# Tilings, Diffraction, and Quasicrystals

**Nonperiodic tilings exhibit a remarkable range of "order types" between periodic and amorphous. The six tilings shown on these pages are a representative sample. How can we characterize these states, and how are they reflected in various measures of order, such as diffraction images? The answers to these questions – which are still wide open – may have applications to contemporary crystallography.**

*Marjorie Senechal*

## Order and Disorder

Not so many years ago, patterns were classified as periodic or nonperiodic, and no further distinctions seemed necessary. But a glance at the tilings shown on these pages (at the upper left of each plate), all of which are nonperiodic, suggests that "nonperiodic" encompasses a broad range of order types, from what we might call "almost periodic" to "quasiperiodic" to "amorphous" (quotation marks are used here because none of these terms has a generally accepted definition).

Nonperiodic tilings – a fairly new subject with a fascinating if brief history [Grünbaum and Shephard 1987] – provide a wealth of examples of orderly disorder. A periodic tiling is one with translational symmetry; roughly speaking, this means that if you displace one of two superimposed copies of the (infinite) tiling through a certain distance in a certain direction, the two tilings will again match up. If a tiling is nonperiodic, any such displacement will cause a mismatch somewhere. Nevertheless, nonperiodic patterns can have a great deal of order. For example, all the tilings shown here have a form of "local" symmetry, called repetitiveness: every *bounded region* in any tiling occurs infinitely often in that tiling. It follows that you can make two superimposed copies of the tiling again coincide – over arbitrarily large regions – by appropriately displacing one of them. This is perhaps easiest to see in the Penrose tiling: choose *any* patch, such as the star made of five thick rhombs, and notice that this configuration appears quite frequently. If you shift a copy of the tiling so that one of these configurations is brought into coincidence with another, the two tilings will coincide at least on that configuration, though they will not match up completely everywhere. The larger the patch, the farther you may have to look for a copy, but copies can always be found. All these tilings were produced by a method called *substitution* (see below); any tiling generated by substitution is repetitive.

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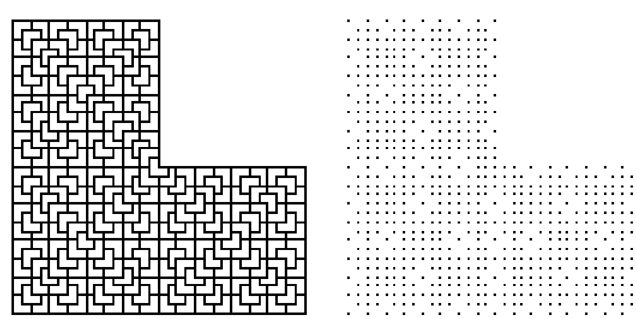
The differences among these nonperiodic, repetitive tilings are more obvious than their similarities, though it is not easy to characterize these differences in a precise way. We will just point out a few of them.

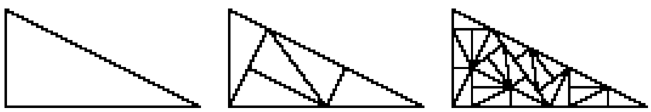
First, notice that the vertices of the tiles of the chair (and also sphinx) tilings occupy some – but not all – of the sites of a two-dimensional lattice. None of the other four tilings has this property.



Second, the prototiles of the Penrose and binary tilings are identical (there are two of them, "thick" and "thin" rhombs) and they occur with the same frequencies (the ratio of thick to thin is the Golden Number $(1+\sqrt{5})/2$, which is also the ratio of their areas), but the Penrose tiling is much more "orderly" than the binary.

Third, the chiral and pinwheel tilings both appear to be somewhat chaotic, in the nontechnical sense of the word. In fact, the chiral tiling is a variant of the Penrose tiling in which some randomness has been introduced in the construction algorithm. There is no randomness in the pinwheel tiling; the apparent disorder is due to the fact that the tiles (in the infinite tiling) occur in infinitely many orientations! No other tiling of the plane with this property is known, at least as of March 1994.

Substitution is a composite operation consisting of decomposition and inflation. The substitution method is applicable when each prototile can be subdivided into smaller copies of itself and the other prototiles (if there is more than one prototile). To generate the tiling, we begin with one tile or a small patch of tiles. We first perform the decomposition on each tile, and then rescale the small tiles to the sizes of the originals. Now we have a larger patch of tiles than the one with which we began. Repeating the decomposition and rescaling, ad infinitum, we obtain – in the limit – a tiling of the entire plane. For example, the prototile of the pinwheel is a right triangle whose edges are proportional to 1, 2, and $\sqrt{5}$; it can be subdivided into five smaller triangles similar to itself. Here is the original triangle and the first two stages of decomposition.



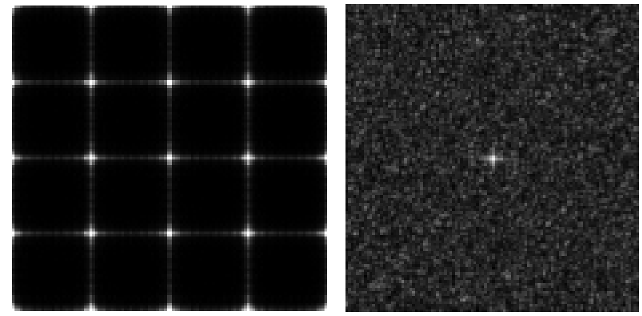
Using *Mathematica*, one can easily iterate such substitutions: the prototype code is that for the Penrose tilings in [Wagon 1991], and the necessary modifications to create the tilings shown here are described in [Senechal].

These six tilings are fairly representative of the different kinds of order that a nonperiodic tiling can have. We can measure this order in various ways. One method is to compute the tiling's diffraction spectrum. (The relation between tilings and their diffraction spectra is the central theme of *Quasicrystals and Geometry* [Senechal], in which these plates will appear.)

## Diffraction: A Measure of Order

Diffraction is not widely used in geometry, but it is an essential tool in physics, chemistry, and crystallography, where it plays a central role in the determination of crystal structures. The structure of a crystal is inferred from its x-ray (and electron and neutron) diffraction images, together with other data; these images give information about symmetry and orderliness needed to establish the positions of the atoms. A diffraction pattern is essentially a density map of the function that specifies the intensities of the diffracted waves. This function does not record the positions of the points, only their *relative positions*; generally speaking, the more point-pairs in the same relative position, the greater the intensity of the diffracted waves, in certain directions. This is why the diffraction image is a good measure of "order." Compare, for example, the diffraction patterns of the Penrose and binary tilings: the differences you see are directly related to the fact that local patterns are more densely packed in the Penrose tilings than in the binary.

Our use of diffraction patterns – to measure the order in a tiling – is somewhat different from the crystallographer's. We are not trying to determine the structure of a tiling: we know what it looks like because we created it ourselves. And, as explained below, we can compute the intensity function in a straightforward way. What we are interested in is the nature of this function, which we call the *spectrum* of the tiling. It is always a sum of "discrete" and "continuous" components, though one of them may be identically zero. The discrete component indicates order, the continuous component disorder. The two extremes are shown in the diffraction patterns produced by a lattice (left) and by a random set of points (right):



Thus, we can characterize the orderliness of the tiling by means of its spectrum.

X-ray diffraction is formally analogous to optical diffraction: the atoms of a crystal scatter x-rays in just the same way that point apertures scatter laser light, and the diffraction pattern produced on a screen in an optical experiment is analogous to the x-ray diffraction pattern that the crystallographer measures by computer or captures in a photograph.

The diffraction patterns shown here simulate with remarkable precision the optical diffraction patterns that would be produced if the vertices of the tiles were point apertures in a metal plate.

To produce these patterns, the set $\Lambda$ of vertices of each tiling was computed by *Mathematica*, and then piped to Fourier, a C program written by Stuart Levy (for the code, see [Senechal] or contact `slevy@geom.umn.edu`). Fourier computes the diffraction pattern in the following way. We assume that each point (vertex) is assigned a unit mass: formally, we place a Dirac delta at each point. The "density function" of the set of points is the sum of these deltas:

$$\rho(\mathbf{x}) = \textstyle\sum_{\mathbf{y}\in\Lambda} \delta(\mathbf{x} - \mathbf{y}).$$

For finite point sets, the intensity function $J(\mathbf{s})$ is the squared modulus of the Fourier transform of $\rho(\mathbf{x})$:

$$J(\mathbf{s}) = \left| \textstyle\sum_{\mathbf{y}\in\Lambda} \exp(-2\pi i\, \mathbf{y} \cdot \mathbf{s}) \right|^2.$$

It is easy to show that $J(\mathbf{s})$ is a trigonometric sum (for more details of this computation, see [Senechal] or [Cowley 1986]). Fourier computes $\sqrt{J(\mathbf{s})}$; its output is a `.pgm` file, the "diffraction pattern."

Images such as these give qualitative, not quantitative results, but even so they are very instructive. In each plate you see, at the top right, the gray map calculated by Fourier from the coordinates of the vertices of that portion of the tiling shown at the left. The image at the bottom left is the same diffraction pattern, with low-level intensities enhanced (using the software tool *xv*). The image at the bottom right is the negative of that on the left.

What can these diffraction images tell us about the orderliness of the tiling? All six diffraction images show spots, but notice that the spots are fewer and fainter for the binary and pinwheel tilings than for the others. This is not a fluke: in fact, one can prove that the spectra of these two tilings have no discrete components. The spots we see may indicate a "singular" component in the continous part of the spectrum, or they may be due to the fact that the number of vertices used in these "experiments" was small (about 500).

The enhanced images help us see what is going on. The first thing to notice, in all cases, is that the regions that appear to be completely dark at the top right are not devoid of structure (as they would be if the tilings were lattice-like). But the nature of this "background" varies from tiling to tiling. For the chair, the sphinx, and the Penrose tilings, the background consists of points of varying brightness: in fact, their spectra do not have continous components. The chiral tiling seems to have a mixed discrete and continuous background, but the continous background is much more evident in the binary and pinwheel tilings. This gives us a rough classification in keeping with our observations about the tilings themselves: the chair, sphinx, and Penrose tilings are more orderly than the chiral tiling, which is more orderly than the remaining two. There are further gradations, some of which can be read from these pictures; for more details see [Senechal].

### Tilings and Quasicrystals

This analysis may have implications beyond tiling theory: nonperiodic tilings are being studied as possible structure models for the strange new crystals, generically known as *quasicrystals*, that have been discovered in the last decade.

The quasicrystal story began in 1984, when an aluminum-manganese alloy was discovered whose diffraction images displayed the symmetry of a regular icosahedron [Shectman et al. 1984]. This news quickly hit the headlines of newspapers as well as scientific journals: it augured a fundamental paradigm shift in our picture of the solid state. For icosahedral symmetry is impossible for a periodic atomic structure, which had been for nearly two centuries the very definition of a crystal. There is good reason to assume that a crystal is a discrete, modular structure, something like a tiling with a small number of prototiles. But it was also assumed that the structure must repeat periodically, like a wallpaper pattern. Look at any papered wall, or at one of the periodic drawings of M.C. Escher, many of which he created at the request of crystallographers for teaching purposes (see, for example, [Schattschneider 1990]). You will find that despite their apparent complexity, all these patterns are based on a simple framework of parallelograms, or in more symmetrical cases of equilateral triangles or squares or regular hexagons (these are the only regular polygons that tile the plane). Accordingly, the only rotational symmetries you will find in such patterns are two-, three-, four-, and six-fold. No other rotational symmetries can appear in a repeating pattern (for example, the plane cannot be tiled by regular pentagons). For the same reason, five-fold symmetry is incompatible with a periodic structure in a crystal. The icosahedron has five-fold as well as three-fold and two-fold symmetry. Evidently, the world of crystals is larger than had been thought!

But if symmetry is not the defining characteristic of a crystal, what is? This question is far from being answered. But over the past decade, more and more examples of crystals with other "forbidden" symmetries have been found in laboratories throughout the world. This suggests that nonperiodicity rather than periodicity may be the hallmark of the solid state. To understand what a broader definition of "crystal" might entail, we need to study the varying degrees of order.

Nonperiodic tilings are an excellent tool for studying nonperiodic solid phases. Notice, for example, that the diffraction patterns of the Penrose (and chiral and binary) tilings show five-fold symmetry! Thus, it is reasonable to assume that some of the properties of the real materials – physical properties such as conductivity and stability, as well as structure – can be modeled by nonperiodic tilings (for detailed examples of how this is being done, see, for example, [DiVincenzo and Steinhardt 1991]). We can also use tilings to explore the structure of order and disorder, and to learn how the order properties of a pattern are reflected in its images and conversely.

*Warning*: Just about everything in this account has been oversimplified, and the necessary ifs, ands, and buts have been deliberately omitted. For caveats and details, see [Senechal]. For more details about pinwheel tilings, see [Radin 1994].

### References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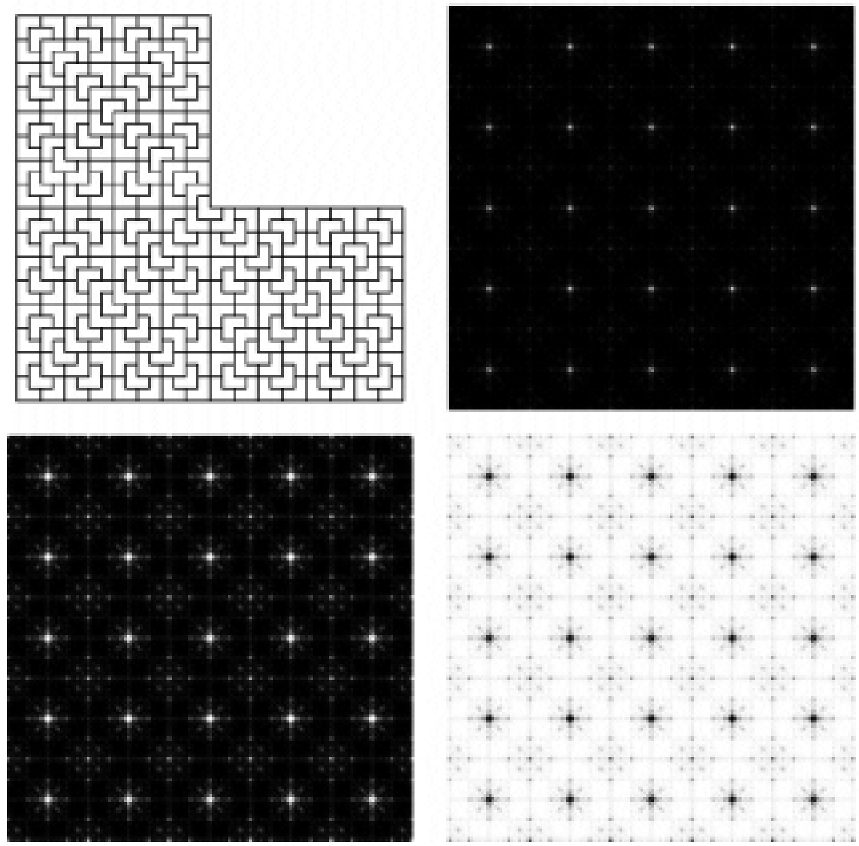
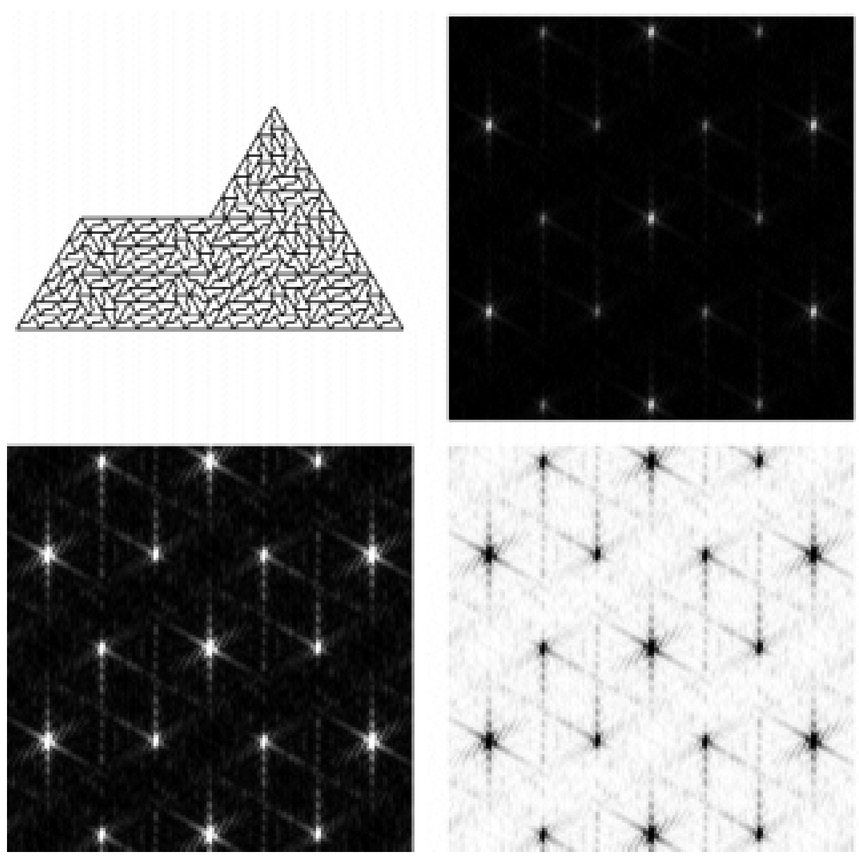
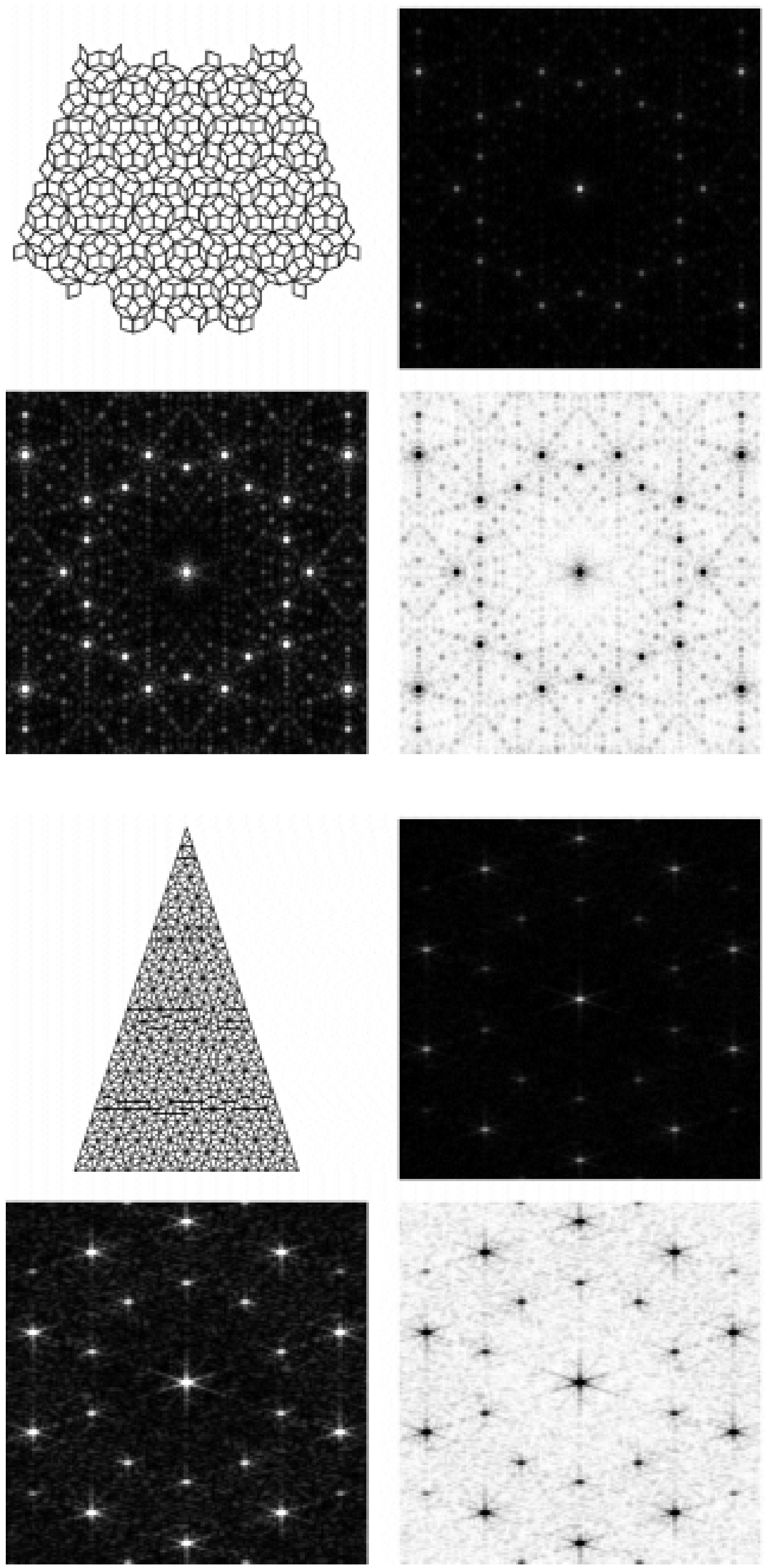
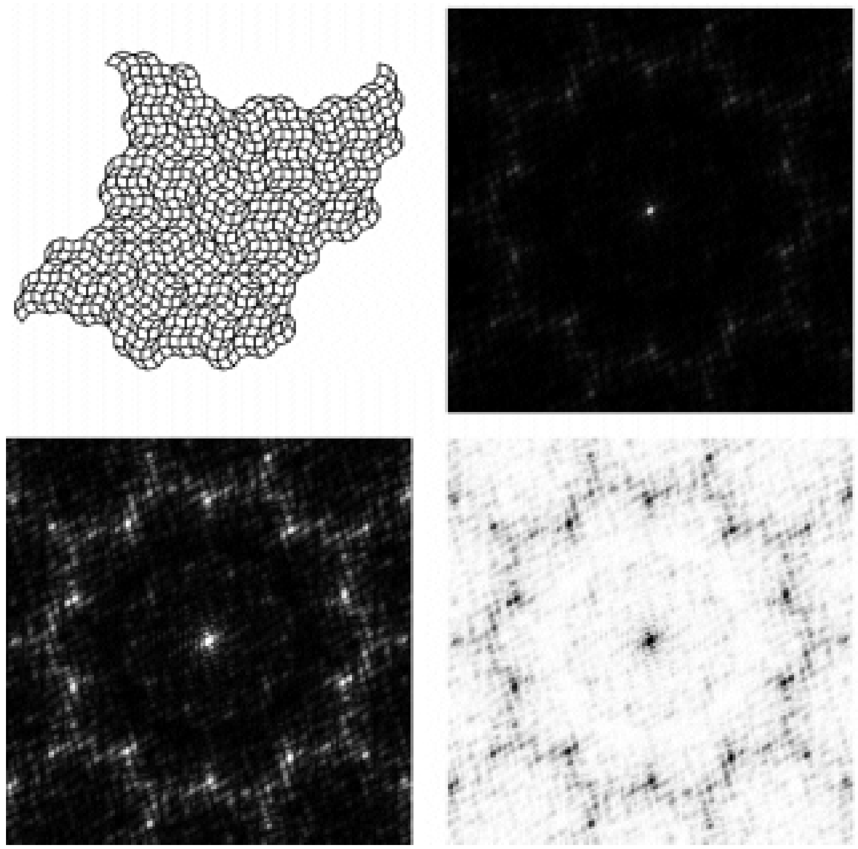


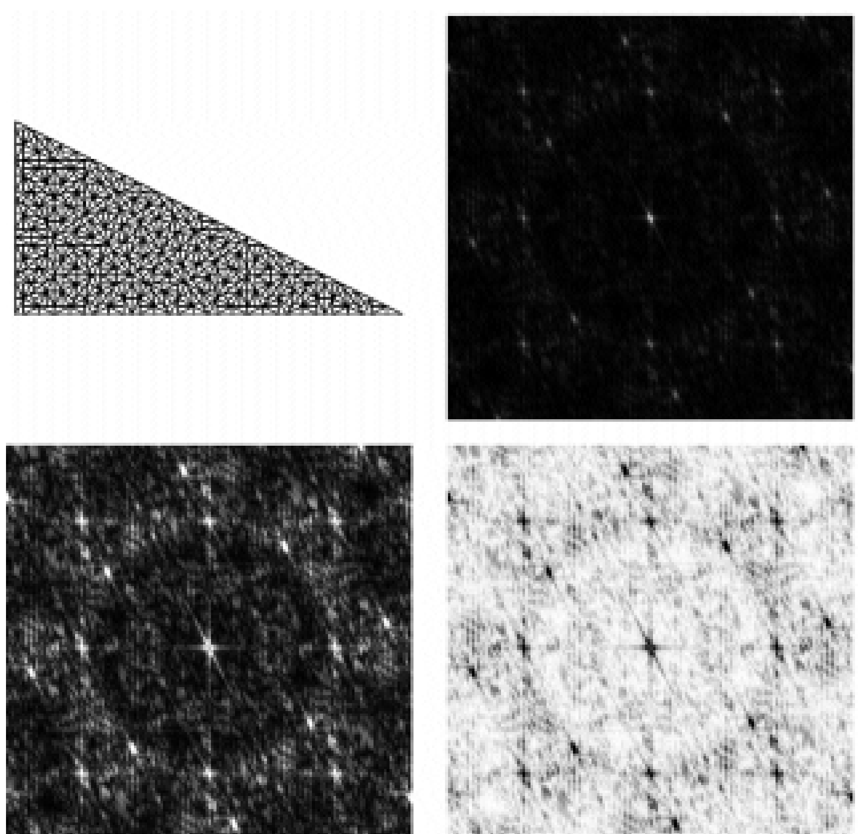


Top: The chair tiling. Bottom: The sphinx tiling. From M. Senechal, *Quasicrystals and Geometry*, Cambridge University Press, 1995.



Top: The Penrose rhomb tiling. Bottom: A chiral tiling. From M. Senechal, *Quasicrystals and Geometry*, Cambridge University Press, 1995.





Top: The binary tiling. Bottom: The pinwheel tiling. From M. Senechal, *Quasicrystals and Geometry*, Cambridge University Press, 1995.