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Crystallography of Quasi-Crystals

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Abstract

The symmetry of quasi-crystals, a class of materials that has recently aroused interest, is discussed. It is shown that a quasi-crystal is a special case of an incommensurate crystal phase and that it can be described by a space group in more than three dimensions. A number of relevant three-dimensional quasi-crystals is discussed, in particular dihedral and icosahedral structures. The symmetry considerations are also applied to the two-dimensional Penrose patterns.

1. Introduction

Recently an Al-Mn alloy was found (Shechtman, Blech, Gratias & Cahn, 1984) that shows a number of uncommon properties. It has sharp peaks in its diffraction pattern, indicating long-range ordering. Its point-group symmetry, however, is not one of the crystallographic ones but contains fivefold axes, a fact that is incompatible with a periodic lattice. The phenomenon was considered so remarkable that its discovery reached the newspapers and it was stated that a new state of matter had been discovered. Fivefold axes had already been found in computer simulations of alloys (Steinhardt, Nelson & Ronchetti, 1983). Perhaps that is not so surprising, because fivefold symmetry was reported much earlier for packings in space. Coxeter refers in his beautiful book *Introduction to Geometry* (Coxeter, 1961, 1980) to an experiment carried out in 1727 by Stephen Hales who studied the form of peas pressed together in a box

and observed the appearance of regular dodecahedra. A similar experiment with lead spheres was performed by Marvin in 1939 (see also Matzke, 1950).

The diffraction pattern is certainly new, but can be considered as a special case of a larger and already intensively studied class of materials: incommensurate crystal phases. Such a phase is characterized by the fact that its diffraction spots are sharp but need for their labelling more than the usual three integer indices. The five points of a regular pentagon are rationally dependent (their sum is zero), but four of them are rationally independent. Therefore, one needs at least four integers for the indices of a pattern with fivefold symmetry. The difference with the crystal phases observed until now is that there is no lattice of main reflections, such as present in, for example, incommensurately modulated crystals. For incommensurate crystal (IC) phases it has been shown (de Wolff, 1977; Janner & Janssen, 1977; de Wolff, Janssen & Janner, 1981) that the symmetry group is a group of transformations in a space with more than three dimensions. Here we shall address the questions how to describe the symmetry in the more general case and to study the possible structures.

In their study of the liquid-solid phase transition, Alexander & McTague (1978) showed in the framework of Landau theory that under certain conditions b.c.c. crystal structures are favoured. In the same paper they pointed out the possibility of a transition to a structure for which the wave vectors are points of a regular icosahedron and which has, consequently, no space-group symmetry. After the

discovery by Shechtman *et al.* (1984), Landau theory was again used to study the latter possibility. It was shown (Bak, 1985*a, b*; Mermin & Troian, 1985; Jaric, 1985) that higher-order terms can be responsible for the stability of such an 'icosahedral' structure.

In the context of the study of the new materials the term quasi-crystal has been coined (Levine & Steinhardt, 1984). The definition of this concept, however, seems to me too restrictive and does not account for the relation with IC phases. It should be noted that also in the domain of IC phases the terminology is not fully satisfactory. A structure is called incommensurate if the points in its Fourier transform contain at least four rationally independent vectors. In other words, when each vector of the Fourier transform is of the form

$$\mathbf{k} = \sum_{i=1}^n z_i \mathbf{a}_i^*, \quad z_i \text{ integers, } n > 3, \quad (1.1)$$

whereas there are no integers m_1, \dots, m_n such that $\sum m_i \mathbf{a}_i^* = 0$. If $n=3$ one has a periodic structure. When the wave vector of an IC phase changes (for example with temperature) in a smooth way it passes an infinite number of rational values, for which the structure becomes periodic. The properties of the crystal will also change in a smooth fashion, such that it is reasonable to consider the dense set of commensurate phases on the same footing as the IC phases and to consider also a higher-dimensional space group as its symmetry group. This fact is in contradiction to the term 'incommensurate phase'. It would be better to have a term covering both incommensurate and commensurate phases.

If $n=3$ in (1.1) the structure is periodic. If $n > 3$ it is no longer periodic but quasi-periodic. A quasi-periodic function is obtained as follows. Consider a function $f(x_1, \dots, x_n)$ which is periodic in each of its n variables: $f(x_i + 1, \dots, x_n) = f(x_1, \dots, x_n)$ *etc.* Take n rationally independent numbers $\alpha_1, \dots, \alpha_n$. Then the function $g(x) \equiv f(\alpha_1 x, \dots, \alpha_n x)$ is called quasi-periodic. Because its Fourier spectrum consists of integral linear combinations of $2\pi/\alpha_1, \dots, 2\pi/\alpha_n$ it may also be called a one-dimensional incommensurate structure. The function with wave vectors of the form (1.1) is clearly a generalization of such a quasi-periodic function. It has the property that for each positive number ε there is a translation such that the transformed function differs from the original one by less than ε , which explains the term 'quasi-periodic'. IC phases are, therefore, also quasi-periodic.

The new class of structures obviously satisfies the requirements for an IC structure. The difference is that modulated and composite structures show a lattice of main reflections. In systems with noncrystallographic symmetry operations this is impossible. We shall call the systems in this class quasi-crystals. In the following we show that the symmetry consider-

ations for modulated IC phases are readily generalized to quasi-crystals.

The organization of the paper is as follows. In § 2 the method to describe the symmetry of modulated crystals is briefly recalled. In § 3 a two-dimensional quasi-crystal is discussed to exemplify the changes that are needed in the formalism. In § 4 this same structure is applied to an interesting covering in the plane, the Penrose tiling. In § 4 a way to describe quasi-crystals is formulated. In § 5 the most important three-dimensional quasi-crystal structures and their symmetries are discussed.

2. IC phases with a lattice of main reflections

For most IC phases studied so far a conspicuous set of main reflections stands out among the diffraction spots and this set belongs to a three-dimensional lattice, the lattice of main reflections. The spots that do not belong to this set are the satellites. Every spot of the diffraction pattern corresponds to a vector \mathbf{k} that can be written in this case as

$$\mathbf{k} = \sum_{i=1}^3 z_i \mathbf{a}_i^* + \sum_{j=1}^d z_{3+j} \mathbf{a}_{3+j}^*, \quad (2.1)$$

where $\mathbf{a}_1^*, \mathbf{a}_2^*, \mathbf{a}_3^*$ form a basis of the lattice of main reflections. The whole collection of all vectors of the form (2.1) forms a set, which we shall denote in the sequel by M^* , that is clearly not a three-dimensional lattice, but that can be considered as the projection on the three-dimensional space of a lattice in $3+d$ dimensions. This is the observation behind the introduction of a higher-dimensional symmetry group. The problem is to find the components in the additional space and to define what is meant by 'distance' in that space. In the following we describe the construction of a basis in the higher-dimensional space [which will be given in (2.5) and (2.7)]. It is based on some simple theorems from group theory.

The relations between the additional components of the wave vectors can be found as follows (Janner & Janssen, 1979). Consider the point-symmetry group of the diffraction pattern. If R is an element of this group K it transforms main reflections to main reflections and satellites to satellites. The action on the vectors $\mathbf{a}_1^*, \dots, \mathbf{a}_{3+d}^*$ is given by

$$R^{-1} \mathbf{a}_i^* = \sum_{k=1}^3 \Gamma_E(R)_{ik} \mathbf{a}_k^*, \quad i=1, 2, 3 \quad (2.2)$$

$$R^{-1} \mathbf{a}_{3+j}^* = \sum_{l=1}^3 \Gamma_M(R)_{jl} \mathbf{a}_l^* + \sum_{m=1}^d \Gamma_I(R)_{jm} \mathbf{a}_{3+m}^*, \quad j=1, \dots, d. \quad (2.3)$$

Therefore, to each element R corresponds a $(3+d)$ -dimensional integral matrix $\Gamma(R)$

$$\Gamma(R) = \begin{pmatrix} \Gamma_E(R) & 0 \\ \Gamma_M(R) & \Gamma_I(R) \end{pmatrix}. \quad (2.4)$$

These matrices form an integral reducible representation of the group K . According to well known theorems in group theory there is a basis in $3+d$ dimensions such that the representation becomes orthogonal and the direct sum of two representations. In other words, there are pairs (R, R_I) of orthogonal transformations, in three and d dimensions respectively, corresponding to each matrix $\Gamma(R)$. The three-dimensional space is the ordinary space, denoted by V_E , and the d -dimensional space is the orthogonal complement, denoted by V_I . In this way the scalar products of vectors in the internal space and the metrical relations there are determined. They are, however, not completely determined. If the representation $\Gamma_I(K)$ has p irreducible components, there are still p free common factors, in each subspace one. (It is clear that nothing happens to the construction if one multiplies all components in V_I by a common factor c .) Apart from this freedom the basis on which the orthogonal transformation R_I has the form $\Gamma_I(R)$ is fixed. We denote this basis of V_I by $\mathbf{b}_{1^*}, \dots, \mathbf{b}_{d^*}$. The basis in $(3+d)$ -dimensional space is then

$$\begin{aligned} \mathbf{d}_i^* &= (\mathbf{a}_i^*, 0), & i &= 1, 2, 3; \\ \mathbf{d}_{3+j}^* &= (\mathbf{a}_{3+j}^*, \mathbf{b}_j^*), & j &= 1, \dots, d. \end{aligned} \quad (2.5)$$

The set M^* is just the projection of the lattice Σ^* spanned by these vectors.

As worked out in detail by Janner & Janssen (1977), one constructs from a three-dimensional quasi-periodic function with Fourier spectrum spanning M^* another function f_s in $3+d$ dimensions as

$$f_s(r, r_I) = \sum \hat{f}(\mathbf{k}) \exp [i(\mathbf{k}r + \mathbf{k}_I r_I)], \quad (2.6)$$

where $\hat{f}(\mathbf{k})$ is the Fourier component of f with vector \mathbf{k} , and $(\mathbf{k}, \mathbf{k}_I)$ is the unique vector of Σ^* that is projected on \mathbf{k} in M^* . Then one can show that f_s is a periodic function with the periodicity of the lattice Σ that is the dual to Σ^* and has basis

$$\begin{aligned} \mathbf{d}_i &= (\mathbf{a}_i, -\Delta \mathbf{a}_i), & i &= 1, 2, 3; \\ \mathbf{d}_{3+j} &= (0, \mathbf{b}_j), & j &= 1, \dots, d, \end{aligned} \quad (2.7)$$

where $\mathbf{b}_1, \dots, \mathbf{b}_d$ is the basis in V_I dual to $\mathbf{b}_1^*, \dots, \mathbf{b}_d^*$ and $\Delta \mathbf{a}_i$ is fixed by the requirement $\mathbf{d}_i \cdot \mathbf{d}_j^* = \delta_{ij}$. Consequently, f_s is invariant under a space group in $3+d$ dimensions with lattice Σ . This space group is called the symmetry group of the IC phase and accounts for selection rules, systematic extinctions *etc.*, just as a three-dimensional space group does for a periodic structure. For quasi-crystals one may generalize this procedure and construct also a corresponding periodic structure in more dimensions.

3. The symmetry of a two-dimensional quasi-crystal

To study the procedure for quasi-crystals we first consider a two-dimensional example. Suppose the

diffraction pattern has fivefold symmetry and all vectors of the pattern are integral linear combinations of the five vectors pointing to the corners of a regular pentagon:

$$\mathbf{e}_n = a(\cos 2\pi n/5, \sin 2\pi n/5). \quad (3.1)$$

Because the sum of these vectors is zero each vector can be expressed as

$$\mathbf{k} = \sum_{i=1}^4 z_i \mathbf{a}_i^*, \quad \mathbf{a}_i^* = \mathbf{e}_i. \quad (3.2)$$

The set M^* of all these vectors is invariant under a fivefold rotation α , a mirror β and the central inversion γ . The action of these elements on the four basis vectors is given by

$$\begin{aligned} \Gamma(\alpha) &= \begin{pmatrix} 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \\ -1 & -1 & -1 & -1 \end{pmatrix}, \\ \Gamma(\beta) &= \begin{pmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix}, \\ \Gamma(\gamma) &= \begin{pmatrix} -1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}. \end{aligned} \quad (3.3)$$

These matrices generate a group isomorphic to $D_5 \times C_2$, the direct product of the non-crystallographic dihedral group D_5 and the group consisting of unity and central inversion. The four-dimensional representation is reducible. The group $D_5 \times C_2$ has four *gerade* and four *ungerade* representations. (The four representations of D_5 in Table 1 can be combined with either +1 or -1 for the central inversion.) The representation $\Gamma(K)$ is the sum of the two *ungerade* two-dimensional representations. The representation carried by the ordinary space, which is an invariant subspace, is the *ungerade* representation stemming from Γ_3 as one sees from the character of the 72° rotation α which is $2 \cos(2\pi/5) = \tau$. The 72° rotation in V_E forms a pair with a 144° rotation in V_I , because the other two-dimensional irreducible component of $\Gamma(\alpha)$ has character $-1 - \tau$, which is $2 \cos(4\pi/5)$.

The basis $\mathbf{a}_1^*, \dots, \mathbf{a}_4^*$ of M^* is the projection of a basis $\mathbf{d}_1^*, \dots, \mathbf{d}_4^*$ of a four-dimensional lattice Σ^* . From the decomposition of the representation $\Gamma(K)$ one knows the pairs (R, R_I) . The basis of Σ^* has to satisfy the requirement that these pairs are represented on this basis by the matrices $\Gamma(R)$. This fixes the basis up to a common scale factor c :

$$\mathbf{d}_i^* = (c\mathbf{e}_i, c\mathbf{e}_{2i}), \quad i = 1, \dots, 4. \quad (3.4)$$

Table 1. Character tables for some dihedral and the icosahedral groups

$D_{5s}, \tau = (\sqrt{5}-1)/2$				
Elements	ε	α	α^2	β
Γ_1	1	1	1	1
Γ_2	1	1	1	-1
Γ_3	2	τ	$-1-\tau$	0
Γ_4	2	$-1-\tau$	τ	0

D_{8s}							
Elements	ε	α	α^2	α^3	α^4	β	$\alpha\beta$
Γ_1	1	1	1	1	1	1	1
Γ_2	1	1	1	1	1	-1	-1
Γ_3	1	-1	1	-1	1	1	-1
Γ_4	1	-1	1	-1	1	-1	1
Γ_5	2	$\sqrt{2}$	0	$-\sqrt{2}$	-2	0	0
Γ_6	2	0	-2	0	2	0	0
Γ_7	2	$-\sqrt{2}$	0	$\sqrt{2}$	-2	0	0

$D_{10s}, \tau = (\sqrt{5}-1)/2$								
Elements	ε	α	α^2	α^3	α^4	α^5	β	$\alpha\beta$
Γ_1	1	1	1	1	1	1	1	1
Γ_2	1	1	1	1	1	1	-1	-1
Γ_3	1	-1	1	-1	1	-1	1	-1
Γ_4	1	-1	1	-1	1	-1	-1	1
Γ_5	2	$1+\tau$	τ	$-\tau$	$-1-\tau$	-2	0	0
Γ_6	2	τ	$-1-\tau$	$-1-\tau$	τ	2	0	0
Γ_7	2	$-\tau$	$-1-\tau$	$1+\tau$	τ	-2	0	0
Γ_8	2	$-1-\tau$	τ	τ	$-1-\tau$	2	0	0

D_{12s}									
Elements	ε	α	α^2	α^3	α^4	α^5	α^6	β	$\alpha\beta$
Γ_1	1	1	1	1	1	1	1	1	1
Γ_2	1	1	1	1	1	1	1	-1	-1
Γ_3	1	-1	1	-1	1	-1	1	1	-1
Γ_4	1	-1	1	-1	1	-1	1	-1	1
Γ_5	2	$\sqrt{3}$	1	0	-1	$-\sqrt{3}$	-2	0	0
Γ_6	2	1	-1	-2	-1	1	2	0	0
Γ_7	2	0	-2	0	1	0	-2	0	0
Γ_8	2	-1	-1	2	-1	-1	2	0	0
Γ_9	2	$-\sqrt{3}$	1	0	-1	$\sqrt{3}$	-2	0	0

$I, \tau = (\sqrt{5}-1)/2$					
Elements	ε	α	α^2	β	$\alpha\beta$
Order	1	5	5	3	2
Γ_1	1	1	1	1	1
Γ_2	3	$-\tau$	$1+\tau$	0	-1
Γ_3	3	$1+\tau$	$-\tau$	0	-1
Γ_4	4	-1	-1	1	0
Γ_5	5	0	0	-1	1

The lattice Σ in direct space has a basis that is dual to (3.4):

$$\mathbf{d}_i = \frac{2}{5}[\mathbf{e}_i - \mathbf{e}_0, (1/c)(\mathbf{e}_{2i} - \mathbf{e}_0)]. \quad (3.6)$$

This lattice belongs to the same Bravais class as Σ^* . There is only one Bravais class in this system and according to Brown *et al.* (1978) to this Bravais class correspond four geometric classes, five arithmetic classes and five space groups (all symmorphic).

An example of a function that is quasi-periodic and the restriction of a periodic four-dimensional function with lattice Σ is a density function

$$f(\mathbf{r}) = \sum_{n=0}^4 \cos(\mathbf{e}_n \cdot \mathbf{r}). \quad (3.7)$$

The corresponding periodic function in four dimensions is

$$f_s(\mathbf{r}, \mathbf{r}_I) = \sum_{n=0}^4 \cos(\mathbf{e}_n \cdot \mathbf{r} + c\mathbf{e}_{2n} \cdot \mathbf{r}_I). \quad (3.8)$$

The symmetry group of this periodic density function in four dimensions is the symmorphic holohedral space group of the lattice Σ . The restriction to the two-dimensional space $\mathbf{r}_I = 0$ is quasi-periodic and can be called a quasi-crystal. A picture of this function is given in Fig. 1.

4. Penrose tilings

Another example of a two-dimensional quasi-crystal with fivefold rotation axes is the so-called Penrose tiling. Penrose (1974) showed that it is possible to cover the plane in an aperiodic way by means of tiles of one of two kinds. The tiles may have the forms known as 'dart' and 'kite' (Fig. 2). Another pair of tiles consists of rhombuses with equal edges, one with

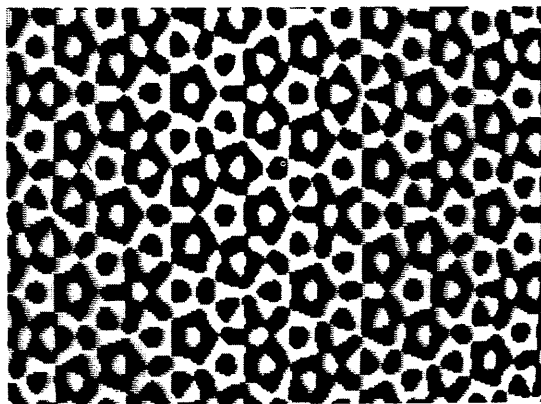


Fig. 1. Restriction of a periodic function in four dimensions to the two-dimensional plane V_E . In the dark regions the value of the function is positive, in the light regions negative. The periodic function is invariant under the holohedral space group of the decagonal lattice.

The lattice Σ^* generated by (3.4) is the decagonal lattice denoted as XIX/I by Brown, Bülow, Neubüser, Wondratschek & Zassenhaus (1978). The distance in the four-dimensional space is determined by the metric tensor g which has components $g_{ij} = \mathbf{d}_i^* \cdot \mathbf{d}_j^*$.

$$g = \begin{pmatrix} A & B & C & C \\ B & A & B & C \\ C & B & A & B \\ C & C & B & A \end{pmatrix} \quad (3.5)$$

with $A = 1 + c^2$, $B = (\tau/2) - [(1 + \tau)/2]c^2$, $C = -(A + 2B)/2$, where $\tau = (\sqrt{5} - 1)/2$ is related to the golden ratio.

angles 72 and 108°, the other with 36 and 144°. A large number of interesting properties for these tilings has been revealed (Gardner, 1977; de Bruijn, 1981). In particular, they are self-similar. In the covering by rhombuses the vertices are of the form $\sum z_n \mathbf{e}_n$ with z_n integer and \mathbf{e}_n as in (3.1). The collection of all points of this form would be dense in the plane but for a Penrose pattern not all quintuples (z_0, \dots, z_4) are allowed. Which quintuples occur may be determined using a theorem by de Bruijn (1981). The same theorem is used to find a periodic pattern in four dimensions such that the intersection with the two-dimensional plane V_E is a Penrose tiling.

De Bruijn considers the vertices of a Penrose pattern as points in the complex plane. They are of the form

$$x = \sum_{j=0}^4 z_j \zeta^{-j}, \quad (4.1)$$

where $\zeta = \exp(2\pi i/5)$. One defines a convex set in the complex plane for every value of a parameter p as

$$V_p = \left\{ \sum_{j=0}^4 \lambda_j \zeta^{2j} \mid 0 \leq \lambda_j < 1, \sum_{j=0}^4 \lambda_j = p \right\}. \quad (4.2)$$

Then the point x (4.1) belongs to a Penrose pattern, specified by five real numbers γ_j with sum 0, if and only if

$$\sum_{j=0}^4 (z_j - \gamma_j) \zeta^{2j} \text{ belongs to } V_p \text{ with } p = \sum_{j=0}^4 z_j. \quad (4.3)$$

This theorem can be used to construct a periodic structure in four dimensions which has as intersection with a two-dimensional hyperplane a Penrose pattern, and has the periodicity of the lattice Σ (3.6). One notices that $\sum z_j \zeta^{-j}$ corresponds to an element $\sum z_j \mathbf{e}_j$ in V_E , whereas $\sum (z_j - \gamma_j) \zeta^{2j}$ corresponds to a lattice point in V_I .

Consider a point S in the four-dimensional space as the origin of the lattice Σ . The point S may be chosen to have an external component zero: $S = \sum s_i \mathbf{d}_i$

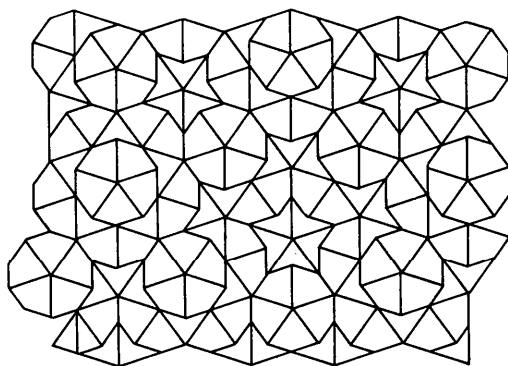


Fig. 2. Tiling of the plane by means of 'darts' and 'kites' (after Gardner, 1977).

and $S_E = 0$. The lattice has point-group symmetry $D_5 \times C_2$. In each unit cell we take five Wyckoff positions: the points $-p(\mathbf{d}_1 + \mathbf{d}_2 + \mathbf{d}_3 + \mathbf{d}_4)/5$ with $p = 0, 1, 2, \dots, 4$. The position with $p = 0$ has site symmetry $D_5 \times C_2$, the others have site symmetry D_5 . Such a point in the unit cell at (z_1, z_2, z_3, z_4) has a component r_E :

$$r_E = \sum_{i=1}^4 (z_i - p/5)(\mathbf{d}_i)_E = \left(\frac{2}{5}\right) \sum_{j=0}^4 z_j \mathbf{e}_j, \quad (4.4)$$

when z_0 is such that $p = \sum_{j=0}^4 z_j$, and a component r_I in V_I :

$$r_I = \frac{2}{5} \sum_{j=0}^4 z_j \mathbf{e}_{2j} + s_I. \quad (4.5)$$

Because s_I can be written as $\sum s_j \mathbf{e}_{2j}$ de Bruijn's theorem states that r_E belongs to a Penrose pattern if r_I is inside a convex domain V_p in V_I defined by

$$V_p = \left\{ \frac{2}{5} \sum_{j=0}^4 \lambda_j \mathbf{e}_{2j} \mid 0 \leq \lambda_j < 1, \sum_{j=0}^4 \lambda_j = p \right\}. \quad (4.6)$$

One can reformulate this condition as follows. Attach to each of the five mentioned Wyckoff positions a two-dimensional pentagon V_p parallel to V_I (Fig. 3). If such a pentagon intersects V_E , the intersection is a point (two perpendicular two-dimensional hyperplanes in a four-dimensional space intersect at a point) which has r_E (4.4) as component in V_E and belongs to the Penrose pattern.

Hence a Penrose pattern may be obtained as the intersection of ordinary space with a periodic structure with disconnected pentagons at five Wyckoff positions per unit cell of a lattice Σ . Different patterns are obtained by choosing another origin S for Σ . An example of the intersection of the given four-dimensional structure with V_E is given in Fig. 4(a). In Fig. 4(b) the intersection points are connected yielding a covering of the plane by rhombuses. The five pentagons in the unit cell are left invariant by the holohedral point group $D_5 \times C_2$ (D_5 leaves each pentagon invariant, the central inversion interchanges the pentagons with $p = 2$ and 3, and also those with $p = 1$ and 4). Therefore, the symmetry group of the Penrose pattern is the holohedral space group discussed in the preceding section.

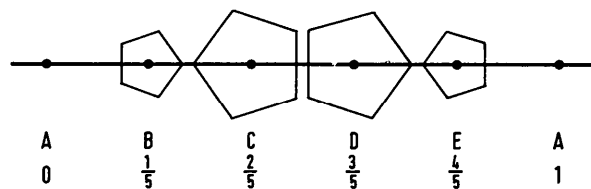


Fig. 3. The five pentagons attached to the five Wyckoff positions $(p, p, p, p)/5$ in the unit cell of the decagonal lattice. The pentagon for $p = 0$ has shrunk to a point.

The crystallographic aspects of Penrose tiling have previously been studied by Mackay (1982), who determined the diffraction pattern by optical means. Kramer (Kramer, 1982, 1985; Kramer & Neri, 1984) has given a crystallographic discussion of aperiodic tilings that is a generalization of that by de Bruijn.

From Fig. 2 one can understand why the diffraction pattern of a quasi-crystal shows sharp peaks although there is no lattice periodicity. In the von Laue approach diffraction is the result from interference between waves reflected from equidistant parallel planes. In the figure one can see five sets of parallel lines. Now there are two different distances between neighbouring lines and their ratio is $1 : \tau$.

5. Quasi-crystals

The fact that the four-dimensional structure corresponding to a Penrose pattern consists of disconnected elements is another difference with modulated and composite structures. In the latter two cases the embedding of the quasi-periodic point structure in a higher-dimensional space consists of lines ($d=1$) or d -dimensional hyperplanes without boundaries. Next to the non-crystallographic point-group symmetry or

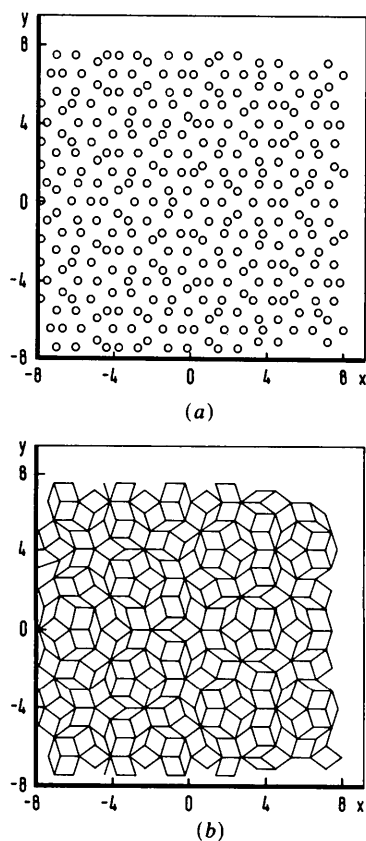


Fig. 4. (a) The intersection of a pattern of disconnected pentagons with $D_5 \times C_2$ symmetry in four dimensions with the two-dimensional plane V_E . (b) The intersection points are the vertices of a tiling of the plane by means of rhombuses.

the absence of a lattice of main reflections, the sharp diffraction spots and the incommensurate diffraction pattern this is an important characteristic of quasi-crystals.

This same phenomenon occurs in a one-dimensional structure which has also been called a quasi-crystal and which we shall call a Fibonacci chain. According to our former definition there are no one-dimensional quasi-crystals because the only two point groups here are crystallographic. Now consider a chain of particles with distance between neighbours either 1 or $\tau = (\sqrt{5}-1)/2$. There are several equivalent prescriptions for the order of the different intervals. For instance, consider the sequence given by $y_n = y_0 + n\tau$. If y_n and y_{n+1} have the same integer part $x_{n+1} = x_n + \tau$, otherwise $x_{n+1} = x_n + 1$. The points one obtains in this way are all of the form $m + n\tau$, and the average distance is $d = 3\tau - 1$. It can be shown that the points can be interpreted as a modulated chain with positions

$$x_n(t) = x_0 + nd + (\tau - 1) \left[\text{frac}(n\tau + t) - \frac{1}{2} \right]. \quad (5.1)$$

The argument t is the phase of the modulation. For $t = \frac{1}{2}$ one obtains a chain with inversion symmetry. Therefore the chain consists of short and long intervals (S and L , respectively) with length ratio $\tau : 1$. An example is the sequence

$$\dots SL SLL SLSLL SLLSLS \dots$$

From (5.1) one sees that the modulation wave vector is $2\pi\tau/d$, the modulation function is the discontinuous function $f(x) = A \text{frac}(x) + B$, and the wave vectors of the Fourier spectrum are of the form (1.1) with $n=2$ and

$$a_1^* = 2\pi/d; \quad a_2^* = 2\pi\tau/d. \quad (5.2)$$

In the standard way one can embed the Fibonacci chain in a two-dimensional space. The chain is again the intersection of V_E with a periodic structure consisting of disconnected line elements on lattice points (Fig. 5a). In contrast to the embedding of the Penrose pattern these elements are not parallel to V_E .

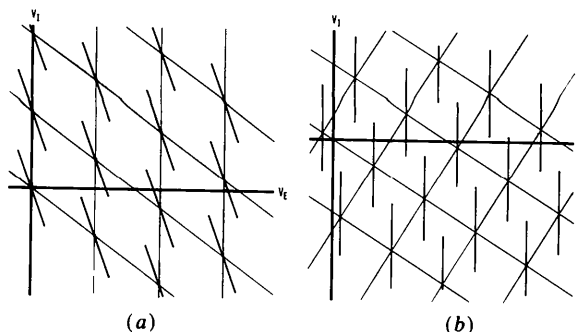


Fig. 5. (a) Embedding of the Fibonacci chain as modulated crystal. (b) An equivalent embedding in two dimensions. The Fibonacci chain is the intersection of the periodic structure consisting of line elements with the line V_E .

For a three-dimensional quasi-crystal the spots of the diffraction pattern are of the form (1.1) and there is no lattice of main reflections. Also in this case the set M^* can be seen as the projection of an n -dimensional lattice Σ^* . The procedure of embedding is a generalization of that for modulated crystals. Consider the point group K that leaves M^* invariant. Then for each element R of K one has

$$R^{-1}\mathbf{a}_i^* = \sum_{j=1}^n \Gamma(R)_{ij}\mathbf{a}_j^*. \quad (5.3)$$

The matrices $\Gamma(R)$ form an integral n -dimensional representation of K that is equivalent to an orthogonal one. Because the orthogonal representations leave the ordinary space generated by $\mathbf{a}_1^*, \dots, \mathbf{a}_n^*$ invariant, $\Gamma(K)$ is $(3+d)$ reducible. The matrices correspond to pairs (R, R_I) . The problem now is to construct a basis

$$\mathbf{d}_i^* = (\mathbf{a}_i^*, \mathbf{b}_i^*), \quad i = 1, \dots, n, \quad (5.4)$$

such that the elements (R, R_I) are represented by $\Gamma(R)$ on this basis.

In the n -dimensional space the dual basis $\mathbf{d}_1, \dots, \mathbf{d}_n$, defined by $\mathbf{d}_i \cdot \mathbf{d}_j^* = \delta_{ij}$, generates a lattice Σ . The action of this lattice on the quasi-periodic structure in V_E gives a periodic structure in n dimensions. The symmetry group of the latter is an n -dimensional space group that again is called the symmetry group of the quasi-periodic structure. The embedding is unique up to a common factor for each irreducible component of $\Gamma(K)$ in V_I .

One can apply this method also to the quasi-periodic Fibonacci chain. The generators of M^* are given by (5.2) which can be seen as the projection of the basis of a two-dimensional lattice. A solution is given by

$$\mathbf{d}_1^* = (2\pi/d)(1, a\tau), \quad \mathbf{d}_2^* = (2\pi/d)(\tau, -a), \quad (5.5)$$

for arbitrary a . Then the dual basis is

$$\begin{aligned} d_1 &= [d/2\pi(2-\tau)](1, \tau/a), \\ d_2 &= [d/2\pi(2-\tau)](\tau, -1/a). \end{aligned} \quad (5.6)$$

The Fibonacci chain is obtained as the intersection of V_E with a periodic structure consisting of line elements parallel to V_I at the lattice points of Σ , whereas the origin of the lattice is an arbitrary point in the two-dimensional space. For $a = 1$ the structure is shown in Fig. 5(b).

6. Some three-dimensional quasi-crystals

The construction in the preceding section is valid for arbitrary dimension. However, from a practical point of view there are restrictions: the description loses its meaning when the dimension n exceeds three by too much. Therefore, we shall restrict ourselves here to the case $n \leq 6$. The generators of M^* form a set that

is invariant under a finite point group. In the Landau-theory formulation the vectors of M^* all belong to the same representation of the Euclidean group $E(3)$ and are, therefore, of equal length. Then the question is: what are the possible structures in three dimensions with a Fourier spectrum that belongs to a set M^* that is invariant under a finite non-crystallographic point group and is of rank less than six (*i.e.* has six or less generators). Because the diffraction pattern is invariant under the central inversion the groups to be considered are direct products of the group consisting of identity and central inversion and a group that is one of the dihedral groups D_5, D_8, D_{10}, D_{12} or the icosahedral group I .

A collection of vectors generating a set M^* invariant under $D_{2m} \times C_2$ (with $m = 4, 5$ or 6) may lie in a plane and form then the corners of a regular $2m$ -gon. The rank of M^* in these cases is four and the corresponding four-dimensional lattice Σ^* belong to the octagonal, decagonal and dodecagonal Bravais classes, respectively, in the terminology of Brown *et al.* (1978). For $m = 5$ it is just the lattice discussed in section three. In this case the structure is essentially two dimensional.

A three-dimensional structure is obtained by adding a vector perpendicular to the $2m$ -gon. The rank of M^* is then five. Generators for the corresponding groups $\Gamma(K)$ are given in Table 2. These representations are $2+2+1$ reducible, as can be seen using the character table (Table 1).

Non-planar generating sets for M^* which are obtained by the action of the dihedral point groups on a single vector are shown in Fig. 6. The rank of M^* is also five in this case. The set invariant under

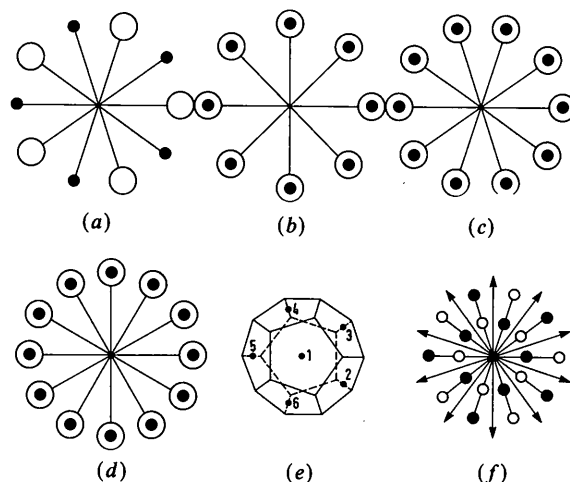


Fig. 6. Generating sets of wave vectors for a quasi-periodic structure. The sets are invariant under a finite point group. (a) $D_5 \times C_2$, (b) $D_8 \times C_2$, (c) $D_{10} \times C_2$, (d) $D_{12} \times C_2$, (e) faces of the dodecahedron, (f) edges of the dodecahedron. Vectors with negative z component are indicated by filled circles, with positive z component open circles. In (f) the arrows have z component zero.

D_5 is not invariant under D_{10} now. The set for D_{12} generates the same M^* as the regular dodecagon plus a perpendicular vector, discussed in the preceding paragraph. For D_8 and D_{10} the vectors of Fig. 6 generate a subset of the corresponding M^* discussed in the preceding paragraph: only those spots occur for which $z_1 + z_2 + z_3 + z_4 + z_5$ is even. Therefore, the corresponding lattices Σ^* are centring of those of the preceding paragraph.

So in total one obtains six Bravais classes for these four dihedral groups. The holohedral point groups $D_p \times C_2$ have subgroups C_p , $C_p \times C_2$ and D_p ($p = 5, 8, 10$ or 12) that belong to the same Bravais class. For each of the corresponding matrix groups one may determine the possible space groups (Zassenhaus, 1948; Brown, 1969; Fast & Janssen, 1971).

We shall treat the icosahedral case in more detail, because it is the case of the Al-Mn alloy. Consider first the case that the set M^* is generated by the 12 vectors that point to the faces of a regular dodecahedron. Among the 12 vectors six are rationally independent.

$$\begin{aligned} \mathbf{a}_1^* &= (0, 0, 1), \\ \mathbf{a}_n^* &= (\sin \theta \cos 2\pi n/5, \sin \theta \sin 2\pi n/5, \cos \theta), \\ n &= 2, \dots, 6. \end{aligned} \quad (6.1)$$

Here $\cos^2 \theta = \frac{1}{5}$. The action of $I \times C_2$ on this basis is given by the matrix group $\Gamma(K)$ with generators

$$\begin{aligned} \Gamma(\alpha) &= \begin{pmatrix} 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 \\ 0 & 1 & 0 & 0 & 0 & 0 \end{pmatrix}, \\ \Gamma(\beta) &= \begin{pmatrix} 0 & 0 & 0 & 0 & 0 & 1 \\ 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 \end{pmatrix} \end{aligned} \quad (6.2)$$

and the central inversion. This six-dimensional representation is reducible: the two irreducible components are the two non-equivalent *ungerade* three-dimensional irreducible representations of $I \times C_2$ (the character table of the icosahedral group I is given in Table 2). This is seen, in the standard way, from the character. For the rotation group I the character of the representation is $\chi(\varepsilon, \alpha, \alpha^2, \beta, \alpha\beta) = (6, 1, 1, 0, -2)$. This is the sum of the characters of Γ_2 and Γ_3 . The rotations in V_E correspond to the irreducible component Γ_3 because there the character is $1 + 2 \cos(\text{rotation angle})$, which is $3, 1 + \tau, -\tau, 0$ and

-1 for $\varepsilon, \alpha, \alpha^2, \beta$ and $\alpha\beta$, respectively. A basis for the representation in the six-dimensional space which projects onto the basis (6.1) is

$$\mathbf{d}_1^* = (\mathbf{a}_1^*, c\mathbf{a}_1^*), \mathbf{d}_n^* = (\mathbf{a}_n^*, -c\mathbf{a}_{2n}^*) \quad (6.3)$$

(arbitrary constant c).

The 20 vertices of the dodecahedron form the centres of the faces of an icosahedron. One obtains 20 vertices of an icosahedron by the action of I on the vector $\mathbf{a}_1^* + \mathbf{a}_2^* + \mathbf{a}_3^*$. Among these 20 vectors of M^* there are again six rationally independent ones. If one denotes these by $\mathbf{v}_1^*, \dots, \mathbf{v}_6^*$ one can express them in terms of the basis (6.1).

$$\mathbf{v}_i^* = \sum_{j=1}^6 S_{ij} \mathbf{d}_j^*,$$

with

$$S = \begin{pmatrix} 1 & 1 & 1 & 0 & 0 & 0 \\ 1 & 0 & 1 & 1 & 0 & 0 \\ 1 & 0 & 0 & 1 & 1 & 0 \\ 1 & 0 & 0 & 0 & 1 & 1 \\ 1 & 1 & 0 & 0 & 0 & 1 \\ 0 & 1 & 0 & -1 & 0 & 1 \end{pmatrix}. \quad (6.4)$$

Because $\det(S) = 1$, the vectors \mathbf{v}_i^* also form a basis for M^* . Hence a set M^* generated by the 12 vertices of an icosahedron belongs to the same Bravais class as the one generated by the 20 vertices of a dodecahedron.

Another lattice is obtained if the set M^* is generated by the 30 edges of the icosahedron. These are the 30 vectors obtained by the action of I on $\mathbf{a}_1^* + \mathbf{a}_2^*$. Also in this case there are six rationally independent ones, which may be expressed in terms of (6.1).

$$\mathbf{f}_i^* = \sum_{j=1}^6 S_{ij} \mathbf{d}_j^*,$$

with

$$S = \begin{pmatrix} 1 & 1 & 0 & 0 & 0 & 0 \\ 1 & 0 & 1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 & 1 \\ 1 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 1 \\ 0 & 1 & 0 & 0 & 0 & 1 \end{pmatrix}. \quad (6.5)$$

Because $\det(S) = 2$ the lattice obtained from the embedding of $\mathbf{f}_1^*, \dots, \mathbf{f}_6^*$ is a centring of that spanned by $\mathbf{d}_1^*, \dots, \mathbf{d}_6^*$. The diffraction spots of such a lattice have components z_i with respect to the basis (6.3) that satisfy the reflection condition: $\sum z_i = \text{even}$.

A related lattice is that spanned by the six vectors \mathbf{w}_i^* :

$$\mathbf{w}_i^* = \sum_{j=1}^6 S_{ij} \mathbf{d}_j^*,$$

Table 2. The generators of the matrix group Γ for some of the dihedral groups, in six Bravais classes

Group	Centring	$\Gamma(\alpha)$	$\Gamma(\beta)$	Decomposition	
D_5	—	$\begin{pmatrix} 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \\ 1 & 0 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \end{pmatrix}$	$\Gamma_1 + \Gamma_3 + \Gamma_4$	
D_8	—	$\begin{pmatrix} 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}$	$\begin{pmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 \\ 0 & 0 & -1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}$	$\Gamma_1 + \Gamma_5 + \Gamma_7$	
D_{10}	$\sum_{i=1}^5 z_i = \text{even}$	—	$\begin{pmatrix} 0 & 0 & 0 & -1 & 0 \\ 1 & 1 & 1 & 1 & 0 \\ -1 & 0 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}$	$\Gamma_1 + \Gamma_5 + \Gamma_7$
D_{12}	$\sum_{i=1}^5 z_i = \text{even}$	—	$\begin{pmatrix} 0 & 0 & -1 & 0 & 0 \\ 0 & 0 & 1 & -1 & 0 \\ -1 & -1 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}$	$\begin{pmatrix} 0 & 1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix}$	$\Gamma_1 + \Gamma_5 + \Gamma_9$

with

$$S = \begin{pmatrix} 1 & 1 & -1 & -1 & 1 & -1 \\ 0 & 0 & 2 & 0 & 0 & 0 \\ 1 & -1 & -1 & -1 & -1 & 1 \\ 0 & 0 & 0 & 2 & 0 & 0 \\ 0 & 0 & 0 & 0 & 2 & 0 \\ -1 & 1 & 1 & 1 & -1 & 1 \end{pmatrix}. \quad (6.6)$$

Here $\det(S) = 32$ and the centring is not equivalent with the former. The diffraction spots have components z_i with respect to the basis (6.3) that are either all even or all odd. Icosahedral quasi-crystals belong either to the Bravais class of the lattice generated by (6.3) or to one of its centring. Quasi-crystals as projections of structures in six dimensions have already been discussed by various authors (Duneau & Katz, 1985; Bak, 1985*a, b*; Elser, 1985; Kramer, 1985). Bak has already noticed the difference between the two icosahedral lattices (6.3) and (6.5) which he calls s.i. and b.c.i., respectively.

Corresponding to each of these Bravais classes are two arithmetic crystal classes: in each Bravais class one with point group $I \times C_2$ and one with I . For each of these arithmetic crystal classes one may determine the possible space groups. Calculation according to Fast & Janssen (1971) shows that the classes with point group $I \times C_2$ have one, two or three space groups, whereas those with point group I have three space groups each. The generators and nonprimitive translations for these space groups belonging to the icosahedral system are given in Table 3. These should describe the symmetry of icosahedral quasi-crystals. Because non-symmorphic symmetries may occur it is worthwhile looking for systematic extinctions.

The basis of the direct lattice Σ corresponding to the reciprocal lattice Σ^* generated by the vectors \mathbf{d}_i^* of (6.3) is readily constructed. It is formed by the vectors

$$\mathbf{d}_1 = \frac{1}{2}[\mathbf{a}_1^*, (1/c)\mathbf{a}_1^*], \quad \mathbf{d}_n = \frac{1}{2}[\mathbf{a}_n^*, -(1/c)\mathbf{a}_{2n}^*]. \quad (6.7)$$

The metric tensor of the six-dimensional lattice Σ^* is given by the scalar products

$$\mathbf{d}_i^* \cdot \mathbf{d}_i^* = 1 + c^2; \quad \mathbf{d}_i^* \cdot \mathbf{d}_j^* = \frac{1}{5}(1 - c^2)\sqrt{5}, \quad i \neq j. \quad (6.8)$$

The direct lattice corresponding to the lattice with basis (6.5) belongs to the Bravais class of the reciprocal lattice with basis (6.6) and *vice versa*. These two direct lattices are centring of that with basis (6.7). The first is a centring given by the two points (000000) and $(\frac{1}{2}\frac{1}{2}\frac{1}{2}\frac{1}{2}\frac{1}{2}\frac{1}{2})$, the second by the 32 points (000000), $(\frac{1}{2}\frac{1}{2}0000)$, $(\frac{1}{2}0\frac{1}{2}000)$, ..., $(0000\frac{1}{2}\frac{1}{2})$, $(\frac{1}{2}\frac{1}{2}\frac{1}{2}\frac{1}{2}\frac{1}{2}\frac{1}{2})$, $(00\frac{1}{2}\frac{1}{2}\frac{1}{2}\frac{1}{2})$, ..., $(\frac{1}{2}\frac{1}{2}\frac{1}{2}\frac{1}{2}00)$. The corresponding arithmetic crystal classes with point group I can be denoted by $P532$, $I532$ and $F532$, respectively, and those with point group $I \times C_2$ by $P5\bar{3}m$, $I5\bar{3}m$ and $F5\bar{3}m$.

For the choice $c = 1$ the basis vectors (6.3) are of equal length and mutually perpendicular according to (6.8). In that case the lattice Σ^* is hypercubic. So the set M^* for an icosahedral quasi-crystal is the projection of a six-dimensional hypercubic lattice. The direct lattice is for $c = 1$ also hypercubic. For a general choice of c the metric tensor has two parameters.

7. Concluding remarks

We have shown that quasi-crystals, which have now been observed experimentally, are special cases of

Table 3. Generators Γ and nonprimitive translations t of the space groups in the two icosahedral six-dimensional Bravais classes

The generators for the groups with point group $I \times C_2$ are the same as for those with point group I plus the central inversion ($p = 0, 1$ or 2).

	$\Gamma(\alpha)$					$\Gamma(\beta)$					$\Gamma(\gamma)$																	
	$\begin{pmatrix} 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & 0 & 0 & 0 & 1 \\ 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 \\ 0 & 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 \\ 0 & 1 & 0 & 0 & 0 \end{pmatrix}$	$\begin{pmatrix} -1 & 0 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 \\ 0 & 0 & 0 & 0 & -1 \\ 0 & 0 & 0 & 0 & 0 \end{pmatrix}$																									
	$t(\alpha)$						$t(\beta)$						$t(\gamma)$															
$P532$	$p/5$	0	$p/5$	0	0	$4p/5$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0		
$I532$	$p/5$	$p/5$	$-p/5$	$2p/5$	$4p/5$	$-p/5$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	
$F532$	$-3p/10$	$p/10$	$p/5$	0	$2p/5$	$4p/5$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	
$P\overline{5}3m$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
	0	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
$I\overline{5}3m$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
$F\overline{5}3m$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
	0	$\frac{1}{4}$	0	0	$\frac{1}{4}$	$\frac{1}{4}$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
	0	0	$\frac{1}{4}$	$\frac{1}{4}$	$\frac{1}{4}$	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0

incommensurate crystals. The symmetry description may be given, similarly to IC phases, by an embedding into a higher-dimensional space. This situation is actually very common in physics. Often the symmetry of a system is larger than the purely Euclidean one. The larger symmetry group may sometimes be interpreted as a transformation group in more than three dimensions. The best known example is the hydrogen atom, where the symmetry (for bound states) is given by the orthogonal group in four dimensions. In IC phases one encounters the same situation, but there the additional dimensions have a simple physical interpretation: that of the phase of the modulation.

The point group on which the higher-dimensional space is constructed is always reducible in a sum of three- and d -dimensional components. The resulting lattice, however, can in principle have a higher symmetry. For example, the two-dimensional structure considered in § 3 gives rise to a decagonal lattice in four dimensions. The embedding, however, still contains an arbitrary factor c , which represents the length scale in the additional space. If the factor c is chosen to be unity, the resulting lattice belongs to the icosahedral lattice in four dimensions, which has a holohedral point group of order 240 (Brown *et al.*, 1978). Experimentally this has consequences. For a reciprocal-lattice vector of Σ^* in a general position, there are 239 other equivalent vectors. The corresponding diffraction peaks should show the same intensity for all 240 vectors, although in three dimensions they do not have the same length, and are not symmetry related. In this way such an irreducible structure would be observable.

Just as for IC phases one could expect new lattice modes. For IC phases these are the phason and amplitudon modes. In principle, quasi-crystals can also show d additional phason modes. Here, however,

the difference between modulated and quasi-crystals may be important. As seen in the examples of the Fibonacci chain and the Penrose tiling, the higher-dimensional structures corresponding to quasi-crystals have disconnected elements. The situation is comparable with that of modulated crystals in the discommensuration domain, where the modulation function also has discontinuities or, at least, a non-smooth behaviour. There the result is that the phason frequency is no longer zero (Janssen & Tjon, 1983). For the same reason one may expect for quasi-crystals a non-zero frequency phason. The reason is that the motion of particles is smooth when the phase of the modulation of a sinusoidally modulated crystal is changed, but that the particles have to jump from one position to another when the 'phase' of a quasi-crystal is changed.

The self-similarity properties of quasi-crystals can be accounted for also by the periodic structure that is by definition already self-similar.

I have not talked about the origin of the quasi-crystalline state. A partial answer is given by the papers using Landau theory mentioned in § 1. A microscopic theory for the stability of the quasi-crystalline state has not been given as far as I know. The stability of the quasi-crystalline state has relations with the problem of the densest packing of spheres. In two dimensions the densest packing of circles is the hexagonal structure, where each circle touches six others. In three dimensions the problem is unsolved. Both f.c.c. and h.c.p. structures have a packing fraction $(\pi/6)\sqrt{2}$ but it has not been shown that this is a maximum. {An upper bound $\sqrt{18}[\cos^{-1}(\frac{1}{3}) - \pi/3]$ has been given by Rogers (1958).} In the case of h.c.p. 12 spheres touch each sphere, but the orientations of the 12 common points are not regularly distributed (there are angles of 60 and 90°). A regular distribution

would give the 12 vertices of an icosahedron and locally icosahedral symmetry, which is, of course, in conflict with periodicity, but as we have seen not with quasi-periodicity.

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The Electron Distribution in Silicon. A Comparison between Experiment and Theory

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Abstract

Deformation and valence-electron densities in silicon are derived *via* Fourier summation and multipole refinement of highly accurate measurements of X-ray structure factors. These results provide a new perspective for the comparison between theory and experiment. The model electron density derived from experiment is in quantitative agreement with recent solid-state calculations, but not with earlier experimental results reported by Yang & Coppens [*Solid State Commun.* (1974), **15**, 1555-1559].

Introduction

Experimental electron distributions for crystalline silicon have been the subject of numerous investigations [e.g. see Scheringer (1980), Fehlmann (1979),

Price, Maslen & Mair (1978; referred to below as PMM), Yang & Coppens (1974; referred to as YC), Aldred & Hart (1973; referred to as AH) and references therein]. In this work we take advantage of recent highly accurate experimental reports on silicon, which, in combination with the earlier measurements of AH, provide data sets of extraordinarily high quality.

The important 222 reflection in silicon was remeasured by Alkire, Yelon & Schneider (1982), with an accuracy better, by a factor of between two and ten, than previous measurements. Alkire *et al.* report F_{222} at room temperature with an accuracy of 0.5%, a measurement of accuracy similar to the AH data (~0.1%).

Teworte & Bonse (1984) measured silicon structure factors for 16 reflections at room temperature with both Ag $K\alpha_1$ and Mo $K\alpha_1$ radiations which were also used by AH. Teworte & Bonse's work verified AH's measurements, and confirmed the claimed accuracy

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