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Average lattices and aperiodic structures

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Abstract. Statistically averaged lattices provide a common basis to understand the diffraction properties of structures displaying deviations from regular crystal structures. An average lattice is defined and examples are given in one and two dimensions along with their diffraction patterns. The absence of periodicity in reciprocal space corresponding to aperiodic structures is shown to arise out of different projected spacings that are irrationally related, when the grid points are projected along the chosen coordinate axes. It is shown that the projected length scales are important factors which determine the existence or absence of observable periodicity in the diffraction pattern more than the sequence of arrangement.

Keywords. Lattice; diffraction; incommensurate; quasicrystals.

1. Introduction

Deviations from strictly periodic and crystallographically allowed symmetries are known and they are commonly referred to as aperiodic structures. The incommensurate phases are aperiodic structures with crystallographically allowed rotational symmetries (Fujiwara 1957; Hirabayashi and Ogawa 1957; Sato and Toth 1961, 1962; McMillan 1976; de Wolff 1977; Amelinckx 1979; Cowley 1979; de Wolff *et al* 1981; Perez-Mato *et al* 1987; Quan *et al* 1987; Simmons and Heine 1987). The quasicrystals are examples of aperiodic structures with crystallographically disallowed rotational symmetries (Shechtman *et al* 1984; Steinhardt and Ostlund 1987). It is desirable to examine whether the different aperiodic structures can be brought under the same frame work in relation to their diffraction properties keeping in mind that an approach that suits diffraction properties might not be suitable for other manifestations like shape or other physical properties. The characteristic feature of incommensurate crystals is the presence of superlattice reflections in their diffraction patterns and that of quasicrystals is their unusual rotational symmetry in addition to the quasiperiodic spacings of reflections. In the present work, focus will be on the role of the numerical length scales forming the spacings in real space, in conjunction with the sequence of arrangement of these spacings in determining these characteristic features. Examples in one and two dimensions to illustrate the importance of length scales are provided.

2. Definition of an average lattice

The concept of an average lattice is best illustrated with an example. Figure 1 shows a grid constructed by two orthogonal vectors. The magnitude of the vectors along X and Y directions are 1·0 and 0·25 respectively. The lattice points are the

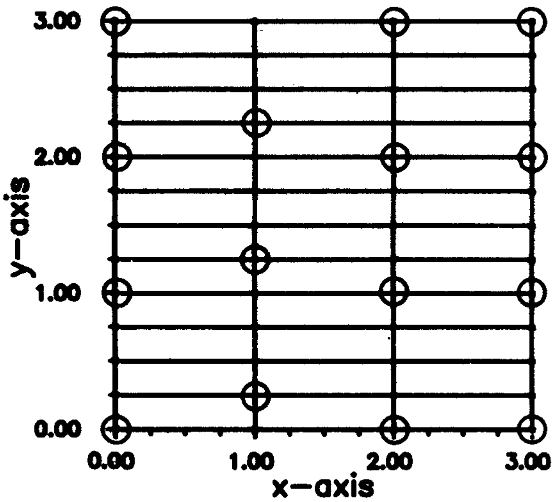

Figure 1. A grid and some scatterers. Partial occupancy of a grid leads to the definition of an average lattice.

grid intersections. The lattice points that are occupied by scatterers are marked by circles. Since the scatterers will be atoms, there exists a minimum separation between them. However, there is no such restriction on grid points. Therefore, in principle, the grid points could be arbitrarily close. However, when a chosen lattice point is occupied, it prevents any other lattice point that is within a certain radius from being occupied. This results in a situation in which the lattice contains a partial occupancy of scatterers. For the present discussion it is necessary to distinguish unoccupied lattice points from vacancies. The unoccupied lattice sites are disallowed sites. Allowed sites which remain unoccupied for other reasons are called vacancies. Note that there are several ways in which the fifteen scatterers shown in figure 1 can be redistributed over the lattice. We will soon see that the major features of a diffraction pattern are dictated by the full lattice and minor features arise out of the nature of the distribution of the scatterers.

In order to show that a distribution as shown in figure 1 is realistic, let us consider the example shown in figure 2a. This figure is a regular square lattice in two dimensions with unit lattice spacing. Suppose one applies a modulation of a type governed by the following set of equations:

$$y = \begin{cases} 0{\cdot}25 & \text{if } \cos wx \geq 0; \\ 0, & \text{otherwise.} \end{cases} \tag{1}$$

The equation implies that when the modulation wave travels along the X-axis, the scatterers get displaced along the Y-axis. Figure 2b illustrates the result of applying the modulation to figure 2a. Some scatterers are displaced by a quantity 0·25 which is the amplitude of modulation. The modulation wavelength is taken to be very large compared to the size of the figure and this leads to an aperiodic modulation within the figure. Actually figure 1 is a subset of figure 2b. Owing to the modulation, the square lattice has transformed into a dense grid with partial occupancy. One can calculate the diffraction pattern of figure 2b by computing the Patterson function assuming unit delta function scatterers at the occupied sites. The diffraction pattern for the set of scatterers in figure 2b is shown in figure 2c. It is seen that the periodicity of reflection with unit spacings along X^* direction

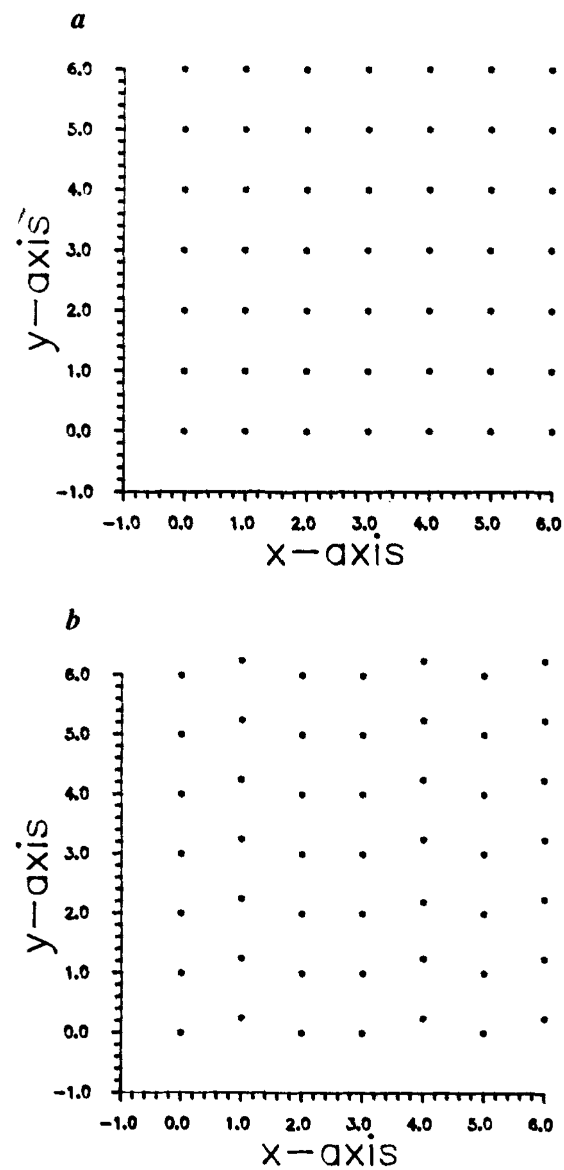

Figure 2a–b. For caption, see p. 786.

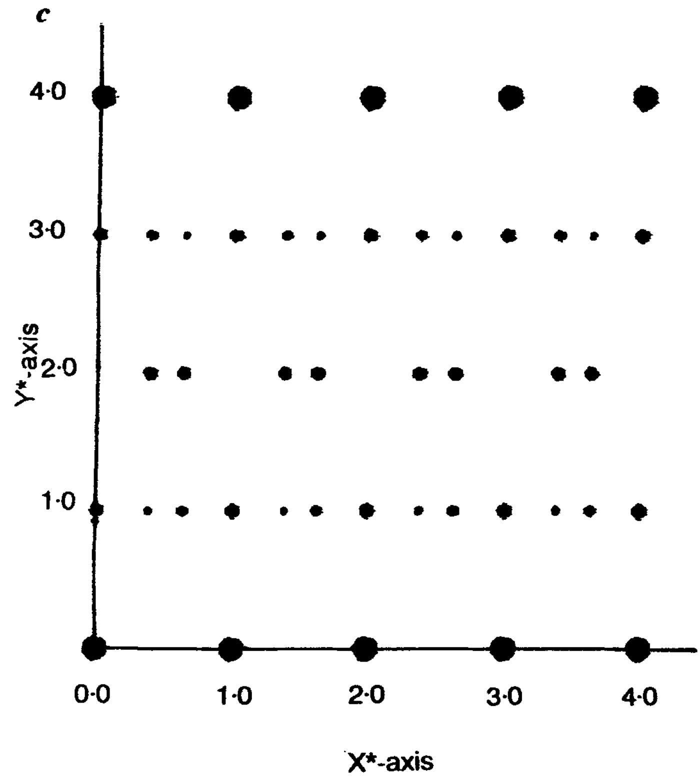

Figure 2. a. Periodic lattice, b. modulation wave along X and modulation along Y (modulation amplitude is rational) and c. diffraction pattern for the distribution of scatterers shown in b.

remains unaffected with respect to what one would obtain without modulation. However, the diffraction pattern is significantly changed along the Y^* direction. The periodicity along Y^* is now 4 units. There is a reciprocal space unit cell of 1×4. Note that it is necessary to consider both the intensity and position of the reflections to define a reciprocal space unit cell (an analogy to real space unit cell which is defined over the translation with respect to the same type of scatterer). The reciprocal space unit cell size is 1×4 owing to the real space unit cell of $1\times0{\cdot}25$ (not drawn on figure 2b). If all the vertices of the grid (spanned by $1\times0{\cdot}25$ in figure 2b) were occupied, then we would not obtain the reflections which occur within the 1×4 cell in the reciprocal space. The inner reflections within the 1×4 cell occur with a periodicity inversely proportional to the modulation wavelength. There are weak satellite reflections by the side of strong reflections which are not observable from the figure, but can be readily obtained from the Patterson.

Let us consider a more general example. Figure 3a shows the result of modulation in figure 2a, applied along X and Y directions. It is seen from the figure that there is no apparent periodicity in the distribution of scatterers. However, the diffraction pattern shown in figure 3b brings out aperiodicity. There is a clear set of reflections within a cell of 4×4 unit square which repeats itself in the reciprocal

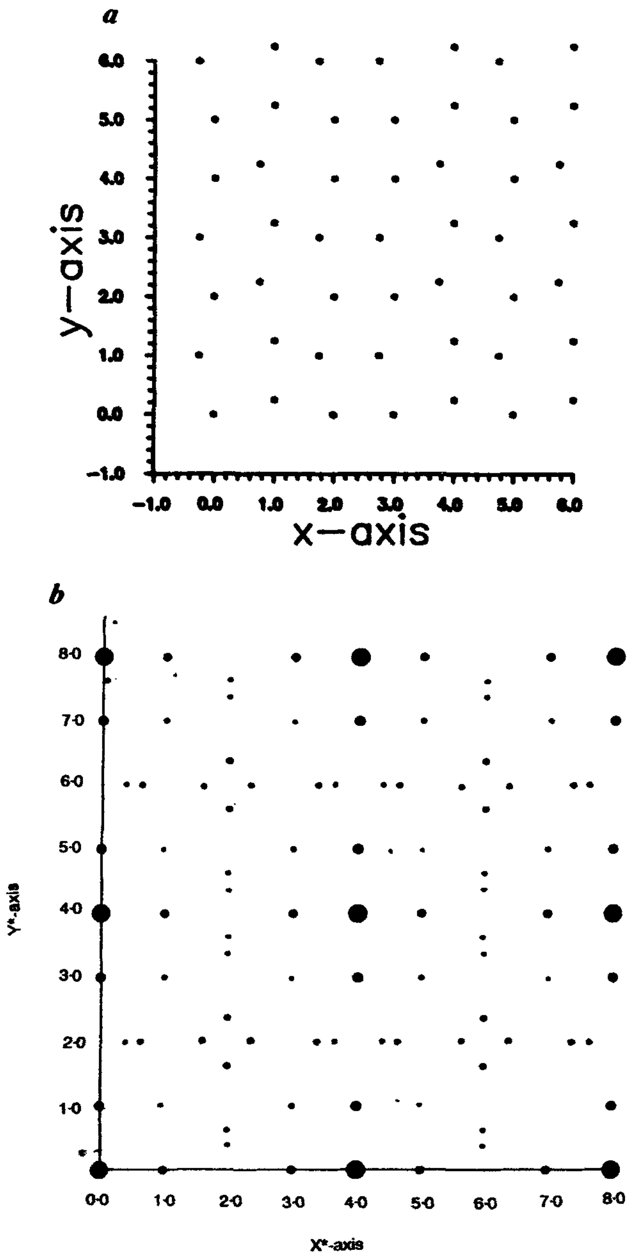

Figure 3. **a.** An aperiodic distribution of scatterers obtained by modulating a periodic distribution gives a partially occupied dense lattice and **b.** periodic diffraction from 3a.

space. This is a classic example, in which the distribution of scatterers do not appear to possess aperiodic order while its diffraction pattern is periodic. In fact, figure 3b is similar to the diffraction pattern observed for incommensurate phases. In the conventional notion of incommensurate structures, the periodic set of strong reflections are usually considered as main reflections and the weak inner reflections are considered to be superlattice reflections. The physical structure is supposed to be composed of two types of occupants, one being a periodically fixed set of scatterers (leading to the main reflections) and the other following a displacive modulation (leading to the superlattice reflections). However, we find that there is no need for a fixed set of scatterers. The distribution of scatterers in figure 3a formed a set of partially filled dense lattice whose basis vectors determined the reciprocal lattice. We are thus in a position to define an average lattice as a regular compact lattice with a distribution of disallowed sites. The term average gains importance from the fact that this distribution can be often unpredictable.

3. One dimensional average lattices

The effect of partial occupancy of a lattice can be better understood if one studies the following one dimensional example. Figure 4a shows a large unit cell structure in which there are three scatterers within a unit cell of 1 unit. For the sake of simplicity, it is assumed that these scatterers are located at 0·25, 0·5, 0·75, 1·25, 1·5 etc with positions 0,1,2 etc as unoccupied. One can readily calculate the diffraction pattern for such a distribution of scatterers by evaluating the structure factor given by

$$S(h) = \exp(-2\pi\, ih * 0{\cdot}25) + \exp(-2\pi\, ih * 0{\cdot}5) + \exp(-2\pi\, ih * 0{\cdot}75)\,. \quad (2)$$

The $S(h)$ values for h-values 0 to 4 will be 3, 1, 1, 1, 3 and these values will repeat for other integral values of h. Alternatively one can calculate the same diffraction pattern by considering the distribution of scatterers (figure 4a) as a difference of two simple lattices constructed by vectors 1 and 4 respectively (as shown in figure 4b). The structure factor of the simple lattice (a = 0·25) is given by

$$S(h') = 1 + \exp(-2\pi\, ih') + \exp(-4\pi\, ih') + \exp(-6\pi\, ih')\,. \quad (3)$$

The structure factor of the other simple lattice (a = 1) is given by

$$S(h') = 1. \quad (4)$$

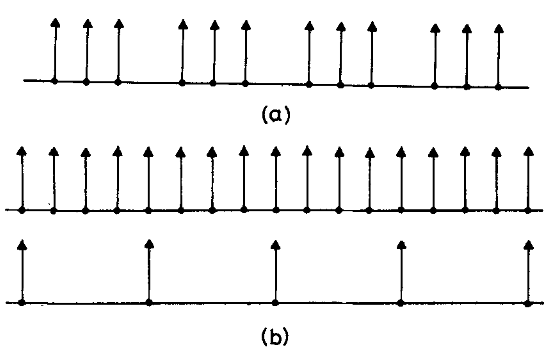

Figure 4. a. A large unit cell with 3 scatterers and **b.** the large unit cell as a difference lattice of two simple lattices with periodicities 1 and 4.

Equation (2) is obtained by subtracting (4) from (3) realizing that $h = 4\,h'$.

In this example it turned out that the unoccupied sites are periodic and therefore the resultant distribution formed a regular periodic structure. However, if the unoccupied sites are not periodic, the resultant structure forms an average lattice structure. The distribution of scatterers in figure 5a is random (with spacings 1·6 and 1). The diffraction pattern for this distribution is shown in figure 5b. It is clearly seen that the reciprocal space periodicity (RSP) of 5 units is inversely proportional to the highest common factor (0·2) between the two length scales forming the sequence. This shows that there is a one-dimensional lattice (a = 0·2) which is partially occupied and the reciprocal space unit cell has been determined by this lattice.

In order to show that the RSP is dependent on the commensuration of two spacings, another example is shown in figure 5c. The two spacings are rationally related (spacings 1·6 and 1) and the sequence of arrangement is the Fibonacci sequence. Figure 5d demonstrates the RSP of 5 units in the reciprocal space. This is again another classic example in which it can be seen that the Fibonacci sequence can lead to a periodic diffraction. Therefore the sequence of arrangement of the length scales is not a sufficient condition for an aperiodic diffraction. The ratio of length scales forming the two spacings are the important factors which determine a unit cell in reciprocal space. Let us consider the other extreme example in which the length scales are irrationally related (spacings are the golden mean τ and 1) but the sequence of arrangement is periodic. One can readily evaluate the structure factor at intervals of $1/(1+\tau)$. The intensities of expected reflections are listed in table 1. From table 1 one finds that there is no periodicity in the reciprocal space if one considers both position and intensity of reflections. The positions of strong reflections follow the Fibonacci sequence $(1, 2, 3, 5, 8)$. Owing to the irrationality between the two spacings, the actual grid has become infinitely dense and this leads to an RSP of infinity.

4. Incommensurate phases and quasicrystals

The diffraction pattern from modulated structures arise out of two factors. The first factor is associated with the amplitude of modulation of the scatterers from a mean position. The second factor is the wavelength modulating wave. The strong periodic reflections occur owing to the amplitude of modulation. The weak inner reflections occur with a periodicity inversely proportional to the wavelength of the modulating wave. The incommensurate phases have rational amplitude of modulation and a large modulation wavelength. If the modulation is periodic and travels along any axial direction then all the reflections from the modulated structure can be readily calculated. Since the modulation need not be along any axial direction, the length scales forming the sequence and the sequence itself could be aperiodic. Figure 6a shows a typical situation in two dimensions. The spacings of projected length scales are aperiodic (related by the golden mean and the sequence is Fibonacci). Figure 6b shows the calculated diffraction pattern. The positions of the reflections are not periodic and related by the golden mean. The intensities also scale up by the Fibonacci sequence. This example illustrates that a diffraction pattern could arise simply by a periodic or an aperiodic distribution of a finite number of length scales.

Regular body centred cubic structures in some alloys possess local icosahedral clusters. Let us consider a typical crystal structure in which undistorted icosahedra are sharing vertices. Every vertex in such a structure is obtainable by a linear combination of the six icosahedral vectors. From such a distribution of vertices it is possible to extract a subset of vertices, which in turn, satisfies the conditions to form a quasicrystal. This can be symbolically represented as,

$$S_{\text{actual}} = S_{\text{crystal}} \cap S_{\text{quasiperiodic}} \,. \tag{5}$$

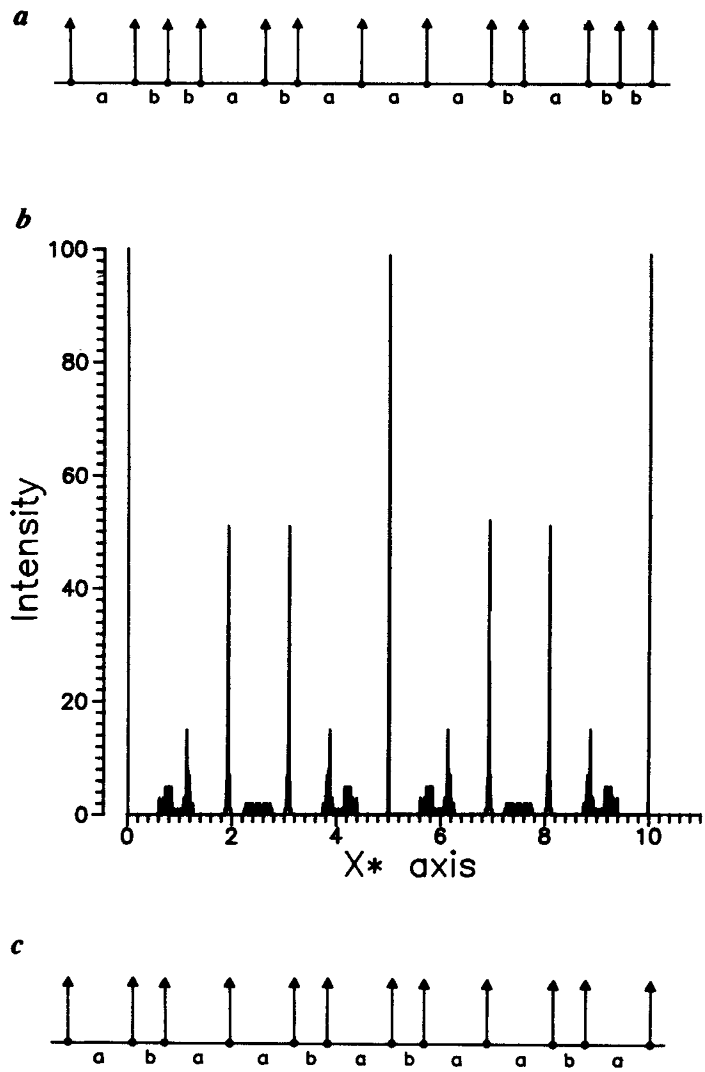

Figure 5a–c. For caption, see p. 791.

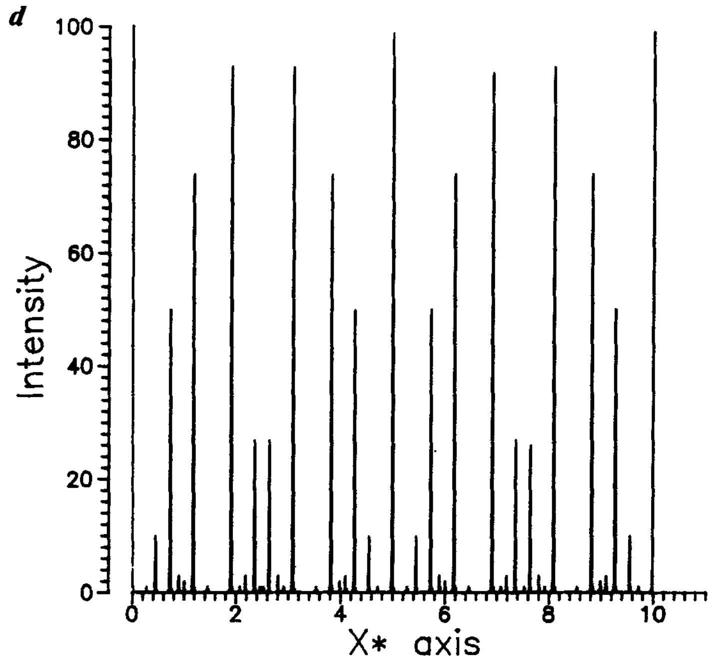

Figure 5. Two length scale average lattices in one dimension and their diffraction patterns: **a.** $a:b = 1{\cdot}6:1$ and the sequence is random, **b.** diffraction pattern that brings out the hidden periodicity in **a**, **c.** $a:b = 1{\cdot}6:1$ and the sequence is Fibonacci and **d.** a periodic diffraction pattern from **c**. Note the periodicity of reflections at intervals of 5 units in both **b** and **d**.

Table 1. The calculated diffraction pattern for a periodic distribution with two spacings 1 and τ.

Position	Intensity	h-value	Position	Intensity	h-value
0·0	100	0	7·639	17	20
0·382	13	1	8·021	99	21
0·764	54	2	8·403	8	22
1·146	80	3	8·785	60	23
1·528	0	4	9·167	74	24
1·91	92	5	9·549	2	25
2·292	37	6	9·931	95	26
2·674	26	7	10·313	30	27
3·056	96	8	10·695	33	28
3·438	3	9	11·077	94	29
3·82	71	10	11·459	1	30
4·202	64	11	11·841	77	31
4·584	6	12	12·223	58	32
4·966	98	13	12·605	10	33
5·348	21	14	12·987	99	34
5·729	43	15	13·369	16	35
6·111	88	16	13·751	50	36
6·493	0	17	14·133	83	37
6·875	85	18	14·515	0	38
7·257	47	19	14·897	89	39

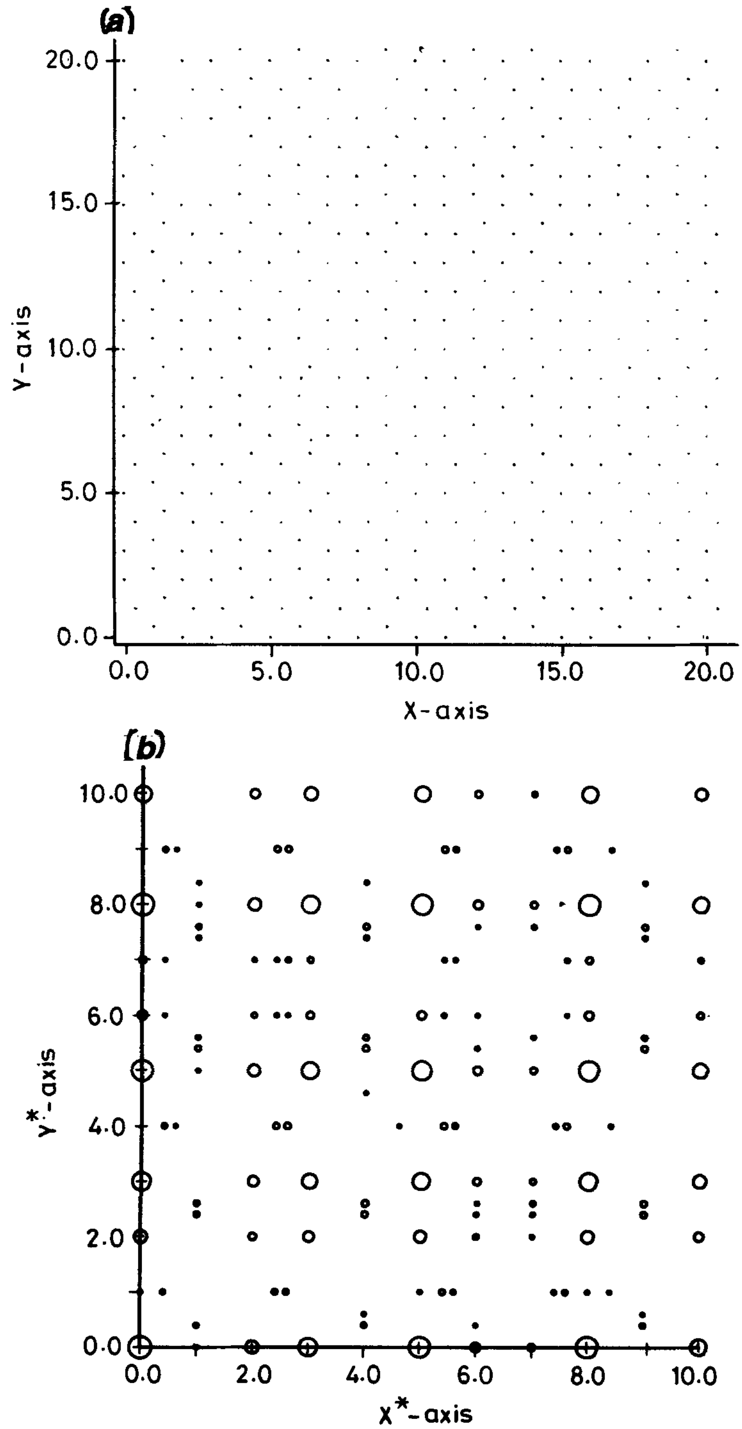

Figure 6. **a.** Two dimensional distribution of scatterers with irrational spacings and aperiodic modulation and **b.** diffraction pattern for the distribution in **a**. Observe that the spacings and the intensities follow the Fibonacci sequence.

The intersection of two sets, one being a collection of vertices in a crystal structure and the other forming a quasiperiodic set, may not be a dense space filling set. Nevertheless, it is seen that one does not require displacement of scatterers from a normal crystal structure to form a quasicrystalline distribution. Accordingly a quasicrystal can result as a special collection of incomplete icosahedra from its parent crystal structure without displacing scatterers. Since the icosahedra could also be sharing faces (instead of sharing edges) in the parent crystal structure, a quasicrystalline structure extracted from it will be different. In this context, it is important to point out that a regular simple cubic structure with two icosahedra per unit cell (sharing three vertices along the ⟨111⟩ direction) can provide a better starting model for extracting a quasicrystalline type structure. This will be discussed in due course.

5. Conclusions

We have presented a different perspective to the problem of aperiodicity in materials, mainly with their diffraction pattern in consideration. Modulations lead to the observed diffraction pattern for incommensurate phases when the modulation amplitude has a rational intercept. Partial occupancy of a crystal structure with local icosahedral symmetry leads to structures analogous to quasicrystalline structures. Since the disallowed sites form a quasiperiodic distribution they are directly related to the vacancy ordered phases (Chattopadhyay *et al* 1987). However, the vacancies are treated as disallowed sites in the present context. This is necessary to obtain the results for the incommensurate phases. The addition of phases in Fourier transform required for diffraction occurs owing to the relationship between different length scales in the distribution of scatterers. The examples considered here may be useful for a careful evaluation of structures that appear to be quasicrystalline (Baranidharan 1993).

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