top), one obtains

$$F(2\ell+2) = b_1\{1 + \exp[i(L+1)qa_1] - \exp(iqa_1L/2)[1 + \exp(iqa_1/2)]\} \times [1 - \exp(iqa_1)]^{-2}.$$

For any integer ν, there are 2ν hut clusters that are distinct through the number of atoms, j, forming the top terrace. A general formula, which can be derived for any integer ν, is with $L = 2\ell + j$ and $j = 1, \ldots, 2\nu$:

$$F(L) = b_1\{1 + \exp[i(L+1)qa_1] - 2f_j(q,L)\} \times \{[1 - \exp(iqa_1)][1 - \exp(i\nu qa_1)]\}^{-1}, \tag{37}$$

where

$$f_j(q,L) = \tfrac{1}{2}\exp(iqa_1L/2)\{\exp[iqa_1(2\nu - j)/2] + \exp(iqa_1j/2)\}. \tag{38}$$

First, we consider the case where the surface atoms of substrate between the huts and the hut clusters themselves contribute to the scattering amplitude. The matrices $\mathbf{M}$ are given by (45). By means of the auxiliary formulae in Appendix C and (28) and (33), we find the structure factor for hut clusters on a substrate [defining $\widetilde{\Gamma_1} = \overline{\Gamma_1}\exp(iqa_1)$, $\widetilde{\Gamma_2} = \overline{\Gamma_2}\exp(-iqa_1)$]:

$$\begin{aligned} \mathcal{S}(q) \propto\ & C_1(q)\{[4(1-\overline{\gamma})(1-\overline{\gamma}\widetilde{\Gamma_2}) - (1-\widetilde{\Gamma_1})(1-\widetilde{\Gamma_2})] \\ & \times (1-\widetilde{\Gamma_1}\widetilde{\Gamma_2})^{-1} + \overline{\eta} - 1 + \text{c.c.}\} \\ & + \{C_{12}(q)[(1-\overline{\Gamma_2})(1+\widetilde{\Gamma_1} - 2\overline{\gamma}) \\ & \times (1-\overline{\Gamma_1}\,\overline{\Gamma_2})^{-1}] + \text{c.c.}\} \\ & + C_2(q)[(1-\overline{\Gamma_1})(1-\overline{\Gamma_2})/(1-\overline{\Gamma_1}\,\overline{\Gamma_2}) \\ & + \text{c.c.}], \end{aligned} \tag{39}$$

where

$$\begin{aligned} C_1(q) &= (b_1^2/8)\{[1-\cos(qa_1)][1-\cos(\nu qa_1)]\}^{-1}, \\ C_2(q) &= (b_2^2/4)[1-\cos(qa_2)]^{-1}, \\ C_{12}(q) &= (b_1b_2/2)[\exp(iqa_1) + \exp(iqa_2)] \times \{[1-\exp(iqa_1)][1-\exp(iqa_2)]\}^{-1} \times [1-\exp(i\nu qa_1)]^{-1} \end{aligned}$$

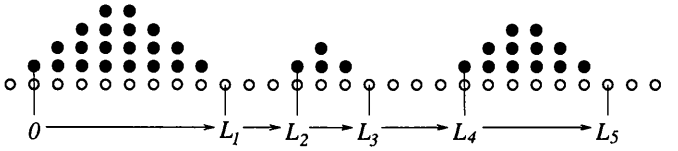

Fig. 2. Huts represented by trapezoidal structures. The allowed number of atoms on the tops is either one (L odd) or two (L even). 0, L_2, L_4 *etc.* label conventional beginnings of the huts, whereas L_1, L_3, L_5 *etc.* do the same for the substrate. Simultaneously, L_1, L_3, L_5 are the widths of the huts, L_2, L_4 *etc.* are the widths of the surface areas.

and

$$\overline{\gamma} = \sum_{\ell=0}\sum_{j=1}^{2}[D_1(2\nu\ell + j)/\mathcal{D}_1]f_j(q, 2\nu\ell + j),$$

$$\overline{\eta} = \sum_{\ell=0}\sum_{j=1}^{2}[D_1(2\nu\ell + j)/\mathcal{D}_1]\cos[(j - \nu)qa_1],$$

where $\overline{\gamma}$ and $\overline{\eta}$ tend to 1 for $q \to 0$.

Next, we consider the case where we assume no scattering from the atoms between the huts. For this case, we put $b_2 = 0$ in (39) and only the first term remains. This is not unexpectedly similar to the alternating filled–empty case given by (31) and (34). However, it is clearly much more complicated both with respect to the prefactor $C_1(q)$ and the way the width distributions enter. A squared cosine factor $C_1(q)$ arises if a unit step slope, $\nu = 1$, is assumed. Equation (39) has a remarkable $1/q^2$ dependence for small q.

3.3. *Structure factor for 'mountains' or a rough (0, 1, 0) surface*

There is another interesting application of our general approach to a partly disordered system. Let us consider a surface consisting of alternating facets of (1, 1, 0) and (−1, 1, 0) planes perfectly elongated in the z direction. We assume that the average slope is zero and that the distribution of ups and downs is equal, *i.e.* on average we have a (0, 1, 0) plane. An example is shown in Fig. 3. We thus consider $\mathbf{q}$ in the x direction. We may choose the horizontal level arbitrarily because shifting it by one vertical unit gives a regular contribution to the scattering amplitude that contributes only to the Bragg scattering, which is not under discussion here. The details of the derivation are given in Appendix *D*. The result is the following very simple and compact formula for the scattering cross section for a rough (0, 1, 0) surface:

$$\mathcal{S}(q) \propto b^2\{2[1 - \cos(qa)]\}^{-2} \times \{(1 - \overline{\Gamma})/(1 + \overline{\Gamma}) + \text{c.c.}\}. \quad (40)$$

Again, the squared cosine factor arises from the assumed unit steps. Upon expansion, this makes $\mathcal{S}(q)$ vary as $1/q^2$ for small q near Bragg peaks.

3.4. *Structure factor for 'terraces'*

We consider finally a system of terraces with arbitrary widths ℓ given by a distribution function $\mathcal{D}(\ell)$. The terraces are separated by steps that are assumed to be only of unit step height, up or down. The steps are supposed to be straight and oriented along $\mathbf{z}$ and we consider again q in the x direction. The terrace heights (m's) and terrace x coordinates (L's) are shown in Fig. 4. The widths of the terraces are related to the coordinates by $\ell_k = L_k - L_{k-1}$. The contribution to the scattering amplitude from the $(k-1)$th and kth terraces is

$$d_1\{m_{k-1}[\exp(iqaL_{k-1}) - \exp(iqaL_k)] + m_k[\exp(iqaL_k) - \exp(iqaL_{k+1})]\},$$

where $d_1 = [1 - \exp(iqa)]^{-1}$. We can rearrange the

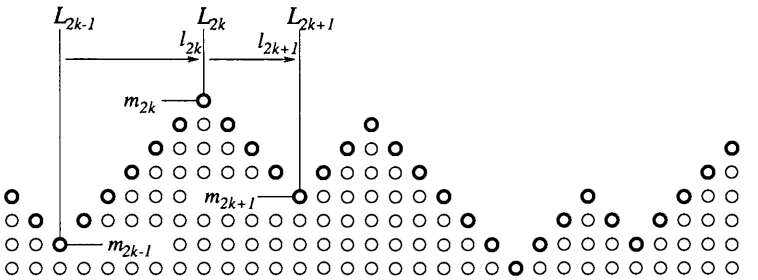

Fig. 3. 'Mountains': the coordinates (L, m) of valleys (odd) and tops (even) are shown.

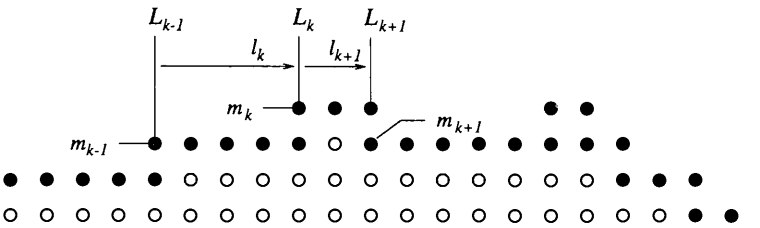

Fig. 4. 'Terraces': x coordinates of steps (L), their (absolute) heights (m) and widths of terraces (ℓ) are shown.

summation order and get

$$\mathcal{A}(q,k) = d_1 \sum_{j=1} \exp(iqaL_j)(m_j - m_{j-1}). \qquad (41)$$

According to our assumption, the height difference is $m_j - m_{j-1} = \pm 1$, which can be considered as an Ising variable, σ_j. For a miscut surface, one generally has only one kind of step and all σ_j have the same sign. However, for generally rough surfaces, the adjacent σ_j and σ_{j+1} can have different signs. Therefore, not only averaging with respect to the widths of terraces must be done, but also with respect to the ensemble of σ's. As in the case of 'mountains', the straightforward calculation of $\mathcal{K}(q,k)$ is preferable (the reader may find more details in Appendix *D*), yielding the scattering for an arbitrarily terraced surface with unit steps

$$\mathcal{S}(q) \propto b^2\{2[1-\cos(qa)]\}^{-1} \times \left\{1 + \sum_{m=1}^{\infty} (\overline{\Gamma}^m + \overline{\Gamma^*}^m) g_m\right\}, \qquad (42)$$

where $\overline{\Gamma} = \sum_\ell \exp(iqa\ell)\mathcal{D}(\ell)/\sum_\ell \mathcal{D}(\ell)$ and $g_m = \overline{\sigma_i \sigma_{i+m}}$. For alternating steps, $g_m = (-1)^m$ corresponding to a stepped (0, 1, 0) surface, it is easily derived that the result becomes similar to the 'mountain' case (40), except that the cosine factor is not squared, as could be expected. Assuming a most simple decoupling scheme for the correlations $g_m = \overline{\sigma_i \sigma_{i+m}} = \overline{\sigma}^2$, one obtains

$$\mathcal{S}(q) \propto b^2\{2[1-\cos(qa)]\}^{-1} \times \{1 + \overline{\sigma}^2[\overline{\Gamma}/(1-\overline{\Gamma}) + \text{c.c.}]\}. \qquad (43)$$

This case corresponds to a rough randomly stepped surface with the average slope $\bar{\sigma}/\bar{\ell}$ relative to the (0, 1, 0) plane, where $\bar{\ell} = \sum_\ell \ell \mathcal{D}(\ell)/\sum_\ell \mathcal{D}(\ell)$ is the mean width of the terraces. If the slope is zero, one cannot determine the width distribution for such a surface in the present scattering set-up. An analytical solution for $\mathcal{S}(q)$ can also be found for an ensemble of correlated σ's. It is based on the transfer-matrix idea, which is very similar to that developed above in (5)–(13). Details of the derivation for the ensemble of σ variables are given in Appendix *E*. The general form of the structure factor for correlated steps on a terraced surface is

$$\mathcal{S}(q) \propto b^2\{2[1-\cos(qa)]\}^{-1}\left\{1 + \left([\overline{\Gamma}/(1-\overline{\Gamma})] \times [(1-t)(1-\overline{\Gamma}) + t\overline{\sigma}^2]/[1-(1-t)\overline{\Gamma}] + \text{c.c.}\right)\right\}, \qquad (44)$$

where t is the independent matrix element of the σ transfer matrix. In a special decoupling regime, $t = 1$, (44) reduces to the form of (43). For zero slope, $\bar{\sigma} = 0$ and $t = 2$, we get the case for alternating steps on a (0, 1, 0) surface, discussed above.

4. Concluding remarks

We have derived the exact result for the (X-ray or neutron) diffuse scattering from different non-overlapping structures in one dimension. The scattering from real objects such as multidomains, multilayers and rough or decorated surfaces is also explicitly given. We have included the effects of the internal structure of the objects. Since the diffuse scattering is emphasized differently near different Bragg peaks, it is possible to use this to single out various distribution functions. It is therefore important to extend the study of diffuse scattering, traditionally done as small-angle scattering, to include that near other Bragg peaks. With these tools, it should be possible to analyze scattering data from a variety of nano- or mesoscale structures and thereby derive the statistical distribution of the equilibrium or non-equilibrium patterns of interest. The scale that can be investigated is set by the attainable experimental q-space resolution. It is required that real-space techniques (or theory) has identified the typical structural elements. The two kinds of technique are therefore in this case necessary and complementary. We have included the possibility of specifying the distribution of all involved structures; however, we have not included correlation between the neighboring patterns. This is sometimes important as well and will be the subject of further work.

It is a pleasure to thank M. Nielsen for discussions concerning nanoscale surface phenomena and D. McMorrow and D. Smilges for bringing the references to Pflanz *et al.* and Fullerton *et al.* to our attention, and H. Richards for linguistic comments. We thank M. Schreckenberg for continued interest. GU acknowledges support by the research program Sonderforschungsbereich 341, Köln-Aachen-Jülich, and the hospitality at Risø National Laboratory, where this work was initiated.

APPENDICES

For the purpose of illustration, we present in the Appendices the set-up formulae for a calculation of the cross section. The algebra involved is straight forward but quite long and tedious. Use of *Mathematica* is of considerable help.

APPENDIX *A*
Alternating phases

For the case of alternating phases, (14) and (27) give, with $\alpha = 1$ and $\beta = 2$,

$$\mathbf{S} = \begin{pmatrix} 0 & \overline{\Gamma_2} \\ \overline{\Gamma_1} & 0 \end{pmatrix}; \quad \mathbf{M} = [2/(1-\overline{\Gamma_1}\,\overline{\Gamma_2})]\begin{pmatrix} \overline{\Gamma_2} & 1 \\ 1 & \overline{\Gamma_1} \end{pmatrix}. \qquad (45)$$

Equations (24) now read

$$\langle L| = \tfrac{1}{2}\left(\frac{b_1(\overline{\Gamma_1}-1)}{1-\exp(-iqa_1)}, \frac{b_2(\overline{\Gamma_2}-1)}{1-\exp(-iqa_2)}\right),$$

$$|R\rangle = \tfrac{1}{2}\begin{pmatrix} b_1(1-\overline{\Gamma_1})/[1-\exp(iqa_1)] \\ b_2(1-\overline{\Gamma_2})/[1-\exp(iqa_2)] \end{pmatrix},$$

which results in the following requisite for calculating $\mathcal{S}(q)$ with (28):

$$\langle F^*(1)F(1)\rangle = \tfrac{1}{4}\left\{\left(\frac{b_1^2(1-\overline{\Gamma_1})}{1-\cos(qa_1)} + \frac{b_2^2(1-\overline{\Gamma_2})}{1-\cos(qa_2)}\right) + \text{c.c.}\right\}$$

$$\mathbf{M}|R\rangle = \begin{pmatrix} \dfrac{b_1}{1-\exp(iqa_1)} - \dfrac{(1-\overline{\Gamma_2})}{1-\overline{\Gamma_1}\,\overline{\Gamma_2}} f_{12} \\ \dfrac{b_2}{1-\exp(iqa_2)} + \dfrac{(1-\overline{\Gamma_1})}{1-\overline{\Gamma_1}\,\overline{\Gamma_2}} f_{12} \end{pmatrix},$$

where $f_{12} = b_1/[1-\exp(iqa_1)] - b_2/[1-\exp(iqa_2)]$.

APPENDIX *B*
Multilayers with intervening layers: ... 1 · 0 · 2 · 0 · 1 · 0 · 2 ...

The matrix elements of **W** are in this case $W_{10} = W_{20} = W_{01} = W_{02} = 2/\mathcal{D}$. Equations (22), (24) give

$$\langle L| = \tfrac{1}{4}\left(\frac{2b_0(\overline{\Gamma_0}-1)}{1-\exp(-iqa_0)}, \frac{b_1(\overline{\Gamma_1}-1)}{1-\exp(-iqa_1)}, \frac{b_2(\overline{\Gamma_2}-1)}{1-\exp(-iqa_2)}\right)$$

$$|R\rangle = \tfrac{1}{4}\begin{pmatrix} 2b_0(1-\overline{\Gamma_0})/[1-\exp(iqa_0)] \\ b_1(1-\overline{\Gamma_1})/[1-\exp(iqa_1)] \\ b_2(1-\overline{\Gamma_2})/[1-\exp(iqa_2)] \end{pmatrix}$$

$$\langle F^*(1)F(1)\rangle = \left[\frac{b_0^2}{4}\frac{1-\overline{\Gamma_0}}{1-\cos(qa_0)} + \frac{b_1^2}{8}\frac{1-\overline{\Gamma_1}}{1-\cos(qa_1)} + \frac{b_2^2}{8}\frac{1-\overline{\Gamma_2}}{1-\cos(qa_2)}\right] + \text{c.c.}$$

$$\mathbf{S} = \begin{pmatrix} 0 & \overline{\Gamma_1}/2 & \overline{\Gamma_2}/2 \\ \overline{\Gamma_0} & 0 & 0 \\ \overline{\Gamma_0} & 0 & 0 \end{pmatrix}$$

$$\mathbf{M} = \frac{2}{1-\frac{1}{2}\overline{\Gamma_0}(\overline{\Gamma_1}+\overline{\Gamma_2})}\begin{pmatrix} (\overline{\Gamma_1}+\overline{\Gamma_2})/2 & \dfrac{1}{\overline{\Gamma_0}} & \dfrac{1}{\overline{\Gamma_0}} \\ 1 & 1 & \dfrac{\overline{\Gamma_0}}{\overline{\Gamma_0}} \\ 1 & \dfrac{\overline{\Gamma_0}}{\overline{\Gamma_0}} & 1 \end{pmatrix}.$$

APPENDIX *C*
Hut clusters

The width of a hut is not restricted. The huts are assumed to have uniform slope determined by terraces of width νa_1. With such an assumption, we even allow a hut to be of unit height but in this last case its width cannot exceed $2\nu a_1$. We remind readers that all the possible geometrical configurations of hut clusters as well as the distances between them must obey some probabilistic distributions.

In order to obtain the set-up formulae, we note that upon averaging the function $f_j(q, L)$ exhibits the following properties:

$$f_j(q,L)\exp(-iLqa_1) = f_j^*(q,L)\exp(i\nu qa_1),$$

$$f_j(q,L)f_j^*(q,L) = \tfrac{1}{2}\{1+\cos[(j-\nu)qa_1]\}.$$

The set-up formulae are then

$$\langle F(1)F^*(1)\rangle = b_1^2\frac{\{[2+\overline{\Gamma_1}\exp(iqa_1) - 4\overline{\gamma}+\overline{\eta}] + \text{c.c.}\}}{8[1-\cos(qa_1)][1-\cos(\nu qa_1)]} + b_2^2\frac{\{1-\overline{\Gamma_2}+\text{c.c.}\}}{4[1-\cos(qa_2)]}$$

$$\langle L| = \tfrac{1}{2}\left(\frac{b_1\exp(-iqa_1)[1+\overline{\Gamma_1}\exp(iqa_1) - 2\overline{\gamma_1}]}{[1-\exp(-iqa_1)][1-\exp(-i\nu qa_1)]}, \frac{b_2(\overline{\Gamma_2}-1)}{1-\exp(-iqa_2)}\right)$$

$$|R\rangle = \tfrac{1}{2}\begin{pmatrix} b_1[1+\overline{\Gamma_1}\exp(iqa_1) - 2\overline{\gamma_1}]/[1-\exp(iqa_1)] \\ [1-\exp(i\nu qa_1)] \\ b_2(1-\overline{\Gamma_2})/[1-\exp(iqa_2)] \end{pmatrix}.$$

APPENDIX *D*
'Mountains' or rough (0, 1, 0) surface and terraces

The defining elements of the 'mountain' case are shown in Fig. 3. The sum over scattering centers whose vertical projection lies between the valley L_{2k-1} and the top L_{2k} is

$$\sum_{n=L_{2k-1}}^{L_{2k}-1}[m_{2k-1} + (n - L_{2k-1})]\exp(inqa)$$
$$= \exp(iqaL_{2k-1})\sum_{n=0}^{\ell_{2k}-1}(m_{2k-1}+n)\exp(inqa)$$
$$= \exp(iqaL_{2k-1})\{m_{2k-1}S_1(\ell_{2k}) + S_2(\ell_{2k})\}, \quad (46)$$

where

$$S_1(\ell) = d_1[1-\exp(iqa\ell)],$$
$$S_2(\ell) = d_2[1-\exp(iqa\ell)] - d_1\ell\exp(iqa\ell),$$
$$d_1 = 1/[1-\exp(iqa)], \quad d_2 = d_1^2\exp(iqa).$$

The contribution of the centers between the top L_{2k} and the next valley L_{2k+1} is

$$\sum_{n=L_{2k}}^{L_{2k+1}-1} [m_{2k} - (n - L_{2k})] \exp(inqa)$$
$$= \exp(iqaL_{2k}) \{m_{2k} S_1(\ell_{2k+1}) - S_2(\ell_{2k+1})\}. \quad (47)$$

We can rearrange (46) and (47) to the following form:

$$\exp(iqaL_{2k-1}) \{(d_1 m_{2k-1} + d_2)[1 - \exp(iqa\ell_{2k})]$$
$$- d_1 \ell_{2k} \exp(iqa\ell_{2k})\}$$

and

$$\exp(iqaL_{2k}) \{(d_1 m_{2k} - d_2)[1 - \exp(iqa\ell_{2k+1})]$$
$$+ d_1 \ell_{2k+1} \exp(iqa\ell_{2k+1})\}.$$

Because $m_{2k} = m_{2k-1} + \ell_{2k}$ and $m_{2k+1} = m_{2k} - \ell_{2k+1}$, these two contributions can be resummed to

$$\exp(iqaL_{2k-1})(d_2 + d_1 m_{2k-1}) - 2d_2 \exp(iqaL_{2k})$$
$$+ \exp(iqaL_{2k+1})(d_2 - d_1 m_{2k+1}).$$

Summation performed over all the 'mountain' peaks finally results in the following scattering amplitude expression:

$$\mathcal{A}(q,k) = -d_2 \sum_k (-1)^k \exp(iqaL_k)$$
$$= d_2 \{\exp(iqa\ell_1) - \exp[iqa(\ell_1 + \ell_2)]$$
$$+ \exp[iqa(\ell_1 + \ell_2 + \ell_3)] - \ldots\}. \quad (48)$$

The direct calculation of $\mathcal{K}(q,k)$ is perhaps easier here than using the general formula (28). We obtain

$$\mathcal{K}(q,k) = \mathcal{K}(q,k-1) + [1 - \cos(qa)]^{-2}$$
$$\times \left\{1 - \overline{\Gamma}/(1 + \overline{\Gamma}) - \overline{\Gamma^*}/(1 + \overline{\Gamma^*})\right\},$$

which gives the compact formula (40) for the scattering from a rough (0, 1, 0) surface.

In the case of 'terraces', the scattering amplitude differs slightly from (48):

$$\mathcal{A}(q,k) = d_1 \sum_k \exp(iqaL_k)\, \sigma_k$$
$$= d_1 \{\exp(iqa\ell_1)\, \sigma_1 + \exp[iqa(\ell_1 + \ell_2)]\, \sigma_2$$
$$+ \exp[iqa(\ell_1 + \ell_2 + \ell_3)]\, \sigma_3 + \ldots\}.$$

Then,

$$\mathcal{K}(q,k) = \mathcal{K}(q,k-1) + \{2[1 - \cos(qa)]\}^{-1}$$
$$\times \left\{1 + \left(\sum_{m=1} \overline{\Gamma}^m g_m + \text{c.c.}\right)\right\},$$

which results in (42).

APPENDIX *E*
Transfer-matrix formulation of σ correlations

Let N be the total number of unit steps among which N_+ and N_- are positive and negative, respectively, and

$$w_+ = N_+/N, \quad w_- = N_-/N.$$

Now, let us consider N pairs of adjacent steps, which may appear in the following sequences: $(++)$, $(+-)$, $(-+)$ and $(--)$. The corresponding numbers will be denoted as $N_{\sigma_1\sigma_2}$ and a definition of double-σ probabilities is a complete analog of (6):

$$w_{\sigma_1\sigma_2} = N_{\sigma_1\sigma_2}/N.$$

We define a decoupling scheme as in (7):

$$w_{\sigma_1\sigma_2} = w_{\sigma_1} T_{\sigma_1\sigma_2} w_{\sigma_2},$$

where $\mathbf{T}$ is the 2×2 symmetric unnormalized 'transfer' matrix

$$\mathbf{T} = \begin{pmatrix} T_{++} & T_{+-} \\ T_{-+} & T_{--} \end{pmatrix},$$

whose elements satisfy two equations [*cf.* (8)]:

$$\sum_{\sigma'} T_{\sigma\sigma'} w_{\sigma'} = 1. \quad (49)$$

The decoupling scheme $g_m = \overline{\sigma}^2$, mentioned above in connection with (43), corresponds to a transfer-matrix description with $T_{++} = T_{--} = T_{+-} = 1$ and $w_+ = (1 + \overline{\sigma})/2$, $w_- = (1 - \overline{\sigma})/2$. The correlation function, g_m, which enters the general equation (42), can be expressed as

$$g_m = \sum_{\sigma_1} \cdots \sum_{\sigma_{m-1}} [w_{+\sigma_1 \ldots \sigma_{m-1}+} + w_{-\sigma_1 \ldots \sigma_{m-1}-}$$
$$- w_{+\sigma_1 \ldots \sigma_{m-1}-} - w_{-\sigma_1 \ldots \sigma_{m-1}+}].$$

A knowledge of $w_{\sigma_0\sigma_1 \ldots \sigma_m}$ would allow a calculation of the right-hand side of (42). The analog of (9) now reads

$$w_{\sigma_0\sigma_1 \ldots \sigma_m} = w_{\sigma_0} T_{\sigma_0\sigma_1} w_{\sigma_1} T_{\sigma_1\sigma_2} w_{\sigma_2} \cdots w_{\sigma_{m-1}} T_{\sigma_{m-1}\sigma_m} w_{\sigma_m}. \quad (50)$$

The important sum

$$G_{\sigma\sigma'} = \sum_{m=1} \sum_{\sigma_1} \cdots \sum_{\sigma_{m-1}} \overline{\Gamma}^m w_{\sigma\sigma_1 \ldots \sigma_{m-1}\sigma'},$$

which enters (42), can be rewritten by introducing the 2×2 matrix $\mathbf{\Theta}$, whose matrix elements are defined as $\Theta_{\sigma_1\sigma_2} = T_{\sigma_1\sigma_2} w_{\sigma_2}$. According to the decoupling scheme

of (50), we obtain

$$G_{\sigma\sigma'} = w_\sigma \sum_{m=0} \left[(\overline{\Gamma}\Theta)^m \overline{\Gamma}\mathbf{T}\right]_{\sigma\sigma'} w_{\sigma'} = w_\sigma \left[(\mathbf{1} - \overline{\Gamma}\Theta)^{-1}\overline{\Gamma}\mathbf{T}\right]_{\sigma\sigma'} w_{\sigma'}. \quad (51)$$

The matrix on the right-hand side of (51) has the form

$$(1 - \overline{\Gamma}\Theta)^{-1}\overline{\Gamma}\mathbf{T} = \{\overline{\Gamma}/[(1 - \overline{\Gamma}T_{++}w_+)(\mathbf{1} - \overline{\Gamma}T_{--}w_-) - \overline{\Gamma}^2 T_{+-}T_{-+}w_+w_-]\} \times \begin{pmatrix} T_{++} + \tau_- & T_{+-} \\ T_{-+} & T_{--} + \tau_+ \end{pmatrix},$$

where $\tau_\pm = \overline{\Gamma}(T_{+-}T_{-+} - T_{++}T_{--})w_\pm$. Using the relation between g_m's and G, *i.e.*

$$\sum_{m=1} \overline{\Gamma}^m g_m = G_{++} + G_{--} - G_{+-} - G_{-+},$$

and expressing T_{++} and T_{--}, using (49), through $t = T_{+-}$, we finally obtain (44).

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