Optical Transforms of Disordered Systems Displaying Diffuse Intensity Loci

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(Received 24 November 1986; accepted 16 February 1987)

Abstract
A method is described of synthesizing a real-space distribution of scattering points which will give rise to virtually any required diffraction pattern. The distribution may be used in the form of an optical diffraction screen to give an immediate visual check on both the real-space and reciprocal-space distributions. The method is applied to two examples which exhibit electron diffraction patterns with diffuse intensity distributed in the form of complex loci in reciprocal space: certain transition-metal niobium sulfide intercalates in which ordering of metal ions and vacancies occurs; and the high-temperature phase of 1T tantalum disulfide in which phenomena due to charge-density waves (CDW) and accompanying periodic lattice distortions (PLD) are observed. It is shown how useful statistical information concerning the local ordering may be obtained from the resulting lattice realizations.

1. Introduction
Optical transform methods (see Lipson, 1973) have been used widely as aids to the interpretation of diffraction phenomena for many years. Despite the advent of powerful computers which can in principle be used to calculate the most complex diffraction patterns the usefulness of the optical method still remains. Perhaps the most important reason for this is that the optical method provides the experimenter with an immediate visual check on both the reciprocal-space and the real-space distributions. Computationally it is possible to obtain the Fourier transform of a mathematical model describing an object even when the model may not actually be physically realizable. For the optical method this possibility is eliminated since the model must of necessity be available in the form of a diffraction screen. While such a model does not necessarily represent the only object that will produce the observed diffraction pattern, it certainly does represent a possible one. At the very least, a realization of a model which gives an optical transform similar to an observed X-ray or electron diffraction pattern must provide a useful starting point in trying to interpret the observations.

In recent years we have sought to develop methods to allow the routine production of optical diffraction screens for use as aids in the interpretation of X-ray diffraction patterns (Welberry & Jones, 1980; Welberry & Carroll, 1982). In the examples to which we have so far applied these methods, the observed scattering effects were caused by substitutional or displacement disorder, in which short-range order was present (e.g. Welberry, Jones & Epstein, 1982; Welberry, 1982). The optical models used for this purpose consisted of photographic images of computer-generated lattices into which disorder had been introduced either at the surface during growth by a simple growth algorithm, or subsequently in the bulk by iterative Monte Carlo procedures. In either case the nearest-neighbour interaction energies formed the basis of the method.

More recently our interest has been drawn to a number of disordered systems which are characterized by the presence of diffuse intensity confined to a geometrical locus in reciprocal space. Such diffuse loci have been reported by numerous workers in a wide range of materials. They may result, for example, from spinodal decomposition in alloys (e.g. de Fontaine, 1984); from the ordering of metal ions and vacancies in some intercalation compounds (e.g. de Ridder, Van Tendeloo, Van Dyck & Amelinckx, 1975); and from compounds containing charge-density waves (e.g. Wilson, di Salvo & Mahajan, 1975). Particularly good examples of diffuse loci are to be found in the paper by de Ridder et al. (1977). The diffuse intensity contours are often remarkably sharp, indicating a type of order extending over considerable distances in real space. Such phenomena are not consistent with any simple model based on near-neighbour interactions but are believed to be attributable to the presence of long-range interactions due to singularities in the electronic part of the interaction energy. The state of order may be said to be intermediate between that of the classical concept of short-range order, and that of the perfect crystal which possesses complete long-range order. For the case of substitutional disorder de Ridder has used the term 'transition state' to describe this state.

In order to produce realizations of a model crystal displaying diffraction patterns of the kind mentioned above, our previous methods which relied on purely short-range interactions are quite unsuitable, and
consequently we sought to develop additional methods which would allow this type of problem to be simulated. For current purposes we invoke the concept of 'concentration waves' for the case of substitutional disorder and 'displacement waves' for phenomena which are evidently of a displacive nature. Such concepts are not new but have been used, for example, in theoretical studies by de Fontaine (1972, 1973). In this paper we describe the production of optical diffraction screens by making use of these concepts, and describe in some detail examples of how the lattice realizations so produced may be used to investigate certain aspects of the real-space distribution.

2. Lattice realizations

We represent the lattice by an array of points with indices \((i, j)\), and specify a set of variables, \(X_{ij}\) say, which define some property which varies from site to site. In previous models \(X_{ij}\) were treated as random variables which mutually interacted via near-neighbour interactions. In the present case, each \(X_{ij}\) is considered to be the sum of simple sinusoidal perturbations of the lattice. In this section we describe how such perturbations may be used to produce lattice realizations which give diffraction patterns displaying the desired effects.

A single modulation of the real-space lattice by a wave of the form \(\exp(2\pi ir.s_p)\) produces in diffraction space a set of satellite peaks symmetrically disposed about each reciprocal-lattice vector \(S\) at the points \(S \pm ms_p\), where \(m\) is an integer. For displacement waves of very small amplitude, or for compositional waves, only the \(m = 1\) peaks have significant intensity. If the modulation is a compositional one, i.e. involving a variation in scattering power, then the satellite intensities are independent of the direction of vector \(S\), but if the modulations involve lattice displacements, then the satellite intensity varies with \(|d.(S \pm s_p)|\), where \(d\) is a unit vector in the direction of the displacement. This direction-dependent property is characteristic of displacement phenomena: but for small displacements the two different types of modulation otherwise produce very similar diffraction effects. (The analogy may be drawn with the AM or FM modulation of radio waves.)

Conversely, if intensity is observed as satellite peaks in reciprocal space it may be inferred that a modulation of the lattice, by a wave of the appropriate wave vector, exists in real space. In general no information on the phase of this wave may be obtained, but if the modulation is incommensurate with the basic lattice the phase may be of no importance, since the maximum of the modulation will necessarily coincide with a node of the lattice at one particular position in the lattice, but elsewhere it will not. Thus, the real-space distribution may be obtained by the direct Fourier inversion of the reciprocal-space distribution with the assumption of an arbitrary phase for the satellite peaks.

We can extend this concept to diffraction patterns which display scattering along continuous loci in reciprocal space if we assume that the scattering within each elemental volume \(dS\) results from a number, \(n_s\), of modulations with wave vectors ranging between \(S\) and \(S + dS\). All other things being equal, the total intensity within each elemental volume will be proportional to \(n_s\) and so in this way account may be taken of the distribution of intensity along the locus. The modulation of the real-space lattice can then be considered to be the sum of contributions from all such waves, the contribution of each being obtainable as before by direct Fourier inversion with the assumption of arbitrary phase. While the particular form of the diffuse locus and the variation of intensity along it will impose certain characteristics of order on the resulting real-space distribution, the choice of phase for each wave will result in a degree of randomness (or disorder) also.

Although structures which are modulated by a single wave often show satellite reflections which are just as sharp as the main Bragg reflections, indicating that the wave is coherent over the whole crystal, the diffuse loci under present discussion appear to be always of a finite width, substantially greater than the typical Bragg-peak width. This would require that wave vectors be distributed not only along the length of the locus but also within a finite distance either side of it. To produce a realization of a lattice containing a sufficient number of modulations to give a reasonably continuous distribution of intensity of this kind, a considerable amount of computing time would be required. An alternative way to treat this problem is to consider that the finite width of the locus is due to the finite extent of the coherence of the wave in real space. That is to say, each modulating wave is multiplied by a factor which limits the extent of the wave in real space. For convenience we use a simple Gaussian for this function and the combined effect on the \(X_{ij}\) of all these modulations is then given by

\[
X_{ij} = \sum_p \exp \left[ -\frac{(r - r_p)^2}{2\sigma_p^2} \right] \exp \left[ 2\pi i(r.s_p + \varphi_p) \right],
\]

where \(r = ia + jb\) defines the particular lattice site, \(r_p\) is the centre of the coherence region in real space of a particular modulation \(p\), \(\sigma_p\) is the half-width of the range of coherence, \(s_p\) is the wave vector for the particular wave, and \(\varphi\) is a random phase.

Use of this particular form is convenient since each modulation need only be applied to the real lattice over a comparatively small area. Consequently, for the same expenditure of computer time a greater number of distinct wave vectors can be utilized, producing a more continuous distribution of intensity in reciprocal space. The form in (1) of the real-space modulation
also means that the contribution to the intensity in reciprocal space will be in the form of a pair of satellite peaks convoluted with the Fourier transform of the Gaussian envelope; i.e. a diffuse spot of half-width inversely proportional to \( \sigma_p \).

In the simulations which are used to illustrate this paper we have found it convenient to use a real-space lattice consisting of 512 \( \times \) 512 sites. We apply each modulation to the lattice in the range of \( \pm 2 \sigma_p \) from a randomly chosen centre. If, for example, \( \sigma_p \) is equal to eight cell spacings, then the modulations are only applied over an area of 32 \( \times \) 32 unit cells, i.e. 1/256 of the total area. In this case a typical realization, utilizing about \( 1.5 \times 10^5 \) individual modulating waves, would on average mean that each real-space lattice site was subjected to modulation by \( \sim 600 \) waves. For the same computation time a higher value of \( \sigma_p \) would mean that a correspondingly smaller number of individual waves could be used, but each lattice site would on average be subject to approximately the same number of waves.

To produce realizations of lattices with perturbations generated in this way we make use of the same procedures as used for previous models (e.g. Welberry & Carroll, 1980). An Optronics P-1700 Photomation system is used as an accurate fine-scale plotter to record the desired image directly onto photographic film. Since the basic increment on the machine is 12-5 \( \mu \)m, 512 unit cells may be written across the film using 16 plotter units per cell. This scale allows reasonable scope for displacement perturbations to be represented but since each increment is \( \sim 6\% \) of the cell spacing, for some purposes it has been found better to use a smaller number of cells with a larger number of steps per cell. Although at any time only a single size of dot may be written, variations in scattering power may be achieved either by writing the dots with variable density or by superposing several centred on adjacent grid points to achieve dots of larger radii (see Welberry & Jones, 1980). In the examples given in this paper we have used only a single dot size.

The array of variables, \( X_{ij} \), produced in the way described above will be on an arbitrary scale, depending on the number of individual modulations used. It is useful to renormalize them so that they have zero mean and unit variance, by computing averages over the whole lattice realization. It should be noted that irrespective of the actual distribution of the individual perturbations (in our case sinusoidal) affecting a single \( X_{ij} \), the central limit theorem states that the distribution of the \( X_{ij} \) will be asymptotically normal (e.g. Spiegel, 1961). For the realizations presented here in which \( \sim 600 \) independent modulations contribute to any individual \( X_{ij} \), the normalized variables may be assumed to a good approximation to be zero-mean unit-variance Gaussian variables.

For use in examples in which binary rather than continuous \( X_{ij} \) are required, we can simply replace each \( X_{ij} \) by a corresponding binary variable \( Y_{ij} \), say, where \( Y_{ij} = 1 \) for \( X_{ij} \geq X_T \), and \( Y_{ij} = 0 \) for \( X_{ij} < X_T \). For \( X_T = 0 \) the ‘site occupancy’ for the binary variables is 50\%, but for \( X_T \neq 0 \) other site occupancies can be achieved. This method of converting Gaussian to binary variables and the resultant effect on the correlation properties has been discussed by Welberry & Carroll (1982).

3. Example involving substitutional disorder

De Ridder et al. (1977) discussed examples of the diffuse scattering in a number of transition-metal niobium sulfide intercalates. In these, the diffuse scattering is attributed to the ordering of the transition-metal ions and vacancies on the octahedral sites between sandwiching layers of NbS\(_2\). In projection down the c axis the problem reduces to that of placing on a simple triangular lattice the required arrangement of metal ions and vacancies which will give a diffraction pattern corresponding to the observed one. For present purposes we concentrate on an example diffraction pattern given in Fig. 2 of de Ridder et al. (1977). In this example the scattering is in the form of diffuse hexagons, forming a honeycomb mesh, with the main Bragg reflections occurring in the centre of each hexagon.

For convenience the simulations presented in this paper have been produced using 50\% occupancy of metal ions since the conversion of Gaussian to binary variables is simpler, but this is by no means a necessary restriction on our method. In Figs. 1(a), (b) and (c) we show optical diffraction patterns of lattice realizations constructed in the way described in § 2, and in Figs. 2(a), (b) and (c) we show small representative portions of the corresponding real-space distributions. Reference to Fig. 2 of de Ridder et al. (1977) will immediately show the similarity of these diffraction patterns to the observed electron diffraction pattern. In each of the examples the realization was constructed by including concentration waves with wave vectors uniformly distributed along the unique segments of the diffuse hexagons. In the three examples (Figs. 2a–c) the only difference is in the value of \( \sigma_p \), the coherence range of the waves in real space. In the original figure of de Ridder et al. it was noticeable that the intensity along the locus was not completely uniform but tended to be greatest at the intersection points (h/3, k/3 and symmetry-related points). Consequently we show in Fig. 1(d) the diffraction pattern of an example realization in which additional waves, all having these particular special wave vectors, have been included in the construction. In this case one-third of the wave vectors were placed at the (h/3, k/3) etc. points, and the remaining two-thirds
were uniformly distributed around the hexagon. For comparison we also show in Fig. 1(e) the diffraction pattern of a realization in which additional waves were similarly included but at the special wave vectors \((0, k/2)\) etc. Small representative portions of these realizations are shown in Figs. 2(d) and (e) respectively.

Having been constructed as described above, the real-space distributions (small representative portions of which are shown in Fig. 2) are available for further scrutiny. It is interesting to analyse these in terms of the local ordering. In Fig. 3(a) we show the 13 different symmetry-inequivalent configurations which are possible on a seven-point cluster on the triangular lattice. In addition to the 13 types of cluster shown there is a related set of 13 in which the black dots (metal ions) and white dots (vacancies) are interchanged. Since in our case the occupancy was 50\% the two sets of clusters occur equally frequently. In Table 1 we list the frequencies with which each of these clusters occurs in the realizations of Fig. 2 together with similar values for some intermediate examples which have not been illustrated. Several points emerge from a consideration of these frequencies.

(i) It is noticeable that certain of the clusters dominate the frequency spectrum while others occur very infrequently.

(ii) In the progression to higher order (i.e. higher \(\sigma_p\)) there is relatively little change in the local ordering, the main change taking place between the first two rows in the table \((\sigma_p = 2; \sigma_p = 4)\). In general, the dominant configurations such as 13, 10 and 2 increase at the expense of other less-frequent ones such as 6, 8 and 12 in the progression to higher order.

(iii) Some configurations, including ones which are relatively frequent, such as 4 and 9, appear to be very insensitive to the degree of order as measured by \(\sigma_p\).

The row labelled 8A in Table 1 gives the frequencies for the realization in which additional waves with wave vectors \((h/3, k/3)\) etc. have been included, corresponding to Fig. 2(d). Here it is seen that the frequencies of configurations 2 and 13 have markedly increased (cf. row labelled 8). Furthermore, when a lattice realization is produced with waves only at these special wave vectors the structure that is generated consists of large domains of the perfect superstructure depicted in Fig. 3(b) (together with domains of its reverse - black and white interchanged). This superstructure is seen to consist entirely of configurations 2 and 13, and the values of \(m_A\) and \(m_B\) for it are \(\frac{1}{3}\) and \(\frac{2}{3}\) respectively. It should be noted that for the reverse structure in which black and white are interchanged the values for \(m_A\) and \(m_B\) are reversed. If a site occupancy other than 50\% is chosen, one or other of

![Fig. 1. Optical diffraction patterns of the realizations shown in Fig. 2 which were used to represent the ordering of metal atoms and vacancies in transition-metal niobium sulfide intercalates. (a), (b) and (c) contained concentration modulations with wave vectors uniformly distributed around the hexagon locus; additional modulations were included at the corners of the hexagon in (d), and in the middle of the edges in (e). The values of the parameter, \(\sigma_e\), defining the coherence range in units of the real-space cell repeat distance were: (a) 2; (b), (d), (e) 8; (c) 32.](image-url)
Table 1. Frequencies ($\times 10^4$) with which each of the cluster types depicted in Fig. 3 occur in the lattice realizations that were used for the optical diffraction examples of Fig. 1, together with some additional intermediate examples

For each of the examples the expected frequencies of a particular cluster and the cluster in which dark and light circles have been interchanged are the same, and the figure quoted is the mean of the two. The numerical part of the sample label given in the leftmost column is the value of $\sigma_r$ in units of the cell translation distance. The examples labelled 8A and 8B correspond to the realizations of Figs. 2(d) and (e) in which additional waves with special wavevectors $(h/3, k/3)$ etc. and $(0, k/2)$ etc. were included. See text for further details.

<table>
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<th>Cluster type</th>
<th>Sample 1</th>
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<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
<th>9</th>
<th>10</th>
<th>11</th>
<th>12</th>
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<td>0</td>
<td>126</td>
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<td>16</td>
<td>54</td>
<td>8</td>
<td>189</td>
<td>288</td>
<td>1</td>
<td>74</td>
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<td></td>
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<td>0</td>
<td>121</td>
<td>0</td>
<td>14</td>
<td>59</td>
<td>5</td>
<td>189</td>
<td>301</td>
<td>1</td>
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<td>0</td>
<td>12</td>
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<td>0</td>
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<td>4</td>
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these two superstructures begins to dominate the real-space distribution and for a site occupancy of 33.3% only the superstructure seen in Fig. 3(b) is observed.

Similarly, the realizations which were made to give additional intensity at the centres of the sides of the diffuse hexagons [i.e. at $(0, k/2)$ etc.], corresponding to Fig. 2(e), gave rise to an increase in the frequency of configuration 10 (see row labelled 8B). When a realization was made with only these waves included, the structure generated consisted entirely of large domains of the structure shown in Fig. 3(c) together with domains of its two symmetry-equivalent orientations.

It is evident therefore that these two special points, $(h/3, k/3)$ and $(0, k/2)$, on the diffuse locus correspond to the two types of ordered superlattice and the presence of the continuous locus of diffuse scattering between them indicates an intimate mixing of the two

Fig. 2. Small representative portions of the optical diffraction masks used to represent the ordering of metal atoms and vacancies in transition-metal niobium sulfide intercalates. The actual masks used to record the diffraction patterns shown in Fig. 1 contained $\sim 6 \times 10^4$ lattice sites. (a), (b) and (c) contained concentration modulations with wave vectors uniformly distributed around the hexagon locus; additional modulations were included at the corners of the hexagon in (d), and in the middle of the edges in (e). The values of the parameter, $\sigma_r$, defining the coherence range in units of the real-space cell repeat distance were: (a) 2; (b), (d), (e) 8; (c) 32.
which necessitates the appearance of other cluster configurations. It is interesting to consider one of these, namely configuration 9, which has a relatively high occurrence frequency in all the realizations shown. Fig. 3(d) shows the superlattice which can be constructed entirely of configuration 9 (and its reverse) clusters. Such a superlattice does not produce superlattice spots on the diffuse locus itself, but a number of superlattice reflections appear close by, as indicated schematically in Fig. 4.

4. Example involving displacement disorder

There has been much interest in recent years in charge-density waves and their associated diffraction patterns, a notable example compound which displays such effects being 1T-tantalum disulfide. This compound has a layer structure in which a layer of tantalum atoms is sandwiched between a double layer of sulfur atoms. The bonding within each sandwich is strongly covalent, with relatively weak van der Waals bonding between. Each tantalum atom has one electron left over from its contribution to the covalent bonding with the octahedrally coordinating sulfur atoms, and these remaining electrons form an essentially two-dimensional metal with a free Fermi surface (e.g. Inglesfield, 1980). The main interest in the compound is that the free Fermi surface causes instabilities, as a result of which phases appear in which incommensurate periodic lattice distortions (PLD) accompanied by charge-density waves (CDW) can exist. In the 1T₁ high-temperature phase of this compound, electron diffraction patterns show a complicated pattern of diffuse spots and rings [see plates in Van Landuyt, Van Tendeloo & Amelinckx (1974); Wilson et al. (1975); Scruby, Williams & Parry (1975)]. In Fig. 5(a) we show for reference an electron diffraction pattern of (3%) vanadium-doped tantalum disulfide which displays the same characteristic scattering pattern as the high-temperature phase of the pure TaS₂. In TaS₂ itself, as the temperature is lowered there is first a transformation to an incommensurate phase (1T₂) displaying sharp satellite peaks, followed later by a rotation of the incommensurate wave vector to 'lock-in' at a commensurate '3-by-1' superlattice vector (phase 1T₃) (Wilson et al. 1975). That the origin of the diffuse scattering in Fig. 5(a) is largely displacive in origin is indicated by the variation of intensity of various features in the pattern with azimuthal angle, producing the characteristic 'bicycle-chain' effect.

Previous workers have drawn attention to two seemingly different features of the pattern – an inner circle of radius \( \sim 0.28 a^* \) on which the six strong diffuse spots are present, and a second more continuously diffuse outer circle of radius \( \sim 0.72 a^* \). It seems likely that emphasis on the inner circle (to

![Fig. 3. Different configurations on the basic seven-point cluster of the triangular lattice, and examples of how combinations of these can form superlattice structures. (a) The 13 symmetry-unrelated configurations. (N.B. a further set of 13 complementary configurations may be obtained by interchanging the dark and light circles. For structures in which the fraction of dark atoms is 50% the frequencies of the two sets are the same.) (b) Superlattice constructed using only configurations 1 and 13. (c) Superlattice constructed using only 10 and its inverse. (d) Superlattice constructed using 9 and its inverse.](image-url)
which the eye is naturally drawn by the presence of the strong diffuse spots) has been influenced by the desire to describe the phenomena in terms of wave vectors in the first Brillouin zone (Scruby et al., 1975). Close scrutiny of the pattern, however, indicates that all of the scattering may be explained in terms of variations of intensity around the larger circles only, and it seems to us more natural to describe the phenomena purely in terms of wave vectors lying on this outer circle. When this is done a particularly simple interpretation is possible.

To model this structure using the same general method outlined in §2 we took the following steps. First we assumed a simple triangular lattice of points to represent the tantalum atoms, and used two sets of continuous variables $X_{ij}$ and $Y_{ij}$ to represent displacements of these along Cartesian axial directions. The lattice was then perturbed by a large number of waves with wave vectors distributed around the circle of radius $0.72a^*$. This was achieved by allowing the number density, $n_w$, of wave vectors to be given by a combination of a Gaussian distribution (to model the diffuse spots) and a uniform distribution (to model the remainder of the circle). No attempt was made to model the observed intensity distribution accurately (dynamical scattering in the electron diffraction makes actual intensities unreliable anyway) — only a pattern displaying the correct basic general features was intended. Similarly, no attempt was made to model accurately the radial width of the diffuse locus, a value of $\sigma_p$ being chosen to give approximately the same degree of diffuseness observed in the electron diffraction pattern. To achieve an azimuthal variation of intensity, giving rise to the 'bicycle-chain' effect, it was found necessary to assume that all waves were predominantly longitudinal. It should be noted that this particularly simple description is not possible if the usual convention of folding each wave vector back into the first Brillouin zone is adopted, since then the waves must be a mixture of both transverse and longitudinal character.

Finally, the synthesized lattice was printed using the Photomation system as described in §2, with the normalized variables $X_{ij}$ and $Y_{ij}$ used to describe the displacements of points from the underlying regular triangular lattice. For the purposes of obtaining the optical diffraction pattern shown in Fig. 5(b) only small displacements were used ($\sim 3\%$ of $a_0$ — this value being dictated by the finite resolution of the Photomation), but since such displacements are barely visible in the realization, for illustration purposes we also generated a realization with displacements enlarged by a factor of $\sim 3$. A small representative portion of this realization is shown in Fig. 5(c), and in Fig. 5(d) we show at a lower magnification a larger portion to reveal the rather broader textural features not evident in Fig. 5(c).

As for the binary example, the actual realization available as a result of the synthesis may be used for further study. We have not attempted to explore all possible ways in which this information might be used but here give one simple example. Since we have normalized the single-site displacement variables $X_{ij}$ and $Y_{ij}$ to have zero mean and unit variance, as described in §2, it is interesting to investigate the way in which variables on neighbouring sites vary with each other. To make a measure of this we compute the averages:

$$\langle X_1 X_2 \rangle; \langle X_1 Y_2 \rangle; \langle Y_1 X_2 \rangle; \langle Y_1 Y_2 \rangle,$$

where the subscripts refer to two neighbouring lattice sites. These quantities form the elements of a $2 \times 2$ covariance matrix and in general the actual values of its elements will depend on the direction of the separation vector between the two sites in relation to the Cartesian axes used to define the directions of $X_{ij}$ and $Y_{ij}$. Computation of the eigenvalues and eigenvectors of these matrices, however, reveals the displacement directions for each pair of sites that are maximally or minimally correlated. In this way it was found for the example shown in Fig. 5 that the eigenvector corresponding to the maximum correlation was in a direction almost parallel (within $\sim 2^\circ$) to the intersite vector and the correlation value was $+0.335(4)$. This evidently reflects the longitudinal character of the displacement modulations. The second eigenvalue, corresponding to displacements approximately normal to the separation vector (i.e.

Fig. 4. Schematic representation of the satellite peaks which occur as a result of the superlattices depicted in Fig. 3. Large black circles represent the main Bragg reflections; small open circles represent peaks due to the superlattice depicted in Fig. 3(b); small crosses represent the peaks due to the superlattice depicted in Fig. 3(c) (and its symmetry equivalents); small dots represent the peaks due to the superlattice depicted in Fig. 3(d) (and its symmetry equivalents). Note the coincidence with or the proximity of all of these satellite peaks to the diffuse locus.
transverse) was \(-0.027(4)\). The errors in these figures (in parentheses) were estimated by comparing the results from different realizations.

In the example illustrated in Fig. 5, one-third of the modulating wave vectors were distributed uniformly around the diffuse circle and two-thirds were distributed using a Gaussian form about the positions of the diffuse spots. For comparison we also computed lattice averages for a realization in which the proportions of the wave vectors distributed in these two ways were changed to one-quarter and three-quarters respectively. In this case it was found that the eigenvectors were virtually unchanged but the longitudinal and transverse correlations were \(+0.412\) and \(-0.067\) respectively.

![Figures](a), (b), (c), (d)

**Fig. 5.** (a) Electron diffraction pattern of vanadium-doped (3%) tantalum disulfide viewed approximately down [0001] showing characteristic diffuse scattering in the form of circles and spots. (b) Optical diffraction pattern of a lattice realization produced to model the features evident in (a). (c) A small representative portion of the lattice realization. For this illustration the displacements from the mean lattice positions have been multiplied by a factor of \(~3\) compared to those used in the optical diffraction mask from which (b) was produced. (d) A lower magnification of a larger area of the same realization as (c) showing the textural features of the distribution.

5. Discussion

What has been described in this paper is a versatile method of synthesizing a real-space distribution of scattering points which will give rise to virtually any required diffraction pattern. We have not been concerned at the present stage with situations where further symmetry information must be utilized – i.e. where modulating waves must conform to some particular symmetry representation of a complex arrangement of atoms within a unit cell. Such a development would appear to be a feasible extension of the present methods but has not so far been attempted.

While it is evident from the present work that the method described enables us to construct a real-space
distribution having a diffraction pattern very similar to a real observed pattern, it is by no means clear that this distribution is unique (in a statistical sense). Further work is in progress to try to establish this point, but at the present time it is worth recalling two features of the present method which may be pertinent.

(i) For wave vectors which are incommensurate with the basic lattice, the assumption of random phases appears to be a reasonable one, since even closely spaced (in reciprocal space) wave vectors can have no meaningful relative phase that can be maintained over any distance in real space. If, however, there are significant numbers of waves with wave vectors that have 'locked-in' to positions corresponding to a real-space superlattice, the relative phases may be important. Even in this last case it was interesting that in the binary example described in § 3, inclusion of only such waves with the special wave vectors \((h/3, k/3)\) or \((0, k/2)\ etc.\) led unambiguously to the corresponding superlattice structures.

(ii) The relationships that result between the \(X_{ij}\) variables in real space come about as a result of the direct and independent Fourier transform of elements of the reciprocal-space distribution. Consequently, since Fourier transformation by definition only involves two-particle information, the real-space distribution generated in this way can contain no higher-order correlation information such as three-particle and four-particle correlations. [For definitions of such higher-order correlation parameters see e.g. Cowley (1968)]. In previous papers (e.g. Welberry, 1977a,b, 1985) we have described a number of lattice distributions which, while giving indistinguishable diffraction patterns, nevertheless possess quite different multi-point properties. It might therefore be anticipated that distributions could exist which would give identical diffraction patterns to those presented here, and which differed in a similar way in their higher-order correlation properties. If this is the case, and this is the only way in which the present distributions lack uniqueness, then it could be said that the realizations produced here represent the simplest distributions consistent with the observed diffraction patterns.

Even with the proviso that the generated realizations may not be unique, it is hoped that having available a plausible real-space distribution will be of benefit to the researcher who, for example, desires a visual picture of what is happening in terms of the local environment in real space, as evident changes occur in reciprocal space. Quite often statements may be found in the literature where an author has attempted to describe in very vague terms the possible structure of a disordered phase in relation to some well documented ordered phase of which it is perhaps a precursor (e.g. McMillan, 1977; Naito & Tanaka, 1982). We believe that the method described here provides the means by which such relationships may be explored in detail.

We are grateful to K. Owen for technical assistance.

References