Acta Cryst. (1986). A42, 36-43

The Diffraction Pattern of Projected Structures

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(*Received* 2 *April* 1985; *accepted* 24 *July* 1985)

Abstract

A method for calculating the properties of structures obtained by projection is developed and applied to a three-dimensional generalization of the Penrose tiling. The diffraction pattern is shown in general to consist of a dense set of delta-function peaks. For the Penrose model the pattern in addition has the symmetry of the icosahedron.

1. Introduction

A global construction of the remarkable nonperiodic tiling of the plane discovered by Penrose (1974) was first given by de Bruijn (1981). He also showed that the construction could be interpreted as the projection of a five-dimensional lattice structure into a two-dimensional subspace. This method was later used by Kramer & Neri (1984) to construct a three-dimensional generalization of the Penrose tiling by projecting from twelve dimensions. The form of the projection in this case was determined by the requirement that the projected basis vectors constitute a representation of the icosahedral group. We thus refer to the three-dimensional Penrose pattern as an icosahedral quasilattice. The term 'quasilattice' was first used by Mackay (1981) in perhaps the earliest speculations about incorporating Penrose patterns into crystallography. This term also seems appropriate in view of the recently proposed classification scheme of Levine & Steinhardt (1984) based on quasiperiodicity.

The aim of the present paper is to give a calculation of the diffraction pattern of the icosahedral quasilattice using the method of projection. We also hope to show that the techniques used lend themselves naturally to the calculation of a number of other properties of projected structures. Of these we include a calculation of the density and the derivation of a formula giving the frequency of occurrence of subpatterns. In the following section we treat the more easily visualized case of a one-dimensional structure obtained by projecting from two dimensions. The tools developed there are readily generalized in § 3 where we present analogous calculations for the icosahedral quasilattice.

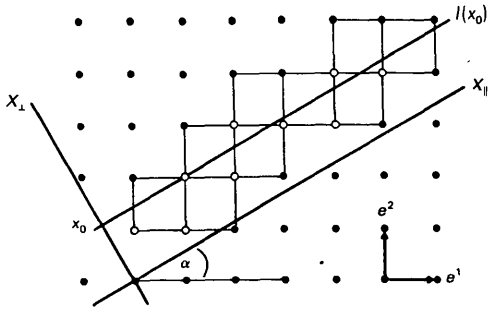

Fig. 1. The construction of the set $\mathcal{S}(x_0)$.

2. A one-dimensional example

The discussion in this section will involve constructions confined to the Euclidean plane. We will let $\mathbb{Z}^2$ denote the square lattice considered as the set of points $\mathbf{n}=(n^1, n^2)$ having integer Cartesian coordinates. Rotated by an angle α with respect to the former coordinate system, we introduce another orthogonal coordinate system with axes labeled $X_\parallel$ and $X_\perp$ (see Fig. 1). The only thing we require about α is that $\tan\alpha$ is irrational. The $X_\parallel$ axis will later assume the role of the 'physical' subspace into which a subset of $\mathbb{Z}^2$ will be projected to give the desired one-dimensional structure.

The selection of a subset of the lattice points is accomplished by first constructing a line $l(x_0)$ parallel to the $X_\parallel$ axis and intersecting the $X_\perp$ axis at the point $x_0 \in X_\perp$ (see Fig. 1). We then consider the elementary square cell of $\mathbb{Z}^2$:

$$C(\mathbf{n}) = \{(x^1, x^2) | x^1 \in [n^1, n^1+1) \text{ and } x^2 \in [n^2, n^2+1)\}.$$

The subset $\mathcal{S}(x_0) \subset \mathbb{Z}^2$ is now given by the definition

$$\mathcal{S}(x_0) = \{\mathbf{n} \in \mathbb{Z}^2 | l(x_0) \cap C(\mathbf{n}) \neq \emptyset\}. \qquad (1)$$

These are just the lower left-hand vertices of all those square cells cut by the line $l(x_0)$ (open circles in Fig. 1).

The last step in the construction involves projecting the set $\mathcal{S}(x_0)$ into $X_\parallel$. If $\mathbf{e}^1$ and $\mathbf{e}^2$ are the standard basis vectors of $\mathbb{Z}^2$, then upon projection

$$\mathbf{e}^i = P_\parallel(\mathbf{e}^i) + P_\perp(\mathbf{e}^i) = e^i_\parallel + e^i_\perp, \quad i = 1, 2,$$

where $P_\parallel$ and $P_\perp$ denote projection operators. A

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matrix representing $P_\parallel$ is given by

$$e_\parallel^i = (P_\parallel)^{ij}\mathbf{e}^j \qquad (2)$$
$$= \begin{pmatrix} \cos^2\alpha & \cos\alpha\sin\alpha \\ \sin\alpha\cos\alpha & \sin^2\alpha \end{pmatrix}^{ij} \mathbf{e}^j.$$

The corresponding formula for $P_\perp$ can be obtained using $P_\perp = 1 - P_\parallel$.

The projected one-dimensional structure $\mathcal{S}_\parallel(x_0)$ can now be expressed as the set

$$\mathcal{S}_\parallel(x_0) = \left\{ \sum_{i=1}^{2} n^i e_\parallel^i \,\middle|\, (n^1, n^2) \in \mathcal{S}(x_0) \right\}.$$

Using the fact that the definition of $l(x_0)$ is given by

$$l(x_0) = \left\{ (x^1, x^2) \,\middle|\, \sum_{i=1}^{2} x^i e_\perp^i = x_0 \right\},$$

a compact definition of $\mathcal{S}_\parallel(x_0)$ takes the form

$$\mathcal{S}_\parallel(x_0) = \left\{ \sum_{i=1}^{2} \text{int}\,(x^i) e_\parallel^i \,\middle|\, \sum_{i=1}^{2} x^i e_\perp^i = x_0 \right\},$$

where int (x) denotes the greatest integer less than or equal to x.

It is quite easy to show that $\mathcal{S}_\parallel(x_0)$ represents the set of endpoints of a filling of the line $X_\parallel$ by a sequence of line segments of two types, one of length $|e_\parallel^1| = |\cos\alpha|$ and one of length $|e_\parallel^2| = |\sin\alpha|$ (see Fig. 2). Owing to the irrationality of the slope of the line $l(x_0)$, the ordering of the two line segments is nonperiodic. Had we imposed further that $\tan\alpha$ was a quadratic irrational, then the sequence obtained would have possessed additional properties associated with quasiperiodicity (Levine & Steinhardt, 1984). Although many of the interesting properties of Penrose patterns appear to be closely related to quasiperiodicity, our subsequent calculations will not rely on these properties.

We will now discuss the sense in which the infinite structure $\mathcal{S}_\parallel(x_0)$ depends on the choice of the parameter x_0. Firstly, we disregard the singular case when $l(x_0)$ passes exactly through a lattice point. Also, it will be more convenient to describe l in terms of its intersection with one of the $x^1 = n$ grid lines of the unrotated coordinate system. If this intersection occurs at $(x^1, x^2) = (n, y_n)$, we denote the line $l(y_n)$. It is clear that adding an integer to y_n only has the effect of shifting the projected structure $\mathcal{S}_\parallel$ by a finite amount. Now, if the fractional part of y_n is replaced by the fractional part of y_{n+m} for any integer m, then we again only shift $\mathcal{S}_\parallel$ by a finite amount. The formula for the sequence of these fractional parts is given by

$$y_{n+m} = (\tan\alpha)m + y_n \mod 1.$$

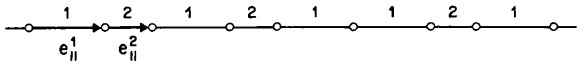

Fig. 2. The filling of $X_\parallel$ with two line segments.

Provided that $\tan\alpha$ is irrational, there is a well known theorem [see, for example, Kuipers & Niederreiter (1974)] that states that the distribution of values y_{n+m} for $m = 1, \ldots, M$ becomes uniformly dense in the interval $[0, 1]$ as $M \to \infty$. Consequently, given the physical assumption of a finite precision in the value of y_n (which leaves a certain small fraction of $\mathcal{S}_\parallel$ ambiguous), all fractional values generate structures that are simply related by shifts. Since x_0 is linearly related to y_n, the sense in which the infinite structure $\mathcal{S}_\parallel(x_0)$ is independent of x_0 should now be clear. Since we confine ourselves to properties of infinite structures, the parameter x_0 will be dropped when not explicitly needed.

One justification of the projection method is provided by the natural appearance in many calculations of objects projected into the orthogonal subspace $X_\perp$. Before proceeding to the diffraction pattern we will consider two such examples. We first calculate the density of points in $\mathcal{S}_\parallel$ and then give a method for obtaining the frequency of occurrence of specific subpatterns within $\mathcal{S}_\parallel$.

In the rotated coordinate system we consider a large rectangular region $\mathcal{R}$ having sides of length $L_\parallel$ and $L_\perp$ in the $X_\parallel$ and $X_\perp$ directions respectively. For each lattice point $\mathbf{n} \in \mathcal{R}$ we define the function

$$\theta_{\mathbf{n}}(x_0) = \begin{cases} 1 & \text{if } l(x_0) \cap C(\mathbf{n}) \neq \emptyset \\ 0 & \text{otherwise.} \end{cases}$$

The value of the integral

$$\int_{-\infty}^{\infty} \theta_{\mathbf{n}}(x_0)\, dx_0 = v$$

is easily seen to be just the length, or measure μ, of the line segment obtained by projecting a unit square cell into $X_\perp$ (see Fig. 3):

$$v = \mu\{P_\perp[C(\mathbf{n})]\} = |\cos\alpha| + |\sin\alpha|. \qquad (3)$$

Up to boundary corrections proportional to the

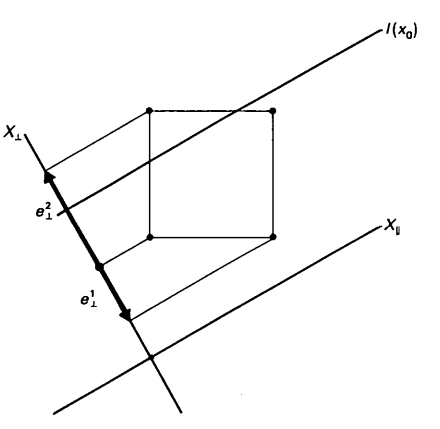

Fig. 3. The projection of a square cell into $X_\perp$.

perimeter $L=2(L_\parallel+L_\perp)$, the area of $\mathcal{R}$ can be expressed as the number of enclosed square cells:

$$L_\parallel L_\perp = \sum_{\mathbf{n}\in\mathcal{R}} 1+O(L) \tag{4}$$

$$= \sum_{\mathbf{n}\in\mathcal{R}} \int_0^{L_\perp} \theta_{\mathbf{n}}(x_0)\,dx_0/v+O(L)$$

$$= \int_0^{L_\perp} (L_\parallel\rho)\,dx_0/v+O(L) \tag{5}$$

$$= (\rho/v)L_\perp L_\parallel+O(L). \tag{6}$$

In (5) we interchanged the order of summation and integration and introduced the density

$$\rho=(L_\parallel)^{-1}\sum_{\mathbf{n}\in\mathcal{R}}\theta_{\mathbf{n}}(x_0). \tag{7}$$

Independent of the above remarks concerning the parameter x_0, it can easily be seen that the limit $L_\parallel\to\infty$ of (7) does not depend on x_0. Comparing (4) with (6) we see that

$$\rho=v=|\cos\alpha|+|\sin\alpha|.$$

Before continuing to the next calculation we will consider an alternative definition of the set $\mathcal{S}(x_0)$. The condition

$$l(x_0)\cap C(\mathbf{n})\neq\emptyset \tag{8}$$

in (1) is equivalent to the condition on the lattice point $\mathbf{n}$ that its projection $P_\perp(\mathbf{n})$ lies within the projection of a particular square cell determined by x_0. Formally, (8) is equivalent to the statement

$$x_0\in P_\perp[C(\mathbf{n})],$$

which, upon translation implies

$$-P_\perp(\mathbf{n})\in P_\perp[C(\mathbf{n})]-P_\perp(\mathbf{n})-x_0$$

or

$$P_\perp(\mathbf{n})\in P_\perp[-C(\mathbf{o})]+x_0\equiv C_\perp(x_0)$$

after an inversion and using properties of $P_\perp$. We thus have the alternative definition

$$\mathcal{S}(x_0)=\{\mathbf{n}\in\mathbb{Z}^2|P_\perp(\mathbf{n})\in C_\perp(x_0)\}. \tag{9}$$

We will now show that in fact $C_\perp(x_0)$ is filled uniformly by the projections $P_\perp[\mathcal{S}(x_0)]$. For convenience, we consider instead of $C_\perp(x_0)$ the interval

$$I_k=\{xe_\perp^2|x\in[k,k+1)\}\subset X_\perp$$

for some integer k. Since $C_\perp(x_0)$ is contained in the union of such intervals with different k, it is enough to show that each of them is filled uniformly. The set of lattice points $\mathbf{n}$ with projections

$$n_\perp=n^1e_\perp^1+n^2e_\perp^2\in I_k$$

is given by the sequence

$$\mathbf{n}_m=(m,n^2(m)),\qquad m=-M,\ldots,M,$$

where $n^2(m)$ is the unique integer multiple of $e_\perp^2$ that places $n_\perp$ into the desired interval I_k. The sequence of projections

$$P_\perp(\mathbf{n}_m)=(k+x_m)e_\perp^2$$

is given by the formula

$$x_m=(e_\perp^1\,.\,e_\perp^2/e_\perp^2\,.\,e_\perp^2)m=-(\tan\alpha)m \mod 1.$$

By the same theorem invoked earlier, the values x_m will be distributed uniformly in the limit $M\to\infty$ provided of course that $\tan\alpha$ is irrational.

If $n_\parallel\in\mathcal{S}_\parallel$ is the projection of some lattice point $\mathbf{n}\in\mathcal{S}$, then the arrangement of points $n'_\parallel\in\mathcal{S}_\parallel$ in the vicinity of $n_\parallel$ is completely determined by the location of $n_\perp=P_\perp(\mathbf{n})\in\mathcal{S}_\perp$ within $C_\perp$. This can be understood if we begin at $\mathbf{n}$ and consider the neighboring lattice points $\mathbf{n}=\mathbf{n}+\mathbf{k}$. To see whether $n'_\parallel=n_\parallel+k_\parallel$ also belongs to $\mathcal{S}_\parallel$ we simply check to see if $n'_\perp=n_\perp+k_\perp$ lies within $C_\perp$. The allowed increments $k_\parallel$ are therefore completely determined by the location of the starting point $n_\perp$ in $C_\perp$.

We will define a 'subpattern' as the projection $P_\parallel(\mathcal{K})$ of a finite set of lattice vectors

$$\mathcal{K}=\{\mathbf{o},\mathbf{k}_1,\ldots,\mathbf{k}_m\}\subset\mathbb{Z}^2,$$

where $\mathbf{k}_1,\ldots,\mathbf{k}_m$ are nonzero and distinct. A subpattern $P_\parallel(\mathcal{K})$ is 'rooted' at $n_\parallel$ if $n_\parallel+P_\parallel(\mathcal{K})\in\mathcal{S}_\parallel$. The question of whether a point $n_\parallel=P_\parallel(\mathbf{n})\in\mathcal{S}_\parallel$ occupies the 'root' position of a subpattern $P_\parallel(\mathcal{K})$ can be answered simply by checking whether

$$P_\perp(\mathbf{n}+\mathcal{K})\subset C_\perp. \tag{10}$$

The set of all lattice points $\mathbf{n}\in\mathcal{S}$ such that (10) holds will be denoted $\mathcal{N}_\mathcal{K}$. After a series of translations, (10) can be rewritten in the equivalent form

$$P_\perp(\mathbf{n})\in\bigcap_{\mathbf{k}\in\mathcal{K}}[C_\perp-P_\perp(\mathbf{k})]\equiv C_\perp(\mathcal{K}).$$

We are now ready to compute the probability that a point $n_\parallel$ drawn at random from $\mathcal{S}_\parallel$ occupies the root of a subpattern $P_\parallel(\mathcal{K})$. Using the above notation, we are interested in knowing the conditional probability

$$p(\mathcal{K})=\text{prob}\,(\mathbf{n}\in\mathcal{N}_\mathcal{K}|\mathbf{n}\in\mathcal{S})$$

$$=\text{prob}\,(n_\perp\in C_\perp(\mathcal{K})|n_\perp\in\mathcal{S}_\perp).$$

According to our earlier discussion, the sample points $n_\perp\in\mathcal{S}_\perp$ are distributed *uniformly* in the interval $C_\perp$. This implies that the probability measure is proportional to the usual 'volume' measure μ, which in this case is just the length:

$$p(\mathcal{K})=\mu[C_\perp(\mathcal{K})]/\mu(C_\perp). \tag{11}$$

We conclude this discussion of probabilities with a simple application of formula (11). For the range of angles $0<\alpha<\pi/4$, the larger of the two intervals

that fills $X_\parallel$ has length $|e_\parallel^1| = \cos\alpha$. We now wish to know the probability that a point drawn at random from $\mathcal{S}_\parallel$ is the common endpoint of two adjacent large intervals. In this case the subpattern is defined by $\mathcal{K} = \{\mathbf{o}, \mathbf{e}^1, -\mathbf{e}^1\}$ and $C_\perp(\mathcal{K})$ is given by the intersection of three intervals:

$$C_\perp(\mathcal{K}) = C_\perp \cap (C_\perp - e_\perp^1) \cap (C_\perp + e_\perp^1).$$

Using the result (3) for $C_\perp$, we have that

$$p(\mathcal{K}) = (\cos\alpha - \sin\alpha)/(\cos\alpha + \sin\alpha).$$

In calculating the diffraction pattern we consider the simple case where identical atoms occupy all the projected points $n_\parallel \in \mathcal{S}_\parallel$. It will be advantageous to view the argument of the structure factor $S(g_\parallel)$ as the projection of a two-dimensional reciprocal-lattice vector $\mathbf{g} \in (\mathbb{Z}^2)^*$. Here we use the convention

$$1 = e^{i\mathbf{g}\cdot\mathbf{n}} = e^{ig_\parallel \cdot n_\parallel} e^{ig_\perp \cdot n_\perp} \tag{12}$$

for all $\mathbf{n} \in \mathbb{Z}^2$ and $\mathbf{g} \in (\mathbb{Z}^2)^*$. In using the Euclidean inner product in (12) we have implicitly chosen to express the elements of $(\mathbb{Z}^2)^*$ in terms of the same basis used for real space. Explicit formulas are given by

$$\mathbf{g} = 2\pi \sum_{i=1}^{2} m^i \mathbf{e}^i$$

$$g_\parallel = 2\pi \sum_{i=1}^{2} m^i e_\parallel^i, \qquad g_\perp = 2\pi \sum_{i=1}^{2} m^i e_\perp^i, \tag{13}$$

where $(m^1, m^2) \in \mathbb{Z}^2$. Identity (12) forms the basis of our calculation of the structure factor (for N atoms):

$$S(g_\parallel) = \sum_{n_\parallel \in \mathcal{S}_\parallel} e^{ig_\parallel \cdot n_\parallel} = \sum_{n_\perp \in \mathcal{S}_\perp} e^{-ig_\perp \cdot n_\perp}$$

$$\simeq [N/\mu(C_\perp)] \int_{C_\perp} e^{-ig_\perp \cdot x_\perp} \, dx_\perp \times (\text{phase}).$$

The last step follows in the limit $N \to \infty$ since, as discussed above, the projections $n_\perp$ uniformly fill up the interval $C_\perp$. Neglecting the phase and using $v = \mu(C_\perp)$ given by (3), we have

$$S(g_\parallel) = N(\sin z)/z, \qquad z = \tfrac{1}{2}|g_\perp| v.$$

The relation between $g_\parallel$ and $g_\perp$ is given by (13). The incommensurate nature of the projection determines a unique $\mathbf{g}$ given $g_\parallel$, which in turn determines $g_\perp$ uniquely. Since the scattering is proportional to the number of atoms, we obtain a set of delta-function peaks. In spite of the fact that these peaks form a dense set, each individual peak is 'isolated' in the sense that one can always find a suitably small neighborhood of the peak position such that all the other peaks in that neighborhood have intensities that are arbitrarily small.

3. The icosahedral quasilattice

We have tried to present the discussion of the one-dimensional example in such a way that generalization to higher-dimensional constructions is straightforward. Here we focus on the icosahedral quasilattice, which is obtained by projecting a subset of the six-dimensional lattice $\mathbb{Z}^6$ into a special three-dimensional hyperplane $X_\parallel$. The orientation of $X_\parallel$ relative to the lattice is determined by the requirement that the projected basis vectors $e_\parallel^1, \ldots, e_\parallel^6$ coincide with the six vertex axes of the icosahedron. We will see that this choice leads to a diffraction pattern having icosahedral symmetry.

A useful way of specifying the orientation of the space $X_\parallel$ is in terms of the representation (2) of the projection operator:

$$(P_\parallel)^{ij} = 20^{-1/2} \begin{pmatrix} 5^{1/2} & 1 & 1 & 1 & 1 & 1 \\ 1 & 5^{1/2} & 1 & -1 & -1 & 1 \\ 1 & 1 & 5^{1/2} & 1 & -1 & -1 \\ 1 & -1 & 1 & 5^{1/2} & 1 & -1 \\ 1 & -1 & -1 & 1 & 5^{1/2} & 1 \\ 1 & 1 & -1 & -1 & 1 & 5^{1/2} \end{pmatrix}^{ij}.$$

Both this matrix and the matrix representing $P_\perp = 1 - P_\parallel$ have rank three and satisfy the required properties

$$P_\parallel^2 = P_\parallel, \qquad P_\perp^2 = P_\perp, \qquad P_\parallel P_\perp = 0.$$

The two sets of projected basis vectors have the same norms,

$$|e_\parallel^i| = |e_\perp^i| = 2^{-1/2}, \qquad i = 1, \ldots, 6,$$

but different pairwise inner products:

$$\cos(e_\parallel^i, e_\parallel^j) = -\cos(e_\perp^i, e_\perp^j), \qquad i \neq j. \tag{14}$$

A calculation of all fifteen combinations

$$\cos(e_\parallel^1, e_\parallel^i) = 5^{-1/2}, \qquad i = 2, \ldots, 6$$

$$\cos(e_\parallel^{2+i}, e_\parallel^{2+j}) = \begin{cases} 5^{-1/2}, & i-j = \pm 1 \bmod 5 \\ -5^{-1/2}, & i-j = \pm 2 \bmod 5 \end{cases}$$

shows that the vectors $e_\parallel^i$ may be identified with the set of six vertex directions shown in Fig. 4. The corresponding identification for the vectors $e_\perp^i$ is shown in Fig. 5. The choice of surrounding one of the vectors symmetrically by the other five was purely a matter of convention. Other conventions result if,

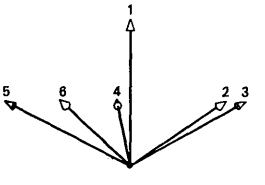

Fig. 4. The projected basis vectors $e_\parallel^i$.

say, one of the basis vectors $\mathbf{e}^i$ is reversed, thereby simultaneously reversing $e^i_\parallel$ and $e^i_\perp$.

The condition (8) used in defining the subset of lattice points $\mathscr{S}(x_0)$ is easily generalized to apply to the present situation. The notation $C(\mathbf{n})$ now represents a six-dimensional hypercubic cell of $\mathbb{Z}^6$ and $l(x_0)$ is a three-dimensional hyperplane parallel to $X_\parallel$ parametrized by $x_0 \in X_\perp$. The construction of the set $\mathscr{S}(x_0)$ suggests the general term *hypercubic covering*. The hyperplane $l(x_0)$ is 'covered' by the hypercubic cells of $\mathbb{Z}^6$ in the sense that every point $x \in l(x_0)$ belongs to some cell of the covering and, conversely, that every cell in the covering contains some $x \in l(x_0)$. The set $\mathscr{S}(x_0)$ is then the set of all lattice points $\mathbf{n}$ of the cells that belong to the covering. The projection $P_\parallel[\mathscr{S}(x_0)]$ gives a particular realization of the icosahedral quasilattice:

$$\mathscr{S}_\parallel(x_0) = \left\{ \sum_{i=1}^{6} \operatorname{int}(x^i) e^i_\parallel \,\middle|\, \sum_{i=1}^{6} x^i e^i_\perp = x_0 \right\}.$$

For the interpretation of $\mathscr{S}_\parallel$ as the vertices of a rhombohedral space filling we refer to the work of Kramer & Neri (1984) where essentially the same construction was used. Here we merely note that the twenty different rhombohedra of the space filling (distinguished by orientation as well as shape) are generated by the combinations of distinct triplets of vectors chosen from the set $\{e^1_\parallel, \ldots, e^6_\parallel\}$.

Before we can proceed with our calculations it will be necessary to know something about the projection into $X_\perp$ of the hypercubic cell:

$$C_\perp = P_\perp[C(\mathbf{o})] = \left\{ \sum_{i=1}^{6} x^i e^i_\perp \,\middle|\, x^i \in [0,1),\ i = 1, \ldots, 6 \right\}.$$

The last line defines a *zonohedron* [see, for example, Coxeter (1963)] generated by six vectors. As in this case, when the vectors $e^1_\perp, \ldots, e^6_\perp$ constitute a vertex set of the icosahedron, the zonohedron goes by the special name *triacontahedron* (see Fig. 6). Thus $C_\perp$ is a triacontahedron with edges taken from the set $\{e^1_\perp, \ldots, e^6_\perp\}$ and 'diameter'

$$|e^1_\perp - \sum_{i=2}^{6} e^i_\perp| = (1+5^{1/2})|e^1_\perp|.$$

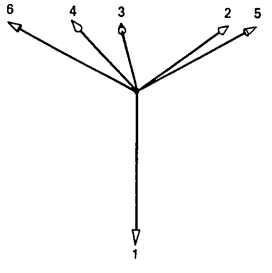

Fig. 5. The projected basis vectors $e^i_\perp$.

A particularly useful representation of the triacontahedron is the decomposition into a disjoint union of twenty rhombohedra [a stereo image is given by Mackay (1982)]. These rhombohedra are the projections of the twenty 3-cells of the 6-cube formed by taking distinct triplets of the six basis vectors. Two such projections of the analogous (but more familiar) 2-cells of the 3-cube are shown in Fig. 7. If the rhombohedral volumes are

$$v_{ijk} = |e^i_\perp \times e^j_\perp \,.\, e^k_\perp|, \tag{15}$$

then

$$\mu(C_\perp) = \sum_{1 \le i < j < k \le 6} v_{ijk} = 8^{1/2}(\sin(2\pi/5) + \sin(4\pi/5))$$

is the volume of the triacontahedron.

The straightforward generalization of our earlier calculation of the density now gives us

$$\rho = \int \mathrm{d}^3 x_0 \theta_{\mathbf{n}}(x_0) = \mu(C_\perp).$$

If we wish to restore the rhombohedra to having *unit* edge length, we must rescale by the factor $8^{-1/2}$ giving

$$\rho_{\mathrm{ISF}} = \sin(2\pi/5) + \sin(4\pi/5)$$

for the density of vertices of the icosahedral space filling.

Analogous to (9), an equivalent definition of the set $\mathscr{S}(x_0)$ sets out to determine the set of all lattice points $\mathbf{n}$ having projections $P_\perp(\mathbf{n})$ that fall inside a particular triacontahedron $C_\perp(x_0)$. As in the one-dimensional case, it can also be shown that the set of these projections $\mathscr{S}_\perp(x_0)$ fills $C_\perp(x_0)$ uniformly (a proof is given in the Appendix). Crucial of course is the fact that the vectors $e^i_\perp$ are linearly independent over the integers. The prescription, then, for computing subpattern probabilities and the diffraction pattern is exactly the same as before.

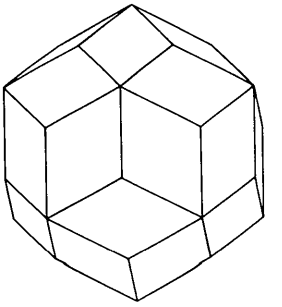

Fig. 6. The triacontahedron.

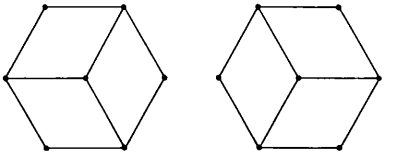

Fig. 7. Two ways of decomposing the projected 3-cube into the disjoint union of projected 2-cells.

Suppose we wish to know the probability that a vertex drawn at random from $\mathscr{S}_\parallel$ has a complete set of neighbors at the twelve icosahedral displacements $\pm e^i_\parallel$, $i=1,\ldots,6$. That is, we are interested in the probability $p(\mathscr{K})$ for the subpattern $\mathscr{K}=\{\mathbf{o},\pm\mathbf{e}^1,\ldots,\pm\mathbf{e}^6\}$. Owing to the high degree of symmetry, it is not difficult to show that the mutual intersection of the thirteen triacontahedra

$$C_\perp(\mathscr{K})=\bigcap_{\mathbf{k}\in\mathscr{K}}[C_\perp-P_\perp(\mathbf{k})]$$

is itself a triacontahedron with diameter $(-1+5^{1/2})|e^1_\perp|$. The probability, given as the ratio of volumes, is thus

$$p(\mathscr{K})=((-1+5^{1/2})/(1+5^{1/2}))^3=\tau^{-6}.$$

An unambiguous indicator of icosahedral symmetry is the diffraction pattern of the quasilattice to which we now turn. As in the one-dimensional example, one considers projections of the reciprocal-lattice vectors. Formulas completely analogous to (13) are also valid in this higher-dimensional case:

$$g_\parallel=2\pi\sum_{i=1}^{6}m^ie^i_\parallel,\ g_\perp=2\pi\sum_{i=1}^{6}m^ie^i_\perp,$$
$$(m^1,\ldots,m^6)\in\mathbb{Z}^6.$$

The counterpart of identity (12) now involves projecting the inner product $\mathbf{g}.\mathbf{n}$ into the two three-dimensional subspaces $X_\parallel$ and $X_\perp$. If identical atoms are now placed at all the vertices of the space filling, the structure factor (up to a phase) is given by the integral

$$S(g_\parallel)=[N/\mu(C_\perp)]\int_{C_\perp}e^{-ig_\perp\cdot x_\perp}\,\mathrm{d}^3x_\perp, \qquad (16)$$

where $C_\perp$ is the triacontahedron or projected 6-cube. Whenever $g_\parallel$ and $g'_\parallel$ are related by an element of the icosahedral group $A(5)$, so are their counterparts $g_\perp$ and $g'_\perp$. It then follows immediately from (16) that $|S(g_\parallel)|=|S(g'_\parallel)|$ since the integration region has the same symmetry. Icosahedral symmetry is also manifest if instead one considers a rotation $a\in A(5)$ of the quasilattice about some vertex $n_\parallel=P_\parallel(\mathbf{n})\in\mathscr{S}_\parallel$. The effect of such a rotation on the set $\mathscr{S}_\perp$ is to rotate it (within $X_\perp$) about the vertex $n_\perp=P_\perp(\mathbf{n})$. Equivalently, the rotation $a(\mathscr{S}_\perp)$ can be described by the redefinition $C'_\perp=a(C_\perp)$. Since $C'_\perp$ and $C_\perp$ differ only by a translation, the modulus of the structure factor is unchanged.

An explicit evaluation of (16) is given in the Appendix. The slice of the diffraction pattern having $g_\parallel\,.\,e^1_\parallel=0$ is shown in Fig. 8. As in the one-dimensional example, the actual pattern is dense with peaks and only the most intense are shown.

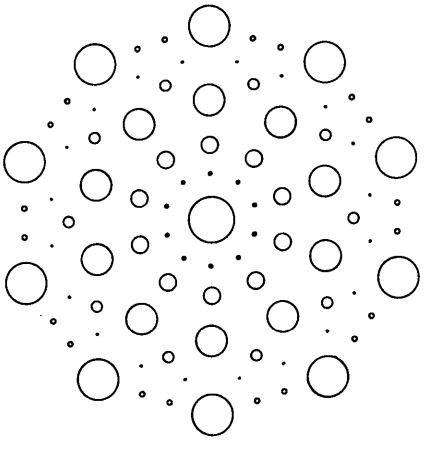

Fig. 8. The fivefold slice of the icosahedral quasilattice diffraction pattern. The circles are centered on the peak positions and have areas proportional to the intensity.

4. Discussion

The discovery by Shechtman, Blech, Gratias & Cahn (1984) of the remarkable alloy of manganese and aluminium that exhibits a point diffraction pattern with icosahedral symmetry has created considerable interest in the possibility of Penrose-tile-based crystallinity. The original proposal that a structure related to the icosahedral quasilattice could account for the observed diffraction pattern is due to Levine & Steinhardt (1984). The diffraction pattern obtained by these authors is very similar to ours but differs in the actual intensity values. This is not surprising since the assumed atomic positions of the two approaches do not agree in detail. The construction of Levine & Steinhardt is intimately related to the notion of quasiperiodicity and is perhaps a natural way of understanding ideas related to 'deflation/inflation' and 'matching rules'. Whereas quasiperiodicity in the projection context is a consequence of icosahedral symmetry, we have seen that a calculational framework exists independent of this property.

Note added: Recently, very similar ideas have been presented by Kalugin, Kitaev & Levitov (1985) and Duneau & Katz (1985).

The author thanks Paul Heckbert of NYIT for providing Figs. 4–6 and Chris Henley for comments. A special debt of gratitude is owed to David Di Vincenzo for having spotted a significant error in an early version of (23).

APPENDIX

1. Uniform density of projections in $C_\perp$

Our argument consists in expressing the distribution as the product of three independent one-dimensional distributions. Also, for convenience, we consider the equivalent problem of showing uniformity of the

distribution inside the rhombohedron

$$I(k^1, k^2, k^3) = \left\{ \sum_{i=1}^{3} (k^i + x^i) e_\perp^i \,|\, x^i \in [0, 1), i = 1, 2, 3 \right\}, \quad (k^1, k^2, k^3) \in \mathbb{Z}^3,$$

rather than the triacontahedron $C_\perp$. We choose to express a general projected lattice point in terms of the same three basis vectors:

$$\sum_{i=1}^{6} n^i e_\perp^i = \sum_{i=1}^{3} a^i e_\perp^i,$$

where

$$\begin{aligned} a^1 &= n^1 + n^5 - \tau(n^4 + n^6) \\ a^2 &= n^2 - n^4 - \tau(n^6 - n^5) \\ a^3 &= n^3 - n^6 - \tau(n^4 - n^5). \end{aligned} \tag{17}$$

Now, if $P_\perp(\mathbf{n}) \in I(k^1, k^2, k^3)$, then

$$k^i \le a^i < k^i + 1, \qquad i = 1, 2, 3. \tag{18}$$

Obviously the statement of uniformity hinges on a properly defined approach to the infinite volume limit (in $X_\parallel$). Here we choose to take a sequence of larger and larger rhombohedra:

$$\mathcal{R}(N^1, N^2, N^3) = \left\{ \sum_{i=1}^{3} x^i e_\parallel^i \,\middle|\, x^i \in (0, N^i), i = 1, 2, 3 \right\}.$$

Thus we consider only the subset of the quasilattice points $P_\parallel(\mathbf{n}) \in \mathcal{R}(N^1, N^2, N^3)$ and then take the limits $N^i \to \infty$, $i = 1, 2, 3$. Expressing the projection of an arbitrary lattice point as

$$\sum_{i=1}^{6} n^i e_\parallel^i = \sum_{i=1}^{3} b^i e_\parallel^i,$$

then

$$\begin{aligned} b^1 &= n^1 + n^5 + \tau^{-1}(n^4 + n^6) \\ b^2 &= n^2 - n^4 + \tau^{-1}(n^6 - n^5) \\ b^3 &= n^3 - n^6 + \tau^{-1}(n^4 - n^5) \end{aligned}$$

with now the conditions

$$0 < b^i < N^i, \qquad i = 1, 2, 3. \tag{19}$$

Combining inequalities (18) and (19) we have

$$-k^i < b^i - a^i < N^i - k^i, \qquad i = 1, 2, 3,$$

where

$$\begin{aligned} b^1 - a^1 &= 5^{1/2}(n^4 + n^6) \\ b^2 - a^2 &= 5^{1/2}(n^6 - n^5) \\ b^3 - a^3 &= 5^{1/2}(n^4 - n^5). \end{aligned}$$

We now propose to choose integers

$$m^i \in \{\text{int}\,[5^{-1/2}(-k^i)] + 1, \ldots, \text{int}\,[5^{-1/2}(N^i - k^i)]\}, \quad i = 1, 2, 3, \tag{20}$$

so that the equations

$$\begin{aligned} m^1 &= n^4 + n^6 \\ m^2 &= n^6 - n^5 \\ m^3 &= n^4 - n^5 \end{aligned}$$

have a (unique) solution for integral n^4, n^5, n^6. This is the case provided $m^1 + m^2 + m^3$ is even. Thus there are four cases to consider: m^1, m^2 and m^3 are all even; exactly one of them is even. In each case, the distribution of values τm^i [which appears in (17)] is uniformly dense mod 1 in the limit $N^i \to \infty$. This is true also for the even and odd subsequences of (20) independently. From (17) we also see that the integers n^i, $i = 1, 2, 3$, are uniquely determined if the a^i are to fall inside the required range (18).

In summary, we have exhibited a one-one correspondence between: (1) lattice points $\mathbf{n}$ with the properties $P_\perp(\mathbf{n}) \in I(k^1, k^2, k^3)$, $P_\parallel(\mathbf{n}) \in \mathcal{R}(N^1, N^2, N^3)$ and (2) triplets of integers m^i with even sum. The m^i belong to independent sequences (20) and imply uniform distributions for each a^i in the limit $N^i \to \infty$. The independence of these one-dimensional distributions implies a uniform density in $I(k^1, k^2, k^3)$.

2. Fourier transform of the triacontahedron

An obvious approach to the evaluation of (16) is to decompose the integration region into 20 disjoint rhombohedra and add the contributions from each of these. Unfortunately, as this decomposition is not very symmetrical, this endeavor not only promises to be extremely tedious, but almost certainly would fail to deliver a formula wherein icosahedral symmetry was manifest. We therefore choose a different approach, beginning with the steps *prior* to (16).

We use the fact that the icosahedral quasilattice may be viewed as a filling of space with rhombohedra generated by basis vectors $e_\parallel^i$, $e_\parallel^j$, $e_\parallel^k$ (Kramer & Neri, 1984). If $r_\parallel(i, j, k)$ denotes the center-of-mass coordinate of the rhombohedron, then

$$S(g_\parallel) = \sum_{1 \le i < j < k \le 6} S_1(i, j, k) S_2(i, j, k),$$

where

$$\begin{aligned} S_1(i, j, k) &= \sum_{r_\parallel(i,j,k)} \exp\left[i g_\parallel \cdot r_\parallel(i, j, k) \right] \\ S_2(i, j, k) &= \sum_{\substack{\varepsilon_\alpha = \pm 1 \\ (\alpha = i, j, k)}} w(\varepsilon_i, \varepsilon_j, \varepsilon_k) \\ &\quad \times \exp\left[i g_\parallel \cdot \left(1/2 \sum_\alpha \varepsilon_\alpha e_\parallel^\alpha \right) \right]. \end{aligned} \tag{21}$$

The 'weight' $w(\varepsilon_i, \varepsilon_j, \varepsilon_k)$ in (21) represents the fraction of solid angle contributed by the rhombohedron with

edges $e_\parallel^i$, $e_\parallel^j$, $e_\parallel^k$ to the vertex

$$x_\parallel = r_\parallel(i,j,k) + \tfrac{1}{2}\sum_{\alpha=i,j,k} \varepsilon_\alpha e_\parallel^\alpha.$$

The six-dimensional analog of identity (12) allows us to replace $\exp(ig_\parallel \cdot x_\parallel)$ with $\exp(-ig_\perp \cdot x_\perp)$, where

$$x_\perp = r_\perp(i,j,k) + \tfrac{1}{2}\sum_{\alpha=i,j,k} \varepsilon_\alpha e_\perp^\alpha. \tag{22}$$

Clearly, $r_\perp(i,j,k)$ is identified with the center of mass of a rhombohedron in $X_\perp$.

Let us determine the distribution of points $r_\perp(i,j,k)$ inside $C_\perp(x_0)$, the triacontahedron. At the expense of introducing an irrelevant overall phase (depending on x_0), we assume the latter is centered on the origin:

$$C_\perp = \left\{ \sum_{i=1}^{6} x^i e_\perp^i \,\middle|\, |x^i| < \tfrac{1}{2}, \qquad i = 1,\ldots,6 \right\}.$$

If all eight vertices (22) lie inside $C_\perp$, then clearly

$$r_\perp(i,j,k) \in R_\perp(i,j,k),$$

where

$$R_\perp(i,j,k) = \left\{ \sum_{\beta=l,m,n} x^\beta e_\perp^\beta \,\middle|\, |x^\beta| < \tfrac{1}{2}, \qquad \beta = l,m,n \right\}$$

is a rhombohedron and l, m, n are just the complementary indices, *i.e.*

$$\{l,m,n\} = \{1,\ldots,6\} - \{i,j,k\}.$$

To evaluate S_1 we note that each rhombohedron with center of mass $r_\perp(i,j,k)$ can uniquely be associated with the vertex

$$r_\perp(i,j,k) - \tfrac{1}{2}\sum_{\alpha=i,j,k} e_\perp^\alpha.$$

Thus, we can apply the notion of a 'subpattern' (a rhombohedron) rooted at this vertex. If the quasilattice has N points, the distribution (in $C_\perp$) of these vertices is uniform with density $N/\mu(C_\perp)$ inside the region

$$R_\perp(i,j,k) - \sum_{\alpha=i,j,k} e_\perp^\alpha$$

and zero elsewhere. Consequently,

$$\begin{aligned} S_1(i,j,k) &= [N/\mu(C_\perp)] \int_{R_\perp(i,j,k)} e^{-ig_\perp \cdot r_\perp} \, \mathrm{d}^3 r_\perp \\ &= [N/\mu(C_\perp)] v_{lmn} \prod_{\beta=l,m,n} \sin z_\beta / z_\beta, \end{aligned}$$

where

$$z_\beta = \tfrac{1}{2} g_\perp \cdot e_\perp^\beta$$

and v_{lmn} is the volume of $R_\perp(i,j,k)$ given by (15).

We now turn to S_2, the sum over the vertices of the rhombohedron. Apart from orientation, the rhombohedron formed by the basis vectors $e_\perp^i$, $e_\perp^j$, $e_\perp^k$ may have one of two shapes called 'prolate' and 'oblate'. To help distinguish between these cases we introduce the quantities

$$\sigma_{ij} = \operatorname{sign}(e_\perp^i \cdot e_\perp^j), \qquad \sigma_{ijk} = \sigma_{ij}\sigma_{jk}\sigma_{ki}.$$

Owing to (14), there is an overall change in sign if we consider instead the analogous quantities defined for basis vectors in $X_\parallel$. With the above definition, the cases $\sigma_{ijk} = +1$ and $\sigma_{ijk} = -1$ correspond to prolate and oblate rhombohedra (in $X_\perp$), respectively.

For the prolate rhombohedron, the two corners on the axis of symmetry subtend the fraction 1/20 of the solid angle while the remaining six corners subtend the fraction 3/20. For the oblate rhombohedron the corresponding fractions are $2\times 7/20$ and $6\times 1/20$. In both cases the vertices on the axis of symmetry (relative to the center of mass) are given by

$$e_\perp^{ijk} = \pm\tfrac{1}{2}(\sigma_{ij} e_\perp^k + \sigma_{jk} e_\perp^i + \sigma_{ki} e_\perp^j).$$

Recalling that the weight $w(\varepsilon_i, \varepsilon_j, \varepsilon_k)$ represents the fraction of solid angle in $X_\parallel$, we find

for $\sigma_{ijk} = +1$:

$$\begin{aligned} S_2(i,j,k) = (1/20)\, 8 \prod_{\alpha=i,j,k} \cos z_\alpha \\ + (7/20 - 1/20)\, 2 \cos(\tfrac{1}{2} g_\perp \cdot e_\perp^{ijk}) \end{aligned}$$

for $\sigma_{ijk} = -1$:

$$\begin{aligned} S_2(i,j,k) = (3/20)\, 8 \prod_{\alpha=i,j,k} \cos z_\alpha \\ + (1/20 - 3/20)\, 2 \cos(\tfrac{1}{2} g_\perp \cdot e_\perp^{ijk}). \end{aligned}$$

With the help of σ_{ijk}, these can be combined into a single formula:

$$\begin{aligned} S_2(i,j,k) = (\tfrac{4}{5} - \tfrac{2}{5}\sigma_{ijk}) \prod_{\alpha=i,j,k} \cos z_\alpha \\ + (\tfrac{1}{5} + \tfrac{2}{5}\sigma_{ijk}) \cos(\sigma_{ij} z_k + \sigma_{jk} z_i + \sigma_{ki} z_j). \end{aligned} \tag{23}$$

References

BRUIJN, N. G. DE (1981). *Proc. K. Ned. Akad. Wet. Ser. A*, **43**, 39–52, 53–66.

COXETER, H. S. M. (1963). *Regular Polytopes*, p. 27. New York: Macmillan.

DUNEAU, M. & KATZ, A. (1985). *Phys. Rev. Lett.* **54**, 2688–2691.

KALUGIN, P. A., KITAEV, A. YU. & LEVITOV, L. C. (1985). *JETP Lett.* **41**, 119–121.

KRAMER, P. & NERI, R. (1984). *Acta Cryst.* A**40**, 580–587.

KUIPERS, L. & NIEDERREITER, H. (1974). *Uniform Distribution of Sequences*, ch. 1. New York: Wiley-Interscience.

LEVINE, D. & STEINHARDT, P. J. (1984). *Phys. Rev. Lett.* **53**, 2477–2480.

MACKAY, A. L. (1981). *Sov. Phys. Crystallogr.* **26**, 517–522.

MACKAY, A. L. (1982). *Physica (Utrecht)*, **114** A, 609–613.

PENROSE, R. (1974). *Bull. Inst. Math. Appl.* **10**, 266.

SHECHTMAN, D., BLECH, I., GRATIAS, D. & CAHN, J. W. (1984). *Phys. Rev. Lett.* **53**, 1951–1953.