Quasicrystals: The View from Les Houches

Marjorie Senechal and Jean Taylor

1. Introduction

Soon after the announcement of their discovery in 1984 [1], quasicrystals hit the headlines. Here was a substance—an alloy of aluminum and manganese—whose electron diffraction patterns exhibited clear and unmistakable icosahedral symmetry (a view along a five-fold axis is shown in Figure 1). A clear and unmistakable diffraction pattern of any sort is evidence of "long-range order": the diffraction pattern is a picture of a Fourier transform. Long-range order is usually synonymous with periodicity, and every periodic structure has a translation lattice. But a simple argument shows that five-fold rotational symmetry is incompatible with lattices in R^2 and R^3: every lattice has a minimum distance d between its points, but if two points at this distance are centers of five-fold rotation about parallel axes, the rotations will generate an orbit with smaller distances between them (Figure 2). By this chain of reasoning, it appeared that the impossible had occurred.

For the past five years, quasicrystals have been studied intensively by metallurgists, solid-state physicists, and mathematicians (few crystallographers have shown much interest in them). The problem has gradually been resolved into three more or less separate questions, not necessarily according to the field of the researcher:

Jean Taylor (left) and Marjorie Senechal

Marjorie Senechal received her Ph.D. from the Illinois Institute of Technology in 1965, where she studied analytic number theory with A. Sklar. She is a professor of mathematics at Smith College, and a member and occasionally director of its History of Science program. Prompted perhaps by her childhood fascination with patterns and weavings, her main research interest has been mathematical crystallography, a field that is a meeting ground for the geometry of numbers, tilings, lattices, polytopes, and discrete groups. She is currently writing a book on quasicrystals. Her nonprofessional interests include eclectic reading, hiking and cycling, and talking on the phone with her two grown daughters. The loom stands unused in a corner, but she hopes to weave again if she can ever find the time.

Jean Taylor was working on her Ph.D. thesis in physical chemistry at Berkeley until she audited a course in differential geometry taught by S. S. Chern. She eventually (1973) received her Ph.D. in mathematics from Princeton University and is now a professor at Rutgers University. Her main research interests concern crystalline minimal surfaces and other variational and growth problems involving anisotropic surface energy. Her other interests (besides quasicrystals) include mountain-climbing, skiing, scuba diving, wine-tasting, flying airplanes, playing with computers, indiscriminate reading, and almost any activity with her kids.

1. *Crystallography.* How are the atoms of real quasicrystals arranged in three-dimensional space?

2. *Physics.* What are the physical properties of substances with long-range order but no translational symmetry?

3. *Mathematics.* What kind of order is necessary and sufficient for a pattern of points to have a diffraction pattern with bright spots?

As Cahn and Taylor pointed out in 1985 [2], to answer Question 3 we must draw on a variety of techniques from many branches of mathematics, including tilting theory, almost periodic functions, generalized functions, Fourier analysis, algebraic number theory, ergodic theory and spectral measures, representations of GL(n), and symbolic dynamics and dynamical systems.

This article is a report on the current status of the problem. We began our discussions while attending a conference on *Number Theory and Physics,* at the Centre de Physique, Les Houches, France in March 1989. One of the foci of that conference was quasiperiodicity and quasicrystals, and during our ten days there we enjoyed extended discussions with a variety of observers and practitioners of this field. But we warn the reader that the view we present here may not be widely shared; in particular, Question 3 is usually not phrased in such generality (see Section 6). And like the view of the Mont Blanc massif from the conference center (Figure 3), the general outline and size of the problem is rather clear, but features that are prominent from our perspective may mask others, including the summit.

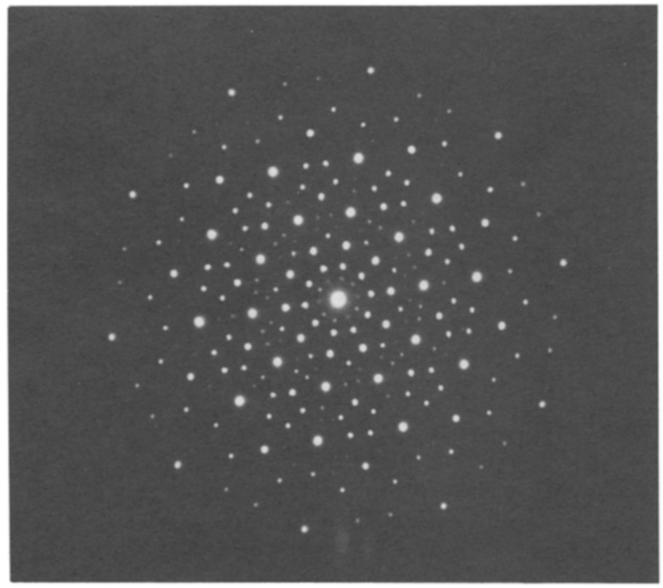

Figure 1. A diffraction pattern of an aluminum-manganese quasicrystal. Its five-fold rotational symmetry produced shock-waves in the world of solid-state science. Photograph courtesy of John Cahn.

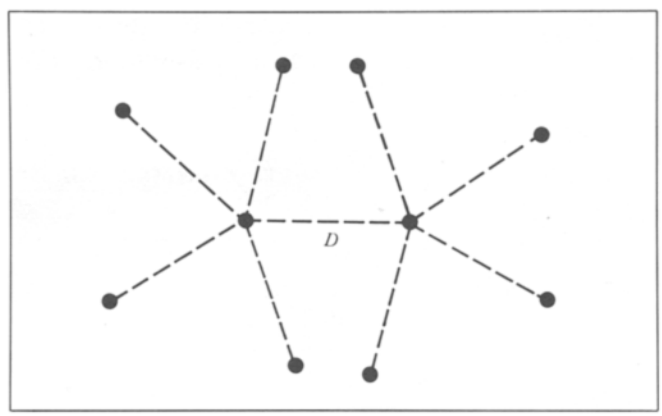

Figure 2. Five-fold symmetry is incompatible with periodicity, because it violates discreteness. Two five-fold centers at the minimum distance *d* generate points whose distance is less than *d*.

2. What Is a Crystal?

The discovery in 1912 that crystals diffract X-rays lent overwhelming experimental support to the hypothesis that crystalline structure is periodic. What could be a better example of Pierre Curie's banal but widely admired Principle of Symmetry: "When certain causes produce certain effects, the elements of symmetry in the causes ought to reappear in the effects produced"? Since then, the lattice has been taken as the definition of the crystalline state.

For the first year or two after their discovery, the question most hotly debated among solid-state scientists was: are quasicrystals crystals? By this was meant, can the structure of these alloys be interpreted within the framework of periodicity (for example, as a mosaic or intergrowth), or is it something truly new? Now, nearly five years later, quasicrystals of varying compositions (aluminum-lithium-copper, uranium-palladium-silicon, and many others) and high perfection have been grown in laboratories all over the world and have been analyzed in great detail, and the mosaic

Figure 3. The Mont Blanc massif, seen from the Centre de Physique, Les Houches, France. Photo by Pierrette Cassou-Nouges.

and intergrowth models have been discarded by essentially everyone but Linus Pauling [3]. It is clear that the question should be interpreted differently. To ask whether quasicrystals are crystals really means to ask what we mean by "crystal." We cannot see inside the solid state; we know it only through the images provided by diffraction, electron microscopy, and other modern techniques. It might be more appropriate to define a "crystal" to be a structure with sufficient long-range order to exhibit images with properties associated with those we call crystalline, such as a diffraction pattern with sharp spots.

A diffraction pattern for a material is essentially a two-dimensional slice of the square modulus of the Fourier transform of its density distribution; it faithfully records the amplitudes of the transform, but gives no direct record of the phases. Geometrically, we can think of a periodic crystal as an orbit of its symmetry group, which is an infinite discrete group with compact fundamental region. It can be shown that every orbit of such a group is a union of a finite number of congruent lattices. In the first approximation, we can take the density distribution of a periodic crystal to be a sum of weighted delta functions located at the nodes of each of these lattices (with the same weight for each point of a particular lattice). The Poisson summation formula implies that the Fourier transform of a lattice sum of deltas is again such a sum. Thus the diffraction pattern of a lattice is a lattice; it is in fact its dual lattice. The original lattices and their weights can be recovered from the diffraction pattern with the help of some techniques for recovering the "phase factor"; this is the experimental and theoretical framework for crystal structure analysis.

Finding a periodic framework on which to hang these structures can be likened to the pre-Keplerian problem in astronomy of trying to explain planetary orbits by decorating the circle with the right number and arrangement of epicycles.

However, the lattice hypothesis is not without its problems. There are crystals with extremely large repeat units, with thousands of atoms in the unit cell. There are crystals that are more or less random stackings of two-dimensional periodic structures. There are crystals in which the lattice is disturbed by a modulation. Finding a periodic framework on which to hang these structures can be likened to the pre-Keplerian problem in astronomy of trying to explain planetary orbits by decorating the circle with the right number and arrangement of epicycles. No number or arrangement will be correct for the quasicrystals! The quasicrystal phenomenon shows us that a diffraction pattern can theoretically show sharp spots even if there is a single nonperiodic but well-defined geometrical pattern that gives rise to it. And although it is widely assumed that the crystal lattice is a global consequence of the play of local interatomic forces, from the standpoint of physics or mathematics this is an open problem. Indeed, Miekisz and Radin have shown that generically one would expect local forces to generate nonperiodic structures [4]. In fact, one can even find crystals almost arbitrarily close to "ideal quasicrystals," in the same way that irrational numbers can be approximated by rational numbers.

Thus the deeper question is: what local ordering properties are necessary and sufficient to produce orderly images?

Two minimal properties that we might require of a locally ordered point set $L \subset R^n$ are discreteness and relative density: there is a minimum distance d between any pair of points of L, and a number $\delta > 0$ such that every sphere of radius δ contains at least one point of L; such an L is sometimes called a *Delone system.* (Incidentally, Delone's name is sometimes spelled Delaunay, reflecting the fact that his forebears went to Russia with Napoleon and stayed on.) A finite region of a Delone system with no additional structure roughly describes the centers of the atoms in a monatomic gas in a closed container. Increasing the structure increases the order. Sufficient local order implies periodicity: Delone and his colleagues proved [5] that there is a number $k = k(\delta,n)$, where n is the dimension of the space in which L lies, such that if the sets $\{x \in R^n: |x - u| \leq k\} \cap L$ are congruent for each $u \in L$, then L is an orbit of a crystallographic group.

The patterns we are interested in have order somewhere between amorphous and periodic. The question is, what intermediate conditions are necessary and sufficient to ensure that L can produce a diffraction pattern with "bright spots"? For example, one condition might be *local isomorphism:* every finite configuration of L occurs in every bounded region of sufficient size. But although local isomorphism is present in all the examples that we know about, there is no proof that it is either necessary or sufficient. Other local ordering conditions can of course be proposed, but not much is known about their effect either. The question remains open.

One obvious difficulty is that "bright spot" is not well defined. We can simplify matters by defining it to mean that there are Dirac deltas in the Fourier spectrum, weighted so that some peaks appear isolated. Then there are two cases to consider: either the entire Fourier spectrum is a set of deltas (possibly dense), or else the spectrum also contains a continuous component. But from the experimental point of view a bright spot need not represent a "real" delta; it might be due to features of the transform that closely approximate delta functions. Point sets with this property are of theoretical as well as experimental interest (see Section 4).

We can formulate these conditions more precisely. Any Delone system D has a density distribution that is a countable infinite sum of weighted Dirac deltas on the points of D; we can assume as a first approximation that all the weights are equal to 1. Then the distribution $\rho(x)$ can be written $\Sigma_{v \in D}\, \delta(x - v)$, where $x \in R^n$; a distribution of this form is sometimes called a *Dirac comb*. We are looking for Dirac combs whose Fourier transforms $\hat{\rho}(t) = \Sigma_{v \in D} \exp(2\pi i t \cdot v)$ are closely related to Dirac combs, where by "closely related" we mean one of the following:

(a) The Fourier transform is precisely a Dirac comb; such a comb is also called a Poisson comb. (An important special case is when the frequencies v at which the delta functions of the Fourier transform occur have a finite basis over the integers; the original density is then said to be *quasiperiodic*.)

(b) The Fourier transform contains a Dirac comb together with a continuous part.

(c) The Fourier transform "looks like" it contains a Dirac comb but does not in fact do so; this can happen when the spectrum has a singular continuous component.

Characterizing the order properties of Dirac combs satisfying (a), (b), or (c), and classifying these combs, is a central problem of quasicrystallography, and involves all of the branches of mathematics listed above. In the absence of a complete answer to our question, we study examples. The two classes of combs that have been studied in most detail are those obtained by projection, and one-dimensional combs. We will also discuss some of the relations between combs and tilings; some interesting recent work is discussed in Section 5.

3. Point Sets Obtained by Projection

Three years before the discovery of quasicrystals, the English crystallographer Alan Mackay [6], long an advocate for a more general crystallography, devised an ingenious experiment. He arranged for an optical diffraction mask to be constructed whose holes were located at the vertices of a tiling by Penrose rhombs (Figure 4). These tilings, which are constructed by juxtaposing the rhombs according to strict matching rules, are nonperiodic. Yet they aren't "disordered": one can discern a great deal of local order. Local configurations with 5-fold symmetry not only occur, they occur all over the place. Indeed, the pattern of vertices has the local isomorphism property discussed above. Moreover, the tilings are self-similar. (These and other properties of the Penrose tilings will be discussed in more detail in Section 5.) As Mackay suspected, the diffraction pattern obtained with this mask was clear and sharp, almost crystalline. And it had the crystallographically forbidden five-fold symmetry.

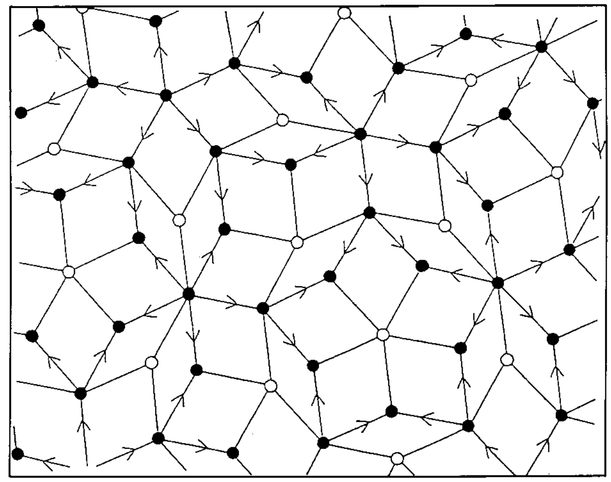

Figure 4. Part of a Penrose tiling by rhombs. Vertex colors and edge arrows must be matched.

De Bruijn's construction. Since the quasicrystal in question, i.e., the set of the vertices of the Penrose tiles, is not a lattice, how can we explain Mackay's experimental results? The necessary insight was supplied by N. G. de Bruijn, in a remarkable set of papers published in 1982 [7]. In these papers, de Bruijn gave a global method for constructing the Penrose tilings that allows us to index the vertices of the rhombs with five integer coordinates $(x_0, \ldots, x_4)$. Thus the vertices can be identified with a subset S of the points of the integer lattice in R^5.

Moreover, de Bruijn showed that

$$1 \leq \sum_{k=0}^{4} x_i \leq 4 \text{ for all } \vec{x} = (x_0, \ldots, x_4) \in S. \quad (1)$$

Since this sum is also the scalar product $(x_0, \ldots, x_4) \cdot (1, \ldots, 1)$, the points of S lie in a region $M \subset R^5$ which projects to a bounded interval on the line containing $\vec{m} = (1,1,1,1,1)$; note that this vector is the body diagonal of the unit 5-cube γ_5. The vertices of the Penrose rhombs are the projections of S onto a plane Π, one of the two invariant planes of the five-fold rotation about that diagonal, which cyclically permutes the five coordinate axes. Both of these planes are irrational: their intersections with the lattice are just $\{0\}$. The tile vertices are integral linear combinations of the projections of the five orthonormal unit coordinate vectors of R^5 onto Π. Not all vectors satisfying (1) are in S; S is the set of points M' of M whose projection onto $\Pi^{\perp}$ lies in the projection of γ_5 onto that subspace. (Any projection of an n-cube is a zonohedron; in this case it is a rhombic icosahedron.)

Katz and Duneau [8], among others, have shown that the projection formalism greatly facilitates the

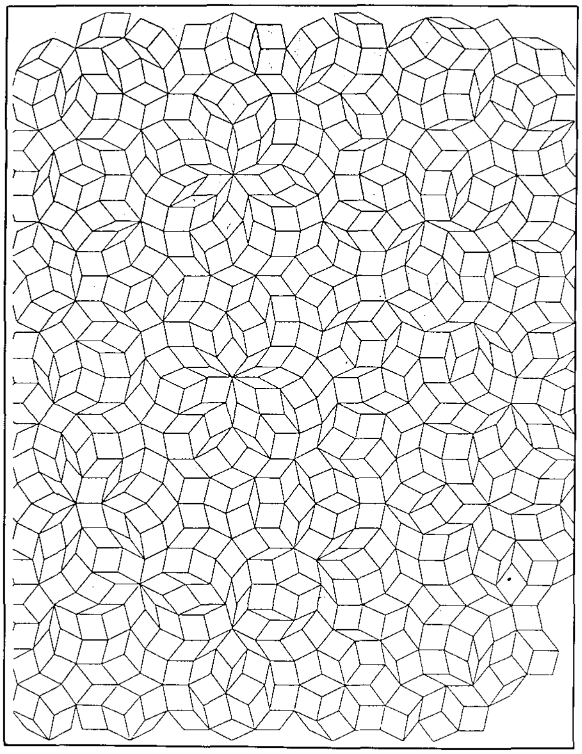

Figure 5. A plane tiling by three kinds of rhombs, projected from R^7. Courtesy of André Katz.

computation of Fourier transforms. The density function of the set S is the product of the density function of the integer lattice in R^5 and the characteristic function of M'. Thus, since the Fourier transform of a product of two functions is the convolution of the Fourier transforms of the individual functions, and since the Fourier transform of the projection of the density function of S is the restriction of the Fourier transform of that function to Π, we can compute the diffraction pattern of the Penrose vertices. (That all of this can be made rigorous has been shown, by rather different arguments, by de Bruijn [9] and by Porter [10].) The Fourier transform is a countable sum of delta functions at a dense set of points in the plane; thus the set of vertices is a Poisson comb. Although the delta functions are dense, we see bright spots in the diffraction pattern, because the amplitude of the transform attains local maxima at isolated points, and at most other points is very small.

The general case. The Penrose tilings are of course very special. To what extent does the property of being a Poisson comb depend on their remarkable properties? Curiously, this dependence is not very strong. For example, while it is easy to construct Poisson combs by projection, as far as we know most of them are not self-similar. (If we translate M in $\Pi^\perp$, the projected pattern will include local vertex configurations that are forbidden by the matching rules of the Penrose tiles.) Or, we can carry out the analogous construction in R^n, producing plane point sets that are the vertices of tilings with local n-fold symmetry for which no matching rules are known (see Figure 5). All of these patterns have the local isomorphism property, however.

More generally, let Λ be a lattice in R^n, and let Π_k be any irrational k-dimensional subspace of R^n (again, irrational means that $\Pi_k \cap \Lambda = \{0\}$). First, let us see under what conditions we can obtain a Delone system in Π_k by projection. Since Π_k is irrational, the orthogonal projection of all of Λ onto Π_k will be nonperiodic, but it will also be dense. We need to find a subset S of Λ that projects to a discrete set (relative density is guaranteed by the fact that Λ is a lattice). We know that there is a minimum distance d between points of Λ: if $\vec{x}$ and $\vec{y}$ are two vectors of Λ, then $|\vec{x} - \vec{y}| \geq d$. We can decompose $\vec{w} = \vec{x} - \vec{y}$ into its Π_k and $\Pi_k^\perp$ components w_k and $w_k^\perp$. If we insist that $|w_k^\perp| < d - \epsilon$ for some $\epsilon > 0$, then $|w_k| \geq d$. This means that we can obtain a discrete set of points in Π_k by requiring that the image of the set $S \subset R^n$ under projection to $\Pi_k^\perp$ lie in an appropriately chosen compact set $T \subset \Pi_k^\perp$; T is sometimes called the *window* for the projection. Thus S lies in the cylinder $M = T \oplus \Pi_k \subset R^n$. The computation of the Fourier transform then proceeds as in the examples above. The projected set always turns out to be a Poisson comb.

There are many variations on the projection theme. The window need not lie in the orthogonal complement of Π_k; the vector space need not be Euclidean. One technique used quite extensively at the moment is to try to replace Λ by a periodic set of connected surfaces in R^n, and to consider the intersection of Π_k with these surfaces. (Again, de Bruijn has provided a firm mathematical basis for much of this, in a different language [11].) It is true that if one has a Poisson comb and its Fourier transform's delta functions have a finite basis, and if one knows the full complex amplitudes of these deltas, then it is possible to reconstruct a periodic density in R^n and a plane Π_k such that the restriction of the density in R^n to Π_k will give the delta functions of the density in R^k. However, it is not at all obvious that there are densities in R^n that consist of "surfaces" of any particular smoothness. Also, although using arbitrary surfaces, rather than those from projecting a lattice, gives a broader class of Poisson combs, it does not always give a noticeably broader class of diffraction patterns, because these combs may differ from the lattice-projection ones only in their intensities and phases, as de Bruijn has noted [11].

A word about symmetry. The most striking thing about the diffraction pattern of the Penrose vertices is its five-fold rotational symmetry; quasicrystals might never have been noticed if this symmetry had not been observed. Indeed, successful crystal structure determination depends on finding directions of high symmetry so that bright spots will appear in the diffraction pattern.

The symmetry we observe in the Fourier transform of a projected pattern depends on the symmetry group G of the lattice Λ and on the choice of Π_k. G is a semidirect product of Z^n and a finite subgroup $P \subset O(n)$, where P is the stabilizer of $0 \in \Lambda$. If Π_k is invariant under P, or under a subgroup of P, the Fourier transform will reflect this. This leads us to the important and interesting problem of determining which lattices in R^n have invariant subspaces of whatever dimension, and how crystallographic groups built on these lattices act on those subspaces. In short, the projection method has opened an interesting chapter in n-dimensional crystallography. To date, those lattices for which G is or contains the icosahedral group have been studied in the most detail (see, e.g., [12]), because they arise in the theory of the three-dimensional Penrose tiles (see Section 5) and in the interpretation of diffraction patterns of real quasicrystals.

But from our point of view, it is the bright spots that are fundamental, not symmetry *per se.* Since bright spots are theoretically present in every projected pattern, we know that their occurrence is not dependent on rotational symmetry. Indeed, it seems that rotational symmetry has nothing *a priori* to do with our problem, except that when we find noncrystallographic rotational symmetry in a diffraction pattern, we know that it was produced by a nonperiodic Dirac comb. On the other hand, bright spots in a diffraction pattern indicate some sort of long-range order or generalized symmetry. This brings us back to the questions raised in Section 2.

4. Order on the Line

The one-dimensional case is the most tractable; here we find examples of all three types of ordering for nonperiodic point sets. We will describe a few of them.

Sequences with average lattices. The standard example of a one-dimensional quasicrystal is the "Fibonacci" sequence of points

$$u_n = n + (\tau - 1)[n/\tau], \tag{2}$$

where $\tau = (1 + \sqrt{5})/2$ is the golden number of classical and modern fame, and $[x]$ is the greatest integer function. (We will explain the relation of this sequence to the classical Fibonacci sequence below.) This sequence can be obtained by projection from R^2 to a line Π with slope $1/\tau$. Let us consider the more general case in which the line has slope α, where α is an irrational number. Using as our window the projection of a unit square of the integer lattice Λ onto $\Pi^\perp$, the cylinder M is the strip bounded by the lines $y = \alpha x$ and $y = \alpha x + \alpha + 1$ (Figure 6) and $M' = M \cap \Lambda$. The points in this strip have coordinates

$$(x,y) = ([n/(\alpha + 1)], n - [n/(\alpha + 1)]),$$

and project onto the points $(x + \alpha y)\vec{u}$, where $\vec{u}$ is the vector $(1,\alpha)/(1 + \alpha^2)$ along Π. Thus the projected points form a sequence

$$p_n = \alpha n - (\alpha - 1)\left[\frac{n}{\alpha + 1}\right], \tag{3}$$

and $p_{n+1} - p_n = 1$ or α. When $\alpha = 1/\tau$, we obtain the sequence above (if we multiply everything by $1/\tau$). The methods of the preceding section can be used to show that all of these sequences are Poisson combs.

The sequences obtained in this way are often called "one-dimensional quasicrystals." But they do not really illustrate the quasicrystal phenomenon, because in fact these sequences are one-dimensional modulated lattices. Modulated crystals were known long before the quasicrystals were discovered, and have been intensively studied for many years.

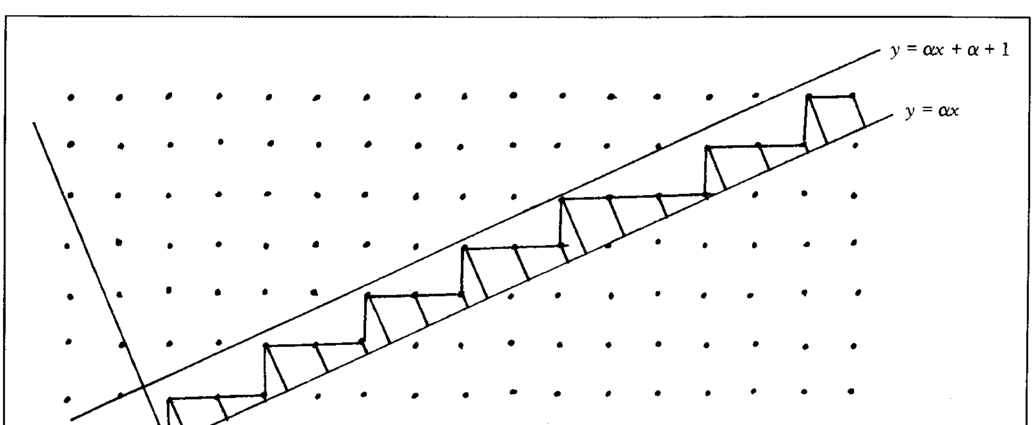

Figure 6. A tiling of the line obtained by projection from R^2. All tilings of this type have average lattices. (Adapted from Ref. [8].)

In what sense are these sequences modulated lattices? Using the equality $[x] = x - \{x\}$, where $\{x\}$ is the fractional part of x, we have

$$p_n = \alpha n - (\alpha - 1)\frac{n}{\alpha + 1} + (\alpha - 1)\left\{\frac{n}{\alpha + 1}\right\}$$
$$= \frac{\alpha^2 + 1}{\alpha + 1}n + (\alpha - 1)\left\{\frac{n}{\alpha + 1}\right\}, \quad (4)$$

or in the case of the Fibonacci sequence (2), $u_n = (2 - 1/\tau)n + (\tau - 1)\{n/\tau\}$. Thus we see that $\{p_n\}$ is built upon the one-dimensional lattice of points of the form $n(\alpha^2 + 1)/(\alpha + 1)$ for $n \in Z$, deviating from it by an amount which is at most $|\alpha - 1|$.

This property, of having a limiting average spacing and a bounded modulation away from the lattice with this spacing, is called having an "average lattice."

One can compute the Fourier transforms of sequences of type (4), or indeed of any sequence of the form

$$v_n = \alpha n + \beta\{\gamma n\} \quad (5)$$

in a straightforward manner [13]; the sequences are always Poisson combs.

In fact, any sequence whose elements are of the form $\alpha n + g(n)$, where $g(n)$ is periodic or almost periodic, is also a Poisson comb. For appropriate choices of parameters, these sequences will be nonperiodic; it is not known whether they can be obtained by projection.

It is not known which of the sets obtained by projection onto subspaces of dimension greater than 1, considered in Section 2, have average lattices. However, some of them do. Duneau and Oguey have recently shown [14] that the set of vertices of a tiling obtained by projection from R^8 to R^2 has an average lattice; the construction applies to certain other tilings as well.

Sequences obtained by substitution. If we interpret the letters a and b to be line segments of lengths τ and 1, respectively, then the sequence u_n discussed above is the limit

$$\lim_{n\to\infty} T^n(w_0),$$

where w_0 is a word of the two-letter alphabet $\{a,b\}$ and T is the map, or substitution rule,

$$T(a) = ab, \quad T(b) = a.$$

When $w_0 = b$, then the length of the word $T^n(w_0)$ is F_{n+1}, where F_n is the n^{th} Fibonacci number ($F_0 = 0$, $F_1 = 1$, $F_{n+1} = F_n + F_{n-1}$); u_n is sometimes called a Fibonacci sequence, although the classical Fibonacci sequence is $T^n(a)$. It is not known which of the more general sequences v_n discussed above can be produced by substitution rules, but obviously, more general substitution rules can be used to produce sequences on the line.

What can be said about the Fourier transforms of substitution sequences? To prove that a Dirac comb is a Poisson comb requires knowing the whole Fourier transform; the only way this has been accomplished so far is to show that the density ρ is the sum of delta functions on a lattice modulated by a periodic or almost periodic function, or that ρ is obtained by slicing through a periodic density in a higher-dimensional space. On the other hand, one can show that a density has property (b) as follows: The density $\rho(x) = \Sigma_{n=0,1,\ldots}\, \delta(x - v_n)$ has Fourier transform $\hat{\rho}(q) = \Sigma_n \exp(2\pi iq \cdot v_n)$. For any frequency q, the sequence of partial sums $\{\Sigma_{n=0}^{N} \exp(2\pi iq \cdot v_n)\}$ is bounded by $N + 1$. If we can show that for some q the sequence grows like cN for some positive c, then asymptotically the sum is a Dirac delta. It is possible to use this technique for some substitution sequences.

Any composition rule T acting on an alphabet of n letters can be represented by an $n \times n$ matrix M with nonnegative integer entries. If there is a $k \in Z$ such that all the entries of M^k are positive, then the Perron-Frobenius theorem tells us that M has a simple eigenvalue θ that is greater in absolute value than all the others. Bombieri and Taylor [13] showed that if $|\theta| > 1$, while all its conjugates have modulus less than one (i.e., if θ is a Pisot-Vijayaraghavan, or P-V, number) then the Fourier transform of the sequence can be computed as a limit of the Fourier transforms of the words $T^n(w_0)$. The transform contains a Dirac comb, because there are frequencies (forming a dense set) for which the sequence of transforms grows like N. In fact, all of these sequences lie in sets that can be obtained by projection.

Every substitution T on a finite alphabet gives rise to a topological dynamical system. By assigning appropriate lengths to the letters of the alphabet, a fixed point of a sequence $T^n(w_0)$ can be interpreted as the list of the successive differences of an increasing sequence of real numbers, and we can study the order properties of such sequences. The dynamical systems associated with substitutions of constant length, and their spectra, have been studied by Queffélec [15]. (Note, however, that the Fibonacci sequence is *not* of constant length.)

Other one-dimensional Poisson combs. Aubry, Godrèche, and Luck [18] studied a family of sequences that, for some choices of parameters, appear to be Dirac combs of type (c).

Let Δ be a subinterval of (0,1) and ω be any positive real number. Consider the sequence w_n of 0's and 1's obtained by setting $w_n = 0$ if $\{n\omega\} \in \Delta$ and $w_n = 1$ otherwise. There are two ways to build a sequence of points on the line from the sequence w_n. One can start with a one-dimensional lattice whose nodes are located at the points $n\omega$, $n \in Z$ and then omit those nodes for which the corresponding w_n is equal to 0. In this way we obtain a lattice with vacancies, which can be shown to be a Poisson comb. Kesten's theorem [17] asserts that this sequence has an average lattice, in the sense defined above, if and only if $\Delta \equiv r\omega$ mod(1) for some $r \in Z$. Thus there exist Poisson combs with no average lattice! The second way to build a sequence is to choose two unequal lengths l_1 and l_2 and let $u_0 = 0$, $u_{n+1} - u_n = l_1 + (l_2 - l_1)w_n$. In this case, it may happen that the Fourier transform has a singular continuous component, i.e., the sequence u_n is a Dirac comb of type (c). As far as we know, property (c) has never been completely established for any example. However, there are cases [18] where the possibility of Dirac peaks can be eliminated using the procedure appropriate to case (b), and the possibility of the transform being absolutely continuous can be essentially eliminated by numerical calculation. The spectrum should therefore contain a singular continuous part; numerical calculations then show it "looks like" a Dirac comb.

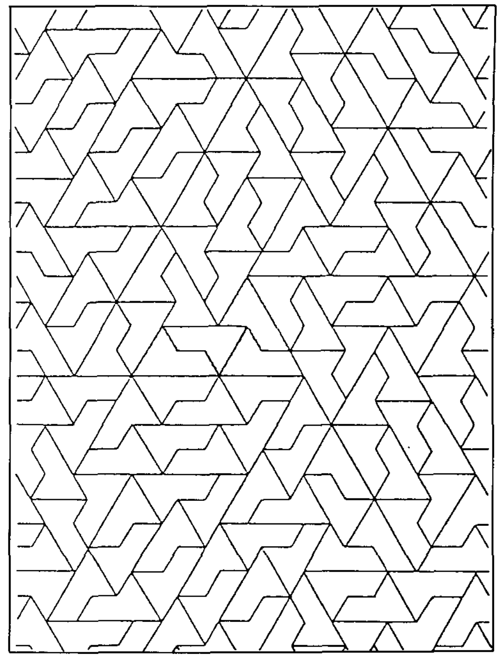

Figure 7. A self-similar tiling for which no matching rules are known to exist. (From Ref. [20].)

5. The Tiling Connection

Crystallographers have used tilings as models for crystal structures for centuries. For example, the lattice can be regarded as a framework for the partition of space into congruent parallelepidal tiles or "unit cells." These fictitious boxes in turn contain congruent, real, atomic configurations.

The diffraction patterns of the first quasicrystals looked remarkably like the one obtained earlier by Mackay. Thus it was natural to ask whether the three-dimensional analogue of the Penrose tilings might be a model for quasicrystals, with the two kinds of tiles playing roles analogous to the unit cells. Further experimental work has shown that this is not the case (see, e.g., [19]). In any case, the tiles in a nonperiodic tiling are not analogous to the unit cells of periodic patterns, although it is frequently asserted that they are. There are infinitely many ways to choose the shape and position of the unit cell for a periodic crystal, all equally valid from the abstract point of view. In contrast, in the few cases in which nonlattice point sets can be associated with tilings by copies of one or a few shapes, the choice of cells is usually unique, and it is by no means clear what the relation between the transforms is when masses are placed at vertices or in the tile interiors.

In fact, it is not clear what aspects of a real structure the tiles in a nonperiodic tiling might represent. Like the Big Dipper and other stellar constellations that one learns to identify as a child, the tiles sometimes appear to be highly artificial from a physical point of view, even when they are convex. For example, the minimum distance between vertices in a tiling by Penrose rhombs is the short diagonal of a thin rhomb; in a reasonable structure model one would expect nearest neighbors to be linked in some way.

Still, the possible connection between tilings and quasicrystal structure continues to be studied, partly because the tilings help us to visualize some kinds of nonperiodic order (this is why four of our eight illustrations are tilings.). It is easy to produce nonperiodic tilings by the projection method. In the special case where T is the projection of the unit n-cube γ_n, the projected points are the vertices of nonoverlapping projections of the k-dimensional faces of γ_n. For suitably chosen subspaces, the number of distinct tile shapes (prototiles) will be small ($O(n)$). Thus we can construct many interesting examples.

What do nonperiodic tilings have to teach us? The Penrose tilings have three important properties:

(1) they have matching rules that force nonperiodicity,

(2) they can be obtained by a substitution process and they are self-similar,

(3) they have strong local order (in fact they are quasiperiodic).

Surprisingly, it appears now that these properties are independent to some extent. There are substitution-produced tilings with matching rules that are not self-similar (several examples are shown in [20]). Figure 7 shows a tiling that is self-similar but for which no matching rules seem to exist; recently it has been shown that this tiling is not quasiperiodic (see below).

Tilings built with the tiles shown in Figure 8, using matching rules, are quasiperiodic but no substitution rule has been found for them.

Matching rules. Why are matching rules of interest in the study of quasicrystals? Evidently they are not needed in order for a tiling to be a Poisson comb. Their importance lies instead in our feeling about what features a good model should have. The projection method says nothing about how the quasicrystal grows—why the atoms order themselves in such a pattern. Some sort of local forcing rules would seem to be an important part of a good model for quasicrystals, since they are an analogue of the local bonding rules that presumably determine the structure.

The matching rules discovered by Penrose and by Ammann ([20]) were found by trial and error. Is there a more systematic way to do this? De Bruijn showed that his indexing system for the Penrose vertices leads to an unambiguous reconstruction of the Penrose rules, but his arguments do not apply if the set M is translated in R^5. Neither has it proved possible to apply it to any of the other plane tilings projected from R^n. This does not mean that no matching rules exist in these cases. For example, Ammann has found matching rules for certain tilings of the plane by squares and rhombs, projected from R^8 (again, see [20]). But as de Bruijn points out [21], although Ammann's rules are expressed locally, the property of an unmarked tiling to be Ammann-markable is not a local property.

It seems to us no more appropriate to define quasicrystals at this stage of our knowledge than to cling to the definition of a crystal as a periodic structure.

More recently, some progress has been made. Using homologous arguments, Katz has developed a method for decorating the tiles of certain projected tilings [22]. He applied it to the "three-dimensional Penrose tiles," thus proving that these tiles can be equipped with matching rules that force nonperiodicity (Figure 8). However, the construction is not a simple one: when decorations are taken into account the two rhombohedra fall into twenty-two classes.

Recently, Danzer has announced the discovery of a set of four marked tetrahedra [23] that tile R^3 only nonperiodically. Although the method by which he found them appears to be less systematic than Katz's, it is of interest because the number of prototiles is small.

Self-similarity. The self-similarity of the Penrose tilings is one of their most remarkable features. But until very recently self-similar tilings have been almost as hard to find as matching rules. In the first place, to be self-similar, a tiling must be a geometric realization of a "fixed point" of a substitution map. Any primitive matrix defines a substitution map, but we do not know of any theory that tells us which substitutions can be realized as tilings. Even when such a realization exists, the tiling need not be self-similar in the sense that the larger configurations into which the tiles are grouped by the action of the substitution map are geometrically similar to the original tiles. Conversely, given a tiling (such as that in Figure 5) it may be very difficult if not impossible to determine whether it is invariant under some substitution map T. Recently, Thurston has developed a method for associating self-similar tilings with fractal tile boundaries to a class of algebraic integers [24]. Substitution rules are implicit in the method, but it is not yet clear to us how to extract them.

Nevertheless, tilings invariant under primitive substitution maps are of interest in our context because they necessarily have the local isomorphism property. Moreover, we can sometimes use the substitution map to prove that a tiling is nonperiodic, a property that may not be obvious. Notice, for example, that the use of matching rules does not guarantee that a tiling is nonperiodic; some other argument must be invoked.

Two different arguments can be used to establish the nonperiodicity of a tiling with the substitution property. First, if the grouping of tiles into larger ones is *unique*, the tiling has a hierarchical structure that must be preserved by any translation. But this is impossible, since repeated iteration of this grouping implies that at some hierarchical level the inradius of the tiles will be larger than any specified translation length.

The other argument might be called a "ratio test" for nonperiodicity. It involves the eigenvectors of the substitution map. Let T be any primitive, integer $n \times n$ matrix, and let $\{a_1, \ldots, a_n\}$ be any finite alphabet. A word w of this alphabet contains x_i copies of the letter a_i. We can think of $\vec{x} = (x_1, \ldots, x_n)$ as a vector of the integer lattice in "configuration space." Then $\vec{x}T$ is another vector in this space; its components are the numbers of copies of each of the letters after one application of T. The components of a *left* eigenvector corresponding to its leading eigenvalue θ are the *relative* numbers of the different letters in the infinite word produced by iterating T. We can now state the ratio test. If T has an eigenvalue θ which is a P-V number, then for any initial configuration vector $\vec{x}_0$, the sequence $\theta^{-n}\vec{x}_0T^n$ will converge to a left eigenvector of θ. If the components of this eigenvector have irrational ratios, then the tiling will be nonperiodic, since in a periodic tiling the relative numbers of kinds of prototiles is given by the numbers in single repeat unit.

If T acts on a tiling, then the prototiles of the tiling play the role of the letters of an alphabet. We assume that they are arranged in such a way that each application of T effects a grouping of the tiles into larger tiles. These tiles need not be similar to the original ones, but if they are, then the relative volumes of the n prototiles after each application of T are the components of a *right* eigenvector of θ. This gives us a way to decide whether a tiling produced by substitution is self-similar; the Penrose tiles pass the test.

Which substitution-invariant tilings produce diffraction patterns with bright spots? There is no definitive answer yet. We have seen that if the tiling can be obtained by projection, then its set of vertices is a Poisson comb. Recently Godrèche and Luck [25] have extended the Bombieri-Taylor method for computing the Fourier transforms of substitution sequences to tilings of the plane by assigning masses to the tiles themselves and expanding the definition of T to take into account the geometry of configurations as well as the numbers of tiles in them. They then showed that Fourier transforms of this density distribution contain Dirac combs even when the matching rules are relaxed. During the Les Houches conference, Godrèche succeeded in showing that the tiling of Figure 7 fits into case (b) (but the possibility that the spectrum also contains a continuous component has not been ruled out). It is especially interesting that in this case there is no finite basis for the frequencies of the delta functions of the Fourier transform, so that the tiling is not quasiperiodic [25].

Local order. The local ordering properties of the Penrose tiles are discussed in [20], so we will not go into detail here. They include local isomorphism, and the fact that the number of different configurations within any finite radius is bounded and grows slowly as the radius increases. These properties hold for all projected and substitution tilings. But it remains an open question to what extent these properties, independently of projection and substitution, can account for the tilings' Fourier transforms.

6. A Word about Definitions

We mentioned at the beginning that we have posed Question 3 more generally than is usually the case.

We did not mention, but many readers will have observed, that we have offered no definition of "quasicrystal." In fact, most other writers define quasicrystals to be projected (or sliced) structures. There may be some justification for this. As we have shown, the projection/slicing method does produce an extremely large class of Poisson combs. Moreover, the models based on this approach are in very good agreement with experiment. But then, experimentally, it may be impossible to distinguish Poisson combs

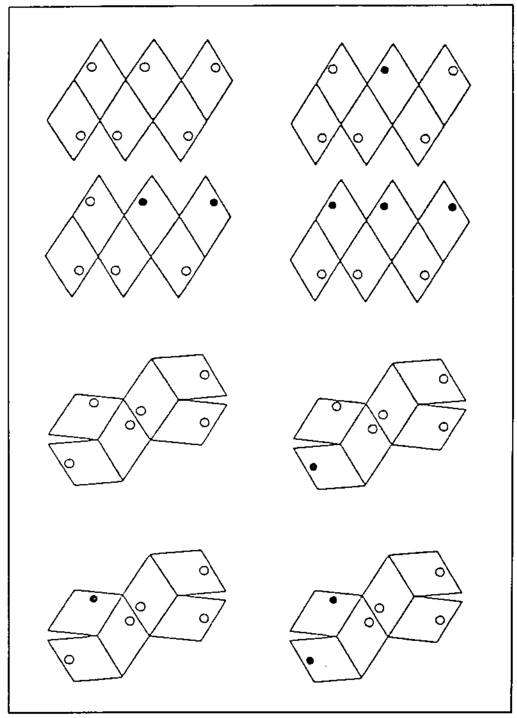

Figure 8. The three-dimensional analogue of the Penrose rhombs are two rhombohedra. When decorated with matching rules according to Katz's scheme, the rhombohedra fall into twenty-two classes. Nets for eight of the rhombohedra are shown here; the others can be generated from this set (see Ref. [22]).

from the other two cases discussed in Section 2. It seems to us no more appropriate to define quasicrystals at this stage of our knowledge than to cling to the definition of a crystal as a periodic structure.

Question 3 is nontrivial mathematically, and it is also nontrivial philosophically. The high-dimensional formalism is only a stop-gap to be used until we understand how quasicrystals grow. String theory notwithstanding, it is reasonable to assume that real quasicrystals, like real periodic ones, grow in R^3, not in R^n. We need a theory that explains how the patterns that we are interested in can be generated at the local level; it is not clear to what extent the deterministic models we have described are physically meaningful. Modeling growth may require a combination of matching rules, modulations, understanding "the sociological behavior of large groups of atoms" [21], and possibly other ideas. It is too early to know what class of patterns will achieve this. In our view, the definition of quasicrystal should be left open until the fundamental questions have been answered.

Acknowledgments: We would like to thank H. S. M. Coxeter, N. G. de Bruijn, and C. Godrèche for helpful comments on this article. Also, the second author would like to acknowledge the support of NSF and AFOSR.

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The essential point of the positivist theory is that there is no other source of knowledge except the straight and short way of perception through the senses. Positivism always holds strictly to that. Now, the two sentences: (1) *there is a real outer world which exists independently of our act of knowing* and (2) *the real outer world is not directly knowable* form together the cardinal hinge on which the whole structure of physical science turns. And yet there is a certain degree of contradiction between those two sentences. This fact discloses the presence of the irrational, or mystic, element which adheres to physical science as to every other branch of human knowledge. The effect of this is that a science is never in a position completely and exhaustively to solve the problem it has to face. We must accept that as a hard and fast, irrefutable fact, and this fact cannot be removed by a theory which restricts the scope of science at its very start. Therefore, we see the task of science arising before us as an incessant struggle toward a goal which will never be reached, because by its very nature, it is unreachable. It is of a metaphysical character, and, as such, is always again and again beyond our achievement.

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