Quasicrystals and geometry

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Marjorie Senechal
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Penrose tilings (Fig. 1) are beautiful. They also suggest significant new mathematics, so it is about time someone wrote a book about them which is readable (in fact, eminently readable) by mathematicians.

The book, *Quasicrystals and geometry*, by Marjorie Senechal, has an even broader goal: to present certain developments in crystallography from the past decade. The developments were generated in the wake of two profound discoveries. The first was the mathematics discovery [1] in 1966 of aperiodic tilings, the origin of Penrose's 1977 examples. The more recent one was the physics discovery [8] in 1984 of quasicrystals. These two discoveries have led to a large volume of interdisciplinary research—among the fields of crystallography, physics, and mathematics, and also between subfields of mathematics, especially discrete geometry and ergodic theory. The interaction of the viewpoints of the different fields has been enormously beneficial to the mathematics which is emerging. We begin with an overview of the terrain, strongly emphasizing those aspects of relevance to mathematics.

Quasicrystals are solids with unexpected properties. The characteristics referred to in this review are all related to properties of the "diffraction patterns" obtained by illuminating quasicrystals by parallel beams of electrons or X-rays. There is no need for us to delve into the details of diffraction, but it will be useful to note the following. When producing diffraction patterns from solid matter assumed fixed in space, three obvious variables of the experiment (Fig. 2) are: the direction S_0 of the incoming beam, the wavelength λ of those waves, and the direction S in which one senses the diffraction. (S_0 and S are unit vectors in $\mathbb{R}^3$.) It turns out that these variables are not independent as far as the results are concerned: the diffraction intensity I that one measures is only a "function" of the composite quantity $s = (S - S_0)/\lambda$. The intensity $I(s)$ is traditionally treated as a function of $s \in \mathbb{R}^3$ in the crystallography and physics literature, although it is usual to assume, on occasion, singular behavior of $I(s)$ which, like the

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well-known "delta functions", are more naturally modeled by distributions or measures. In particular, we will sometimes need to refer to examples for which $I(s)$ is supported on some countable set of points in $\mathbb{R}^3$; using the notation of real analysis, we will then say that $I(s)$ is "purely discrete". In signal analysis $I(s)$ would be called the (power) spectrum of the configuration, and we will use that term for later convenience.

The interest in quasicrystals all stems from two facts about the spectra $I(s)$ of the first quasicrystals that were identified: $I(s)$ was purely discrete; and there were axes through $s = 0$ about which $I(s)$ was invariant under 10-fold rotation, which is not possible for any "crystal". For us the chief characteristic of a crystal is the fact that the atomic nuclei in it are arranged "periodically"; that is, their configuration consists of unit cells which are repeated, as in Fig. 3. It is this feature which is incompatible with a 10-fold symmetry of $I(s)$. In fact, it was this incompatibility which showed that these materials were not crystals. We have yet to determine what the atomic-level configurations are for these materials; we just know, from the incompatibility, that they are not periodic in the manner of crystals.

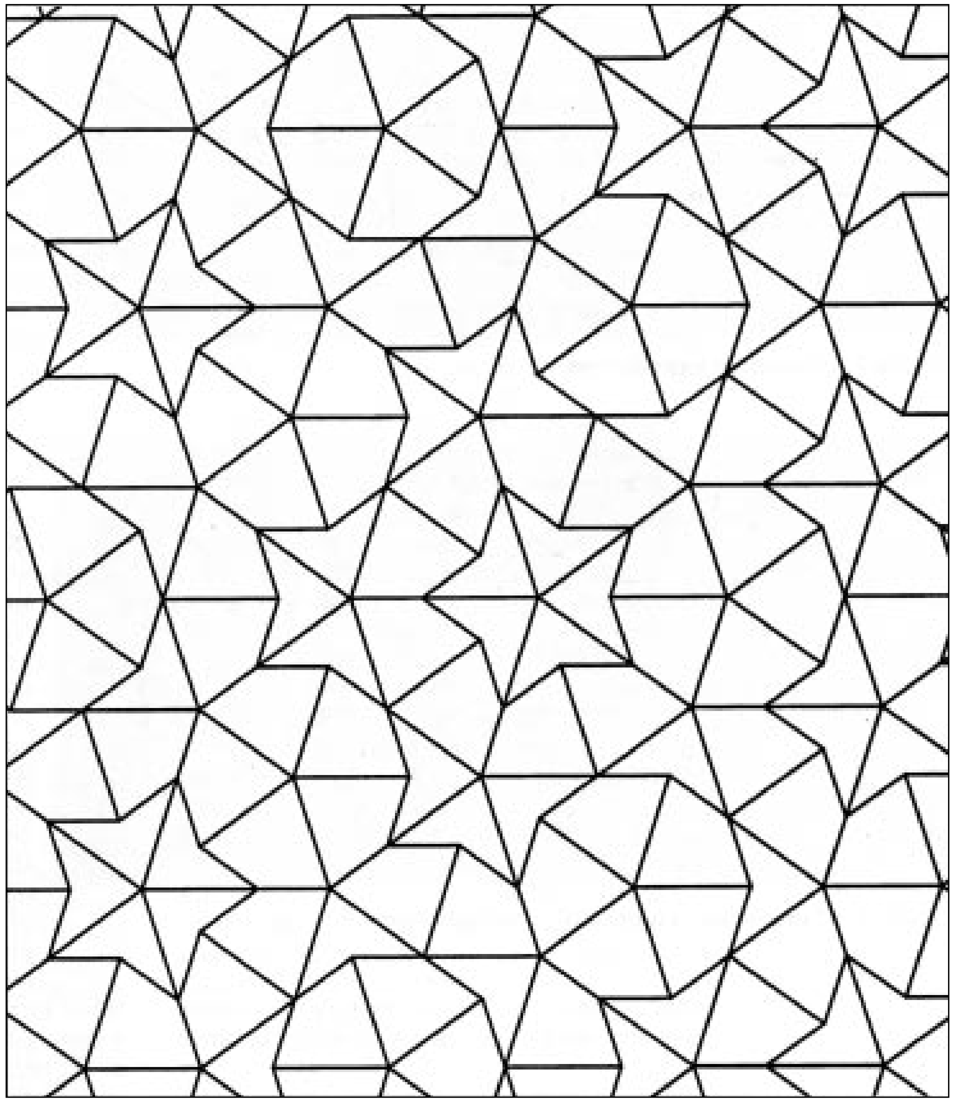

Figure 1. A Penrose "kite and dart" tiling.

The discovery of quasicrystals produced two surprises. On the one hand it had been widely thought that there are very general physical reasons why all (equilibrium) solids *had to be* crystals, so these noncrystalline substances should not have existed. Independently, it had been widely thought that only the *periodic* atomic configurations of crystals could have purely discrete spectra, so even if there were solids which were not crystalline, their spectra would have been expected to be nondiscrete.

Crystallography is devoted to the "inverse problem" of determining the structure of an atomic configuration from its spectrum. This process is greatly simplified by qualitative assumptions on the nature of the internal structure of the solid: in particular, assuming it is periodic is *very* useful and very widely used. The discovery of quasicrystals caused a profound stir in crystallography, since it affected some of its basic assumptions. This naturally led to a belated desire to understand what kind of atomic configurations could possibly produce the purely discrete spectra with which they are often confronted. In particular, crystallographers wanted to know what symmetries such atomic configurations could have, since symmetries of configurations become symmetries of their spectra and are heavily involved in the methods of crystallography. The book under review is chiefly motivated by this problem. (Crystallographers have been so affected by the discovery of quasicrystals that they have literally altered their definition of "crystal"!)

In condensed-matter physics (the subfield of physics devoted to understanding solids and fluids) the impact of the discovery of quasicrystals was large but not as profound as it was in crystallography, since quasicrystals do not seem to have particularly useful properties. The widespread attention paid to their discovery was simply due to unexpectedness. (Although it was a shock for physicists to realize they did not really understand the fundamental nature of so

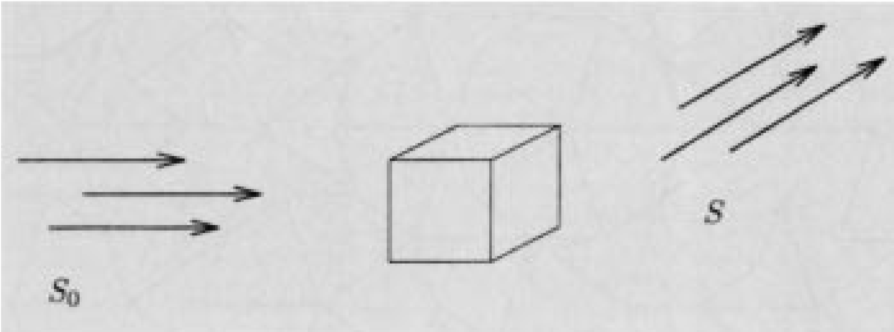

Figure 2. Diffraction experiment.

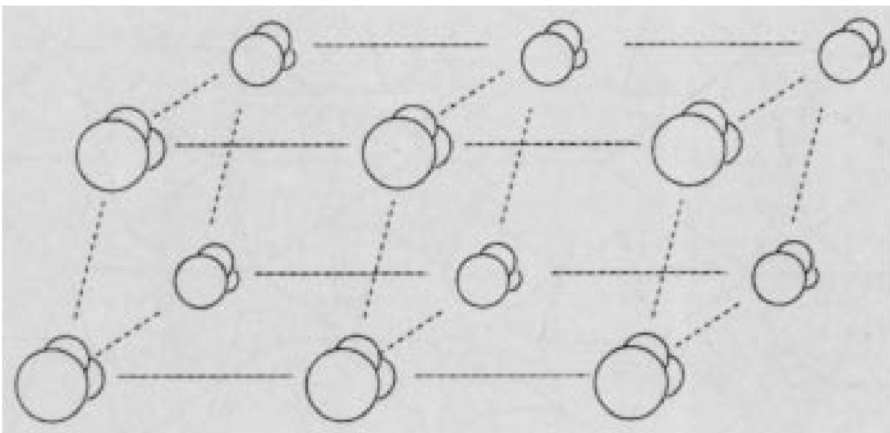

Figure 3. 12 unit cells of a periodic configuration.

basic an object as a solid, history has taught them repeatedly that their knowledge is provisional.)

We now come to the other basic discovery relevant to Senechal's book— "aperiodic tilings". First we need some notation. A collection of polygons is a tiling of the plane if the polygons cover the plane without gaps and do not overlap. (Much of what we say about tilings of the plane by polygons goes over to tilings of space by polyhedra.) There is an intuitive connection between tilings and configurations of points. The connection can be made more concrete by associating with a tiling the configuration of its vertices or by the Voronoi construction which associates a tiling to a configuration. However, such specific connections are not important here; it is enough to think of tilings and configurations as similar structures.

The discovery of aperiodic tilings was precipitated by a question of the philosopher Hao Wang in the early 1960s [9]: Is it possible to have a finite number of polygons such that congruent copies of them can tile the plane but *only* nonperiodically? (A tiling is "periodic" if it consists of repetitions of a unit cell, as in Fig. 4. We have generalized Wang's question slightly, using hindsight.)

The unexpected answer of yes by Wang's student Robert Berger [1] led, over the last thirty years, to the new subject of aperiodic tiling. Treating nonperiodicity as a form of "unexpected complication", the subject evolved to analyze what other forms of complication could be forced by a finite set of polygons; that is, what other aspects, not true of periodic tilings, could be true of *all* tilings built by copies of a given finite set of polygons. (The generalization to tilings of space by polyhedra is clear.)

Wang was a philosopher, and his motivation in asking the question was connected with the algorithmic nature of tilings. The first step in such an analysis is to enumerate the polygons in tilings and thus think of tilings as functions on the integers. One of the first generalizations of Wang's idea was the example by Dale Myers [5] of a finite set of polygons, congruent copies of which could tile the plane but only "nonrecursively". A tiling is recursive if it can be the output or result of an algorithm. Now, although it was the motivation for the birth of aperiodic tiling, this line of analysis, concerned with the *complexity* of the tilings forced by a finite set of polygons, has not yet proved very fertile. Most work has been concerned with two other forms of complication: tilings, forced by a finite set of polygons, which are not *ordered* as are periodic ones, and tilings which have *symmetries* not possible of periodic ones.

We discuss next these aspects of order and symmetry in tilings, beginning with order. The most mathematically developed measure of orderliness comes from probability theory (from which it was imported by ergodic theory and physics). We begin with an intuitive introduction, in the context of tilings.

Imagine we can see near us the finite portion of some tiling, and assume that although we know everything about the tiling, we do not know where we are in absolute terms within the tiling. We measure the degree of order of the tiling by the *extent to which we can infer, from the features near us, features of the tiling far away from us.* We use a probabilistic approach to justify such inferences, as follows.

There are two extremes of order. The usual model of extreme disorder is a sequence of independent coin flips. One can easily use this to give various definitions of a "random tiling", any of which would have the property that knowing some finite portion of the tiling would be of no value in predicting the details of the tiling further away. A reasonable model for extreme order would be any periodic tiling (consisting of repetitions of a unit cell), in that once one knows the location of a unit cell, one has complete knowledge of the rest of the tiling.

Between these two extremes we need a way to analyze the order of tilings. With the two extremes in mind, we think of the fact that some particular polygons occupy particular positions in a certain tiling of interest as an "event". For such an event R we use for its "frequency" $Freq(R)$:

$$Freq(R) \equiv \lim_{N\to\infty} \frac{\begin{array}{c}\text{the number of occurrences}\\ \text{of } R \text{ in a box of volume } N\end{array}}{N}$$

(not worrying here about existence of limits, etc.) Now assume we know some finite number of polygons near us (event P) and we want to know if some other finite set of polygons is at a location with relative position given by the vector t (event Q_t). We say our tiling is *disordered* if, for any P and Q,

$$Freq(P\&Q_t) - Freq(P)Freq(Q) \xrightarrow[t\to\infty]{} 0.$$

In ergodic theory this is closely associated with the concept of mixing, and various refinements are known. In fact, ergodic theory as a subject was created to analyze such frequencies. The basic theorem of the subject is the pointwise ergodic theorem of G. D. Birkhoff, which in this setting is simply a way to compute such frequencies by averaging over tilings instead of over the occurrences of patterns in a tiling.

Examples have been found of finite sets S of polygons which force tilings with various degrees of order. For instance, an S is known for which in all tilings and for all P and Q:

$$\frac{1}{T^2}\int_{|t|\le T} |Freq(P\&Q_t) - Freq(P)Freq(Q)|\, dt \xrightarrow[T\to\infty]{} 0,$$

which comes close to the above definition of disorder. But it is still an important unsolved problem to produce a set S which only allows disordered tilings.

Crystallographers like to use a different criterion of order, associated with diffraction. They are particularly interested in configurations for which the spectrum is purely discrete. To understand why this is an order property, it is again useful to turn to ergodic theory. In the ergodic theory of configurations or tilings (in which the "dynamical" group is that of translations in $\mathbb{R}^d$) there is also a notion of spectrum. Since we have made a point of using the same term *spectrum* for both diffraction and ergodic theory, it should come as no surprise that the two notions are the same (though the details only appeared in the literature recently [2]). We will, of course, use the same symbol for it, $I(s)$. It turns out that in ergodic theory the spectrum is one of the main tools used to analyze order, and in particular it is easy to show that discrete spectrum implies order in the sense discussed above. Crystallographers are interested in the interrelations between this order property and symmetry, which we address next.

Most of the effort in aperiodic tilings is connected in some way or other with symmetry and has a curious history. After Berger published his example, which used over 20,000 different (noncongruent) polygons, a number of people became interested in simplifying the result, in particular by reducing the number of different polygons. In the original examples it was natural (for their algorithmic interpretation) to use only polygons which would appear in the tilings in a single orientation, never rotated. But driven by the criterion of reducing the number of different polygons allowed, researchers (for instance, Raphael Robinson [7]) found it advantageous to have polygons appear in several orientations in the tilings. These were followed by the "kite and dart" tilings (Fig. 1) of Roger Penrose [3]. The Penrose tilings consist of copies of only two polygons (Fig. 5), which appear in ten different orientations. They have an interesting feature which

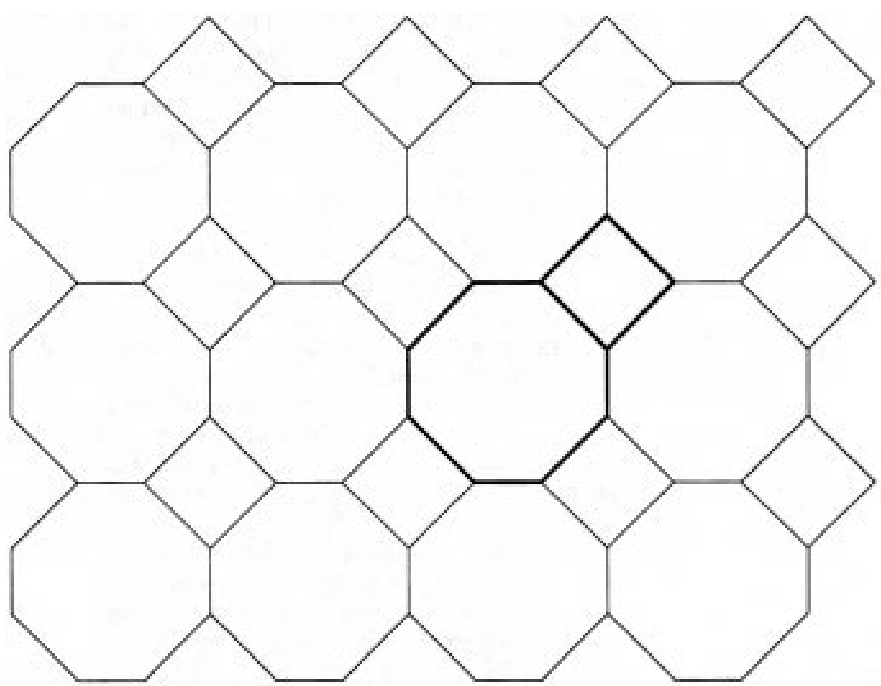

Figure 4. Periodic tiling, with unit cell in dark lines.

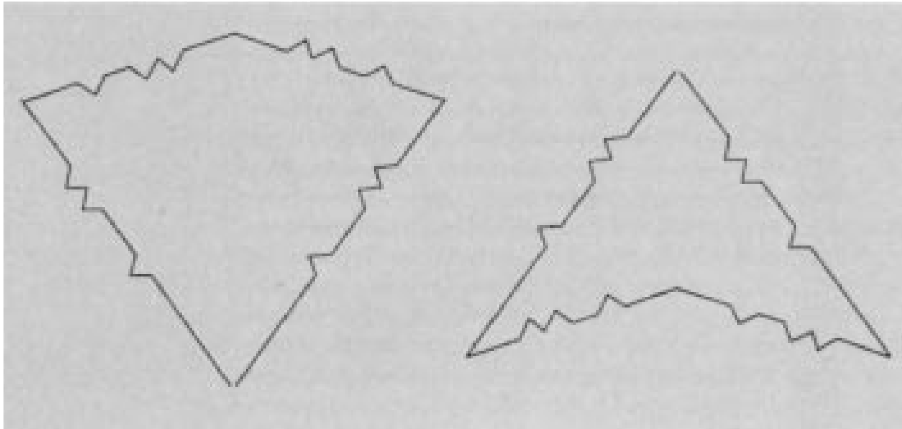

Figure 5. Kite (left) and dart (right).

attracted attention: essentially, each tiling consists of *arbitrarily large* overlapping regions for each of which there is a point about which the region is invariant under rotation by $2\pi/5$. The kite and dart tilings are said to exhibit "approximate 5-fold symmetry". It was well known that this is impossible for any periodic tiling, and this has spawned much research trying to understand what approximate n-fold symmetries could hold for *all* the tilings made of copies from some finite set of different polygons. Such tilings are of interest in part to help understand what the atomic configurations might be for real quasicrystals. This is the main open problem underlying the presentation of the book under review.

There is an intuitive similarity between the (statistical mechanics) model of the way particles gather together in solids and the jigsaw-like manner that governs how polygons gather together in tilings. This has led physicists to see in aperiodic tiling a simple model to understand how noncrystalline materials might be created, in particular materials with atomic configurations with spectra of unusual symmetry.

Crystallographers, as a whole, are less interested than physicists in the fundamental issue of how quasicrystals could possibly arise from interparticle forces. So they have extended their use of tilings as models of matter by dropping the condition of aperiodicity. In its place they insert as an *assumption* a strong form of order; they consider tilings with discrete spectra and try to determine which approximate n-fold symmetries are possible. This path is thoroughly examined by Senechal. Most of the examples found with interesting approximate n-fold symmetries have been produced by a method which obtains the tilings by projection from a higher-dimensional periodic tiling. This technique, and the examples, are well described in the book.

There is another wrinkle in the symmetry game. When a particle configuration produces a diffraction pattern, what is sensed about the configuration is not the complete information about the positions of all the particles, but only certain statistical facts about the configuration. In particular, in order for the spectrum of a configuration (discrete or not) to exhibit n-fold rotational symmetry about some axis it is sufficient *but not necessary* for the configuration to have the associated approximate n-fold symmetry. What is both *necessary and sufficient* is that the configuration have the associated "statistical n-fold symmetry", in the sense that every finite subconfiguration appear in the full configuration, in a given orientation, with the *same* frequency as it would if the configuration were rotated by $2\pi/n$ about the axis [6]. (The frequency of appearance of a finite subconfiguration is defined in the same way as was the frequency of a finite collection of polygons in a tiling: by counting how many times it occurs in a big box, dividing by the volume of the box, and taking the limit of the fraction as the box gets arbitrarily large.) This has led to a study of tilings with statistical rotational symmetries, with some unexpected developments. For instance, it turns out to be possible for a tiling, using congruent copies of finitely many different polygons, to be statistically symmetric under rotation *by all angles.* This idea can also be mixed with aperiodicity to produce a finite set of polygons such that *all* the tilings using such polygons have some n-fold statistical rotational symmetry—even that of "infinite n" which was just mentioned. Statistical symmetry has also led, through the search for new effects in three-dimensional tilings, to new results on subgroups of $SO(3)$.

As might be guessed from the appearance of frequencies in its definition, statistical symmetry is closely connected to ergodic theory. Thus we see that ergodic theory is useful in analyzing both the order and symmetry properties of structures like configurations or tilings. It seems to be the natural mathematical framework for the various aspects of aperiodic tiling. Though it is a subjective judgement, we go further and claim that those results from aperiodic tilings which are already of significance in other parts of mathematics have used ergodic theory at least as inspiration. As a prime example we note results of Shahar Mozes [4] which have used tilings to discover a very surprising connection between two traditional parts of symbolic dynamics, "subshifts of finite type" and "substitution subshifts".

We have given an overview of a very broad area of research and mentioned at a few points how the material is treated in the book under review. But it is now appropriate to concentrate on the book to get a feel for it as a whole.

The book divides naturally into two parts. The first half discusses configurations of points, and the second half discusses tilings. The first half contains the presentation of the main theme, the order and symmetry properties of these structures. The second half is a wonderful catalog of examples of interesting tilings.

In the first half the study of configurations of points is expressly motivated as part of a model for diffraction from an atomic configuration. There is a strong emphasis on configurations which can be obtained from a projection technique from some higher-dimensional lattice, $\mathbb{Z}^n$. The reason for this preference is that such configurations can easily produce configurations with unusual approximate n-fold rotational symmetries, of interest in crystallography. There is a good treatment of some of the algebraic aspects of symmetry groups, to the extent needed to see what types of examples can be produced by the projection technique.

The notion of order as a property of configurations is not explored in depth. Instead, it is simply declared that it is an indication of order when a configuration has purely discrete spectrum and that this feature is a desirable one to assume in configurations which will be analyzed. This is in keeping with the practical interests of crystallography, where it is important to know what configurations of particles can produce purely discrete spectrum.

The spectrum itself is introduced as it would be in a physics or crystallography text. The spectrum of an infinite configuration $C = \{x_j : 1 \leq j < \infty\}$ is approximated by that of a finite part C_N of C. The approximate spectrum $I_N(s)$ is:

$$I_N(s) = \frac{1}{N} | \sum_{j=1}^{N} e^{is\cdot x_j} |^2 = \frac{1}{N} \sum_{j=1}^{N} \sum_{k=1}^{N} e^{is\cdot (x_j - x_k)}.$$

The desired spectrum $I(s)$ of the full configuration C is then the limit of $I_N(s)$. The limit is a delicate matter; recall that $I(s)$ must be treated as a measure or distribution. Algebraic manipulation of sums such as $I_N(s)$ can be revealing, and scientists are adept at this. But it is precisely to understand the subtleties that can arise in their limits that often motivates mathematics, in particular the spectral analysis developed in functional analysis and ergodic theory.

This is the point at which the author had to make a significant choice. To make the book accessible to a wide audience of scientists, crystallographers, and mathematicians, the author decided to minimize the mathematical background assumed of the readers. This meant omitting any serious discussion of spectral analysis or ergodic theory, which would have shown how the material fits into mathematics.

The second half of the book is concerned primarily with aperiodic tilings. There is an interesting chapter on some aspects of the Penrose tilings, in particular how to obtain them from projection from the five-dimensional lattice, $\mathbb{Z}^5$.

To me, Chapters 7 and 8 are worth the price of the book. The former contains an extensive discussion of the known aperiodic tilings and analyzes tilings made by projection. The latter contains computer programs and their graphic output for numerical simulation of the spectra of various tilings. (The spectrum of a tiling is here defined as the spectrum of the configuration of its vertices.)

In summary, *Quasicrystals and geometry* is concerned with a variety of phenomena which are expressed equivalently in terms of configurations or tilings. The main characteristics of these structures which are studied are order properties and symmetry properties, both separately and as they affect one another. The book is aimed at a very broad scientific audience, and the level of mathematics is kept low accordingly. There is, however, significant mathematics coming out of this area. Those wanting to pursue this would need to hunt a bit in more specialized literature. However, there is a large number of appealing examples— well described, illustrated, and referenced— which should fire the imagination and entice new blood into related research.

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