The Structure of Quasicrystals

Quasicrystals are neither uniformly ordered like crystals nor amorphous like glasses. Many features of quasicrystals can be explained, but their atomic structure remains a mystery

by Peter W. Stephens and Alan I. Goldman

When aluminum, copper and iron are melted together and cooled, they can solidify to form a grain in the shape of a perfect dodecahedron, a geometric solid whose 12 faces are regular pentagons. Although this dodecahedral grain looks like a crystal, it is not. Crystals are composed of identical building blocks called unit cells, each containing precisely the same distribution of atoms and each fitting together with its neighbors in the same way. A dodecahedral grain cannot be constructed from atoms in unit cells of a single shape whether they be small cubes or even dodecahedrons. The dodecahedral grain is a quasicrystal.

Indeed, all probes of atomic-scale structure show that quasicrystals are not made up of repeated unit cells. It is clear that these exotic new materials cannot be crystals, but it is not immediately apparent just what they are. As physicists, chemists and materials scientists have investigated the structure of quasicrystals, they have come to realize that periodic crystals, whose atomic structures they have studied over the past 78 years, are but a subset of the possible types of ordered solids.

Since 1984, when Dan S. Shechtman and his colleagues at the National Institute of Standards and Technology (NIST) discovered the first quasicrystal,

workers have fabricated many different species of quasicrystalline alloys [see "Quasicrystals," by David R. Nelson; SCIENTIFIC AMERICAN, August 1986]. They have learned how to improve samples to the point that they can quantitatively study issues that were only idle speculations seven years ago. The study of quasicrystals has advanced three theories about their structure: the Penrose, glass and random-tiling models.

The Penrose model—derived from the work of mathematician Roger Penrose of the University of Oxford—suggests that quasicrystals are composed of two or more unit cells that fit together according to specific rules. The model accurately describes some of the basic properties of quasicrystals, but it has difficulty explaining how these rules might be related to atomic growth processes.

The glass model, in contrast, relies on local interactions to join clusters of atoms in a somewhat random way. According to the model, all the clusters have the same orientation, but because of random growth, the structure contains many defects.

It now seems that the two models are converging toward a third, the random-tiling model, which combines some of the best features of its predecessors. In the past few years the structure of quasicrystals has been one of the most hotly debated topics in solid state physics. The resolution of this debate may lead to a theory of quasicrystalline structure and guide the development of materials with unusual structural and electrical properties.

To produce the first quasicrystals, Shechtman and his colleagues at NIST melted together aluminum and manganese and then squirted the molten metals against a rapidly spinning wheel, thereby achieving a cooling rate of about one million kelvins per second. This abrupt cooling process, called quenching, can "shock" the alloy

into a variety of novel structures, or phases. To understand these unusual phases of matter, one must first have a grasp of some of the principles of basic crystallography.

A crystal can possess only certain symmetries because there are a limited number of ways that identical unit cells can be assembled to make a solid. For instance, a salt crystal is composed of cubic unit cells that stack to form cubic grains. Consequently, the salt crystal has fourfold rotational symmetry: when the crystal is simply rotated through a quarter turn around the appropriate axis, atoms of the rotated crystal occupy the same positions as those of the unrotated crystal. Crystals
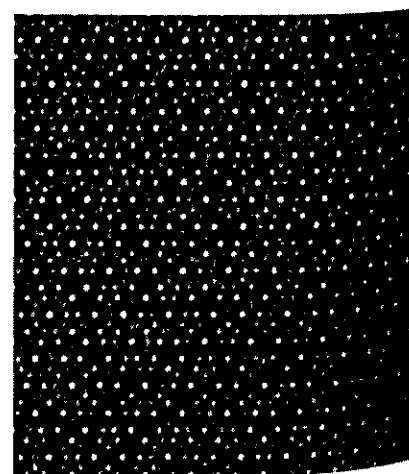
QUASICRYSTAL (*right*) can be distinguished from other forms by its unusual symmetry. The material can form grains in the shape of dodecahedrons, solids whose 12 faces are regular pentagons. The dodecahedral grain has fivefold symmetry, that is, when rotated by one fifth of a circle about an axis through one of the faces, its appearance is unchanged. The grain is composed of aluminum, copper and iron and is about 300 microns in diameter. The electron micrograph (*above*) shows a slice through several atomic layers, also exhibits fivefold symmetry.

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can have only twofold, threefold, four-fold and sixfold symmetry.

A crystal can never have, say, five-' symmetry, because a single unit that has fivefold symmetry, such as a dodecahedron, cannot be assembled to completely fill space. There will always be gaps between the dodecahedral unit cells.

To determine the structure of a crystal, investigators use an indirect, but well understood, technique. Atoms in a crystal are arranged in families of parallel planes. Each such plane acts as a mirror to incoming X rays, electrons and other rays or particles that trav-el through space as a wave. Each plane reflects the incident waves very weakly. But if the reflected waves from each member of a family of planes combine in phase, the total intensity of the reflected wave can become quite strong. This phenomenon is called diffraction; it occurs whenever any type of wave interacts with an ordered structure of the appropriate spacing.

When a crystal is bombarded by a beam of X rays or electrons, the angles through which the waves are diffracted reveal the shape and dimensions of the unit cells of the crystal. The diffracted waves can be recorded, for example, on a photographic emulsion, where they appear as a pattern of bright spots.

All the symmetries of a crystal are reproduced in its diffraction pattern. A crystal with sixfold rotational symmetry will produce a diffraction pattern that also has sixfold symmetry. Because no crystal can have a fivefold symmetry axis, one would not expect to see a diffraction pattern that has fivefold rotational symmetry.

Nevertheless, when Shechtman and his colleagues illuminated a grain of the aluminum-manganese alloy with electrons, they found a diffraction pattern that had fivefold rotational symmetry.
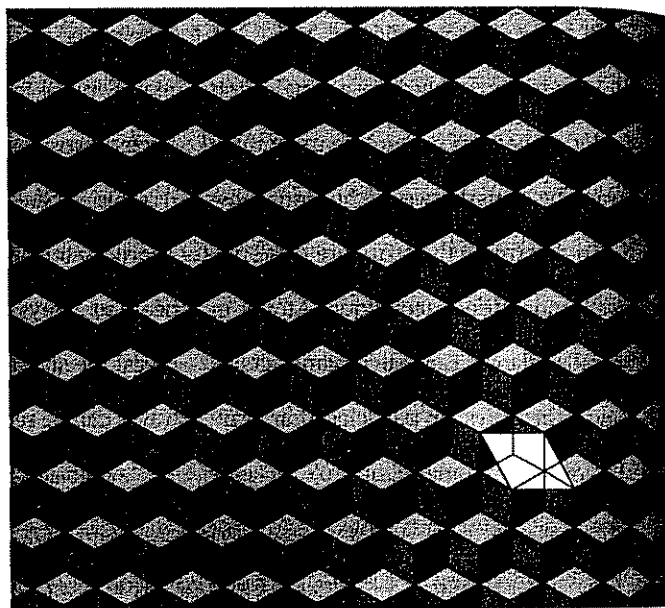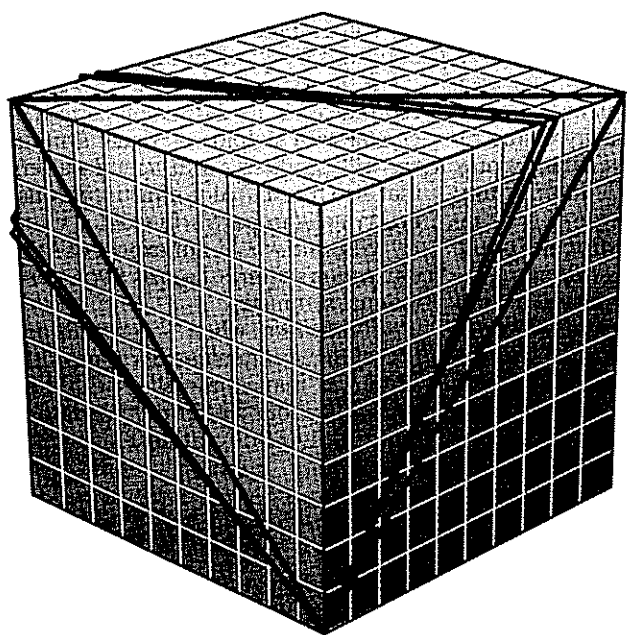
PERIODIC OR QUASIPERIODIC pattern can arise when a plane cuts through a stack of cubes (*left*). The green plane intersects the cubes to form the green pattern at the right. It is periodic in the sense that the pattern can be assembled by duplicating and positioning a unit cell (*white area*) in an orderly fashion. The blue pattern is also periodic and has a unit

Indeed, by rotating the sample by the appropriate angles, they deduced that the alloy had six fivefold symmetry axes. In technical terms, the alloy has icosahedral symmetry, because an icosahedron is a 20-sided solid that has six fivefold rotational axes in the same orientation as the alloy.

The icosahedral alloys are only one of many families of quasicrystals discovered during the past seven years. All these materials have symmetries that are "forbidden" in conventional crystals. Leonid A. Bendersky of NIST found that aluminum and manganese can form a material that is periodic along one direction and has tenfold rotational symmetry in the perpendicular plane. Workers have also recently fabricated quasicrystals with eightfold and 12-fold symmetry. In some sense, these quasicrystals provide a link between quasicrystalline and crystalline order. They also demonstrate that the phenomenon of quasicrystallinity extends far beyond ideas about icosahedral symmetry and the stability of specific icosahedral clusters of atoms.

The first model for quasicrystals emerged from the mathematics of tiling—a field advanced by Penrose and others during the 1970s [see "Mathematical Games," by Martin Gardner; SCIENTIFIC AMERICAN, January 1977]. Penrose examined how two or more shapes could be assembled in a quasiperiodic way to tile a plane, that is, to cover it completely with shapes that do not overlap. Each of these qua-

siperiodic tilings, now known as Penrose tilings, could be constructed using a set of instructions called matching rules [see box on page 51].

In 1982 Alan L. Mackay of the University of London calculated the diffraction properties of a theoretical quasiperiodic structure. He demonstrated that if atoms were placed at the corners of each shape in a Penrose tiling, they would give rise to a diffraction pattern that had tenfold symmetry. Then, in 1984, Peter Kramer and Reinhardt Neri of the University of Tübingen extrapolated the concept of two-dimensional Penrose tilings to three dimensions. Dov Levine and Paul J. Steinhardt of the University of Pennsylvania had also been considering the possibility of alternative forms of atomic order based on Penrose tilings.

Building on this early work, Levine and Steinhardt proposed the Penrose model for quasicrystals only six weeks after Shechtman and his colleagues published their famous paper. They put forth a three-dimensional generalization of the Penrose tiling that described the structure of the aluminum-manganese alloy, and they showed that the pattern of diffraction peaks calculated from their model agreed well with Shechtman's results.
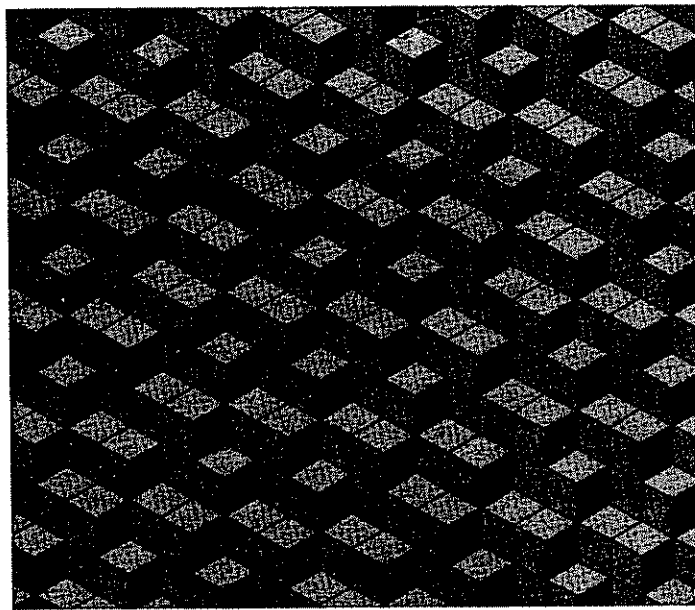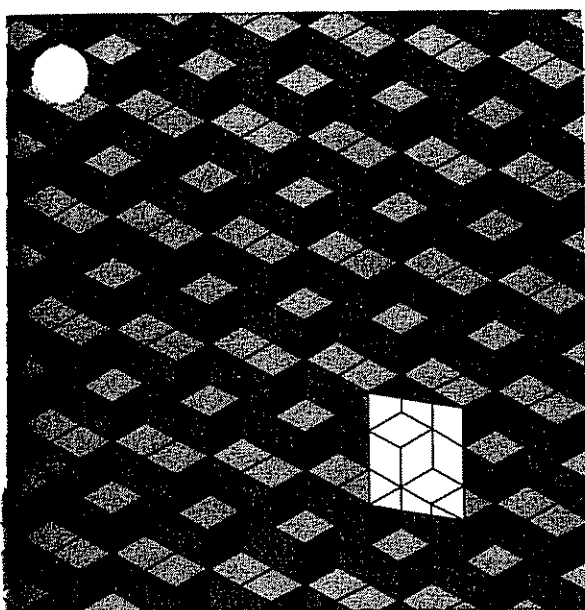
Penrose quasicrystals are constructed from a set of unit cells and specific matching rules that govern how they fit together. These rules are more complicated than the identical repetition of identical unit cells that form a crystal. Three important features distinguish

a Penrose quasicrystal from a crystal.

First, a Penrose quasicrystal contains many regions that explicitly show forbidden rotational symmetries, that is, fivefold. Second, a Penrose quasicrystal is built from two or more unit cells rather than a single unit cell as suffices for periodic crystals. Third, a Penrose quasicrystal does not exhibit equally spaced rows of lattice points as the periodic structures do. Nevertheless, the diffraction pattern resulting from a Penrose quasicrystal has an array of sharp spots, in agreement with the experimental observations.

One can elegantly describe the structure of Penrose quasicrystals and their corresponding diffraction patterns if one thinks of Penrose structures as resulting from a slice through a higher-dimensional periodic lattice. This concept is easiest to visualize in two dimensions.

Imagine a two-dimensional lattice composed of points that sit at the corners of squares in a grid. A horizontal line of these points is covered by a strip. If the covered points are projected onto a line that is parallel to the strip, the projected points will be equally spaced along the line. Those points define a periodic sequence because they divide the line into equal-size segments. To produce a quasiperiodic sequence, the strip must be tilted with respect to the lattice so that it has a slope equal to an irrational number [*see illustration on page 50*]. (A number is irrational if, when expressed as a decimal, no set of consecutive digits repeats itself indefi-

cell. The red pattern, which cannot be constructed from a single unit cell, is quasiperiodic. Whether a pattern is periodic or quasiperiodic depends on the angle of the cut. Whereas the slope of the green plane with respect to the stack of cubes and the slope of the blue plane are equal to rational numbers, the slope of the red plane is an irrational number.

nitely, for example, the number π, or 3.1415....) If the points that are covered by the strip are then projected onto a line parallel to the strip, the projected points will divide the line into a quasiperiodic sequence of long and short segments.

This sequence serves as a one-dimensional model of a quasicrystal if one imagines that an atom is placed at each point that divides the line into long and short segments. Like other quasicrystals, the quasiperiodic sequence has unusual diffraction properties. One might guess that the one-dimensional model would generate a blurry diffraction pattern because the atoms are not periodically spaced. But careful calculation proves otherwise. In fact, the quasiperiodic sequence produces a sharp diffraction pattern as a consequence of periodicity of the two-dimensional parent lattice.

The diffraction pattern derived from a quasiperiodic sequence consists of a dense set of weak and strong peaks [see illustration on page 50]. In experiments, one detects only the strongest of these peaks. Even so, one can still show that the quasiperiodic sequence is not a crystal because of the aperiodic spacing between the observable peaks.

To make quasiperiodic structures in two dimensions, one needs a lattice in e of even higher dimension. For ple, a plane that cuts through a stack of cubes at an angle can form a quasiperiodic two-dimensional structure if the slopes between the plane and the cube axes are irrational num-

bers [see illustration above]. (Indeed, a two-dimensional Penrose tiling that has fivefold symmetry can be produced by a projection from a five-dimensional lattice.)

Using similar reasoning, theorists have described icosahedral quasicrystals as a three-dimensional cut through an abstract six-dimensional lattice. This kind of cut leads to the type of quasiperiodic structures proposed in the Penrose model. By describing quasicrystals in this way, one can understand how quasicrystals can have unusual symmetries and yet not contradict the precepts of crystallography. Whereas the possible symmetries of the diffraction patterns of crystals are limited by the possible symmetries of the crystals themselves, the symmetries of quasicrystals derive from those of a higher-dimensional parent lattice.

In general, a quasiperiodic structure will give rise to a diffraction pattern that has great order because of the periodic order in the higher-dimensional parent lattice.

Although the Penrose model is very successful in predicting the diffraction patterns generated by icosahedral alloys, it gives few clues about how physical reality is related to multidimensional spaces and matching rules. In particular, although the Penrose matching rules are local, a great deal of planning ahead is required to construct a perfect Penrose quasicrystal. Growing such a quasicrystal requires that atoms in very distant

unit cells would have to interact in some manner to communicate their positions and relative orientations. This idea is contrary to all generally accepted notions about crystal-binding forces, which are relatively short range.

Another objection to the Penrose model is that it fails to account for the considerable disorder evident in almost all quasicrystals. This disorder appears in their structural, electrical and diffraction properties. For instance, one consequence of the Penrose model is that a perfect quasicrystal should conduct electricity as well as an ordinary metallic crystal. In fact, all quasicrystals produced in the laboratory have conducted electricity rather poorly.

A more important sign of disorder is revealed in the X-ray diffraction patterns from icosahedral alloys. In many cases, these show broadened peaks in contrast to the perfectly sharp peaks predicted by the Penrose model. Broadened diffraction peaks are a sign of disorder in many crystalline materials. Common sources of disorder—such as small grain size, defects or strain—produce well-known signatures in diffraction patterns. But none of these signatures seem to match the peak broadening exhibited by quasicrystals.

The X-ray diffraction results, in fact, point to a new form of structural disorder called phason disorder, which is unique to quasicrystals. If one compares a Penrose tiling or a quasiperiodic sequence with a conventional crystal, one sees that the quasiperiodic structures have the ability to generate a new
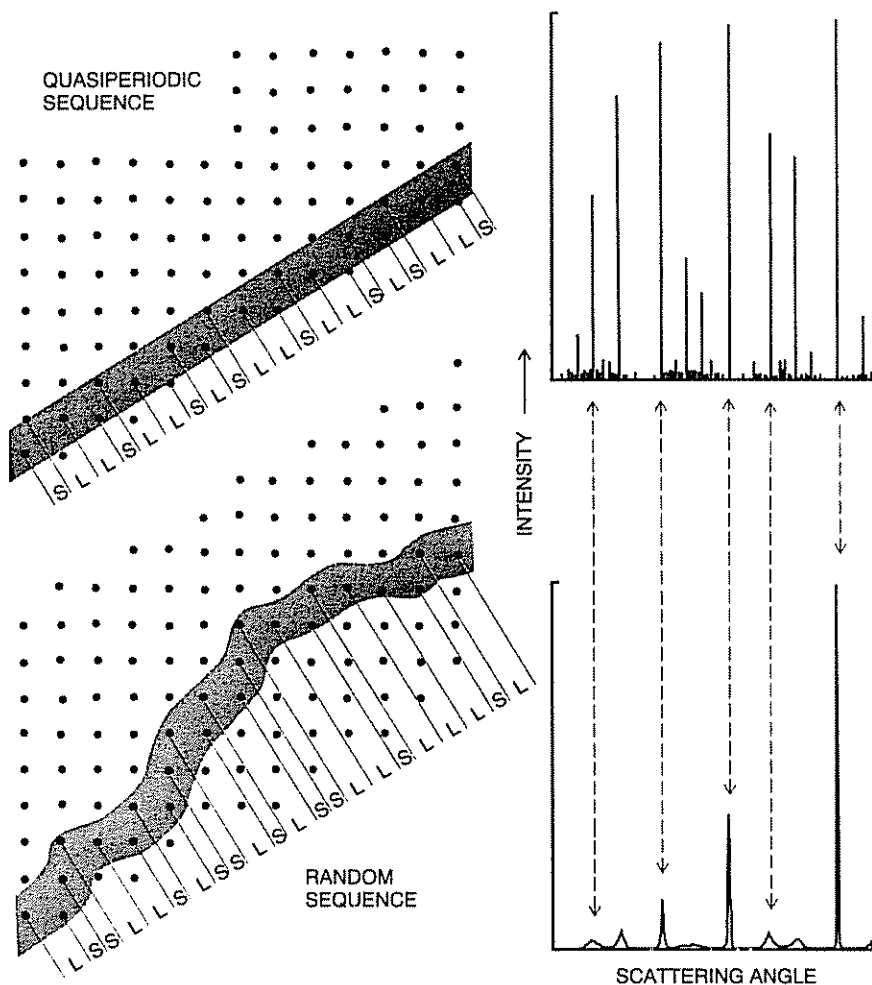
QUASIPERIODIC SEQUENCE of long and short segments (*top left*) is formed when a strip covers a lattice at a slope equal to an irrational number. Specifically, the slope is the inverse of the golden mean, or about 0.618. The sequence yields a sharp diffraction pattern (*top right*). A meandering path through the lattice produces a random sequence (*bottom left*). Despite this disorder, the diffraction pattern of the random sequence (*bottom right*) is similar to that of the quasiperiodic sequence.

form of disorder during growth: a defect can arise when the wrong kind of unit cell, or line segment, falls in a particular place. A few isolated mistakes will not affect the diffraction properties of an entire sample, but if many such mistakes plague a sample, they will disturb the diffraction patterns.

As an extreme example, imagine that the long and short segments in the one-dimensional quasiperiodic sequence are rearranged in a completely random fashion. Surprisingly, this random sequence gives rise to a diffraction pattern that is quite similar to the pattern derived from the original quasiperiodic sequence. The diffraction peaks of the random sequence are found at the same positions as those from the quasiperiodic sequence but are broader. In fact, the widths of these peaks are inversely related to the strength of the corresponding diffraction peak from the quasiperiodic sequence, so that only

the more intense peaks remain. Nevertheless, the existence of relatively sharp diffraction peaks from the random sequence indicates that quasiperiodicity can survive disorder.

Such ideas led the authors to suggest in 1986 that the icosahedral alloys had an inherently defect-ridden structure. Our proposal became known as the icosahedral glass model. Randomness is important to the glass model in two ways. First, it removes the necessity of arcane matching rules and gives a more plausible explanation for quasicrystalline growth. Second, the disorder introduced through randomness closely mimics that evidenced by the peak broadening of the diffraction patterns. Interestingly, soon after the discovery of quasicrystals, Shechtman and Ilan Blech of Israel Institute of Technology–Technion in Haifa suggested that icosahedral quasicrystals were composed of icosahedral clusters that were random-

ly connected. This theory was refined by the authors to the point where we could reproduce the experimental observation of diffraction peak broadening [*see illustration on page 53*].

The attractiveness of the glass model extends beyond its ability to incorporate disorder. Certain crystalline alloys contain icosahedrally symmetric atomic clusters, which are plausible building blocks for the glass model. The size of those clusters is within 1 percent of that required to match the experimental diffraction patterns.

As these theoretical ideas were being developed in the late 1980s, materials scientists and chemists were busy in their laboratories discovering dozens of new icosahedral alloys. Some of the materials were variations of the aluminum-manganese alloys, but investigators also synthesized new families, such as aluminum-zinc-magnesium, uranium-palladium-silicon and nickel-titanium.

One of the most important results to come from these new materials was the discovery that quasicrystalline phases could be thermodynamically stable. The quenching process initially used by Shechtman and others produced very small grained quasicrystalline phases that, when heated, transformed irreversibly into common crystalline phases. Unfortunately, this metastability prevented workers from improving the quality of samples by heat treatment and other metallurgical techniques. Hence, the first quasicrystals had grain sizes of only a few thousandths of a millimeter, making many kinds of experiments impossible.

Several materials discovered in the past few years, however, retained their quasicrystalline structure up to their melting point. Hence, workers could prepare much larger samples by conventional crystal growth techniques. In this way, they have recently made alloys that have single grains as large as 10 millimeters in size.

Astonishingly, when these first stable quasicrystals of aluminum, lithium and copper were grown slowly enough to form large, faceted surfaces, they still suffered from the same degree of phason disorder as did their cousins formed by quenching. The discovery of phason disorder in these materials seemed to support the icosahedral glass model.

Although the icosahedral glass model is more successful at predicting the diffraction patterns, it goes too far in its attempt to incorporate disorder. It leaves too many gaps or tears in the structure where icosahedral clusters

How to Grow a Penrose Tiling

A Penrose tiling can be constructed from two kinds of rhombuses that have been decorated with colored bands.

To make a Penrose tiling, one fits these rhombic tiles together according to the following "matching" rule: two rhombic tiles can be placed side to side only if bands of the same color join continuously across their interface. Some examples are shown below.

In growing a Penrose tiling, one starts with a single tile and adds each subsequent tile to the outer boundary. One assumes that once the tile has been positioned, it cannot be removed or shifted. (This growth process is somewhat similar to the formation of crystals, in which groups of atoms firmly attach themselves to the surface of a "seed" crystal.)

Growing a Penrose tiling is complicated by the fact that a given position on the boundary can often accommodate either of the two kinds of tiles. For example, a fat rhombus or two thin ones could be fit in the space at the top of these five tiles.

When these situations arise, one could choose among the possibilities at random. But such choices can lead to other problems. The rhombic tiles can be assembled so that they obey the matching rule but prevent other tiles from being added to the boundary. For example, neither a fat rhombus nor a thin rhombus can fit into the space at the left in the following configuration

In general, if a wrong choice is made, the tiling will stop growing at some later point.

Can one develop a procedure that will allow Penrose tilings to grow indefinitely? Following the work of John H. Conway of the University of Cambridge and Nicolaas G. deBruijn of Eindhoven University of Technology, George Onoda of the IBM Thomas J. Watson Research Center and co-workers have taken the first step toward a solution

To understand their method, one must first realize that each vertex in an infinite Penrose tiling is surrounded by one of eight combinations of tiles as shown below.
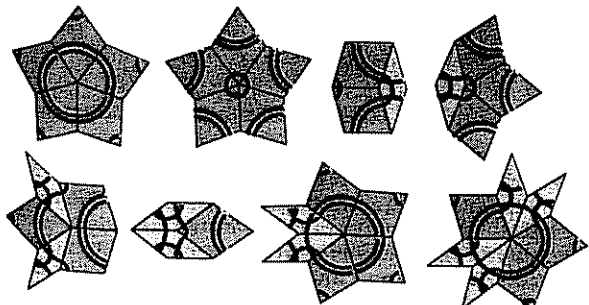
If a vertex on the boundary of a growing pattern will accommodate one or more tiles in only one way that is consistent with one of the eight vertices, then it is said that the vertex is forced. For example, the vertex indicated below by a dot is forced.

The boundary of a growing pattern will always contain at least one forced vertex if one starts with a particular seed in which the matching rules have been violated in certain places. Such a seed is represented by the white region below. The region contains two matching-rule violations that occur between the tiles shaded in light blue and yellow As tiles were added to forced vertices on the boundary of the white region, the tiling began to grow, and new forced vertices were created on the new boundary. (Each forced vertex on the boundary in the pattern below is labeled with an F.) Because this growth process can be continued indefinitely, the tiling can be enlarged to any size. It will be a perfect Penrose tiling except for the original defects in the seed.
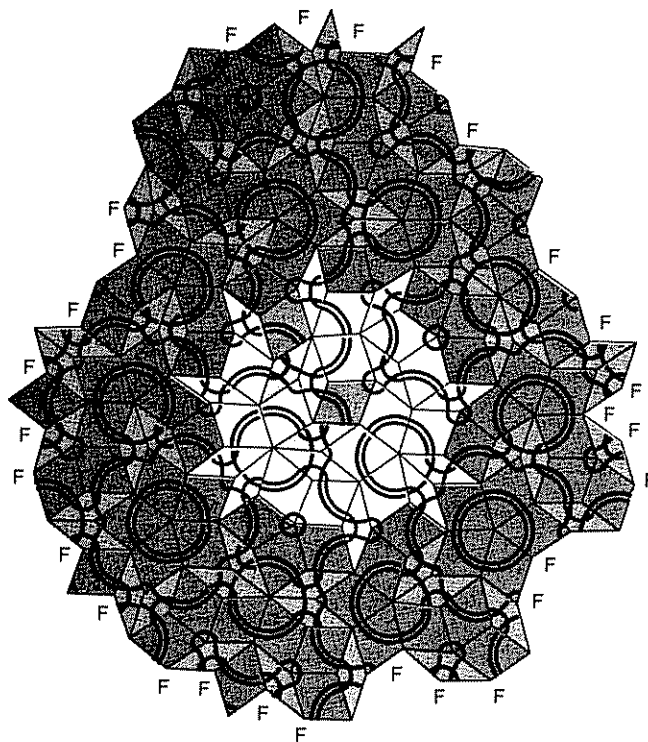
This procedure for growing Penrose tilings simulates only some aspects of real atomic growth One can sensibly assume that forced vertices on the boundary represent sites where atoms attach easily to some surface On the other hand, it seems unreasonable that some vertices will wait for an indefinite time before they become forced It also seems odd that although several violations of the matching rules must occur in concert to produce an appropriate seed, additional mistakes must be avoided Perhaps these issues will be resolved by further research
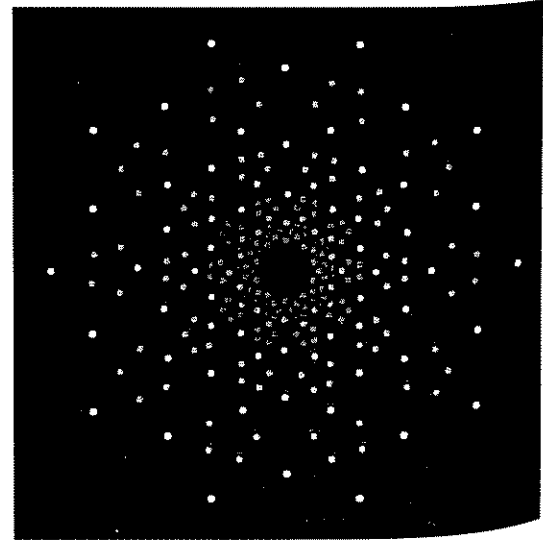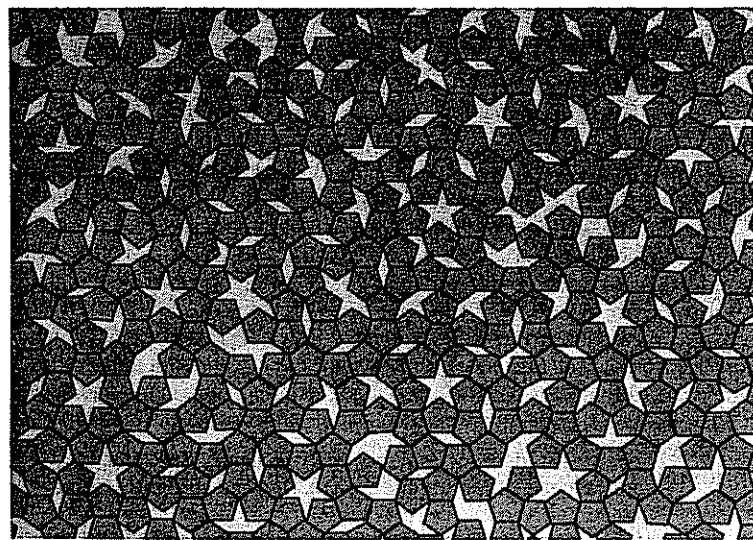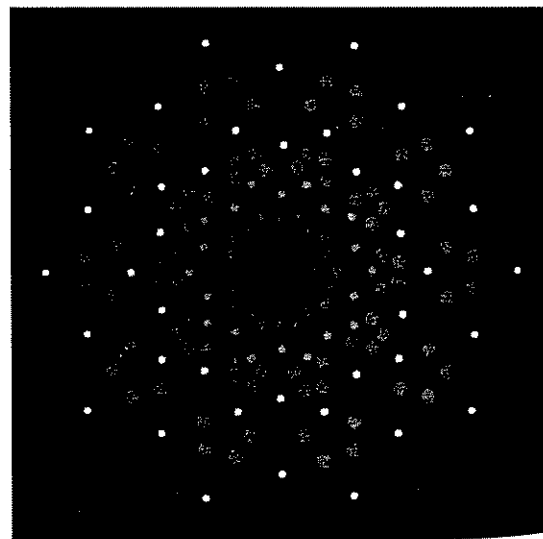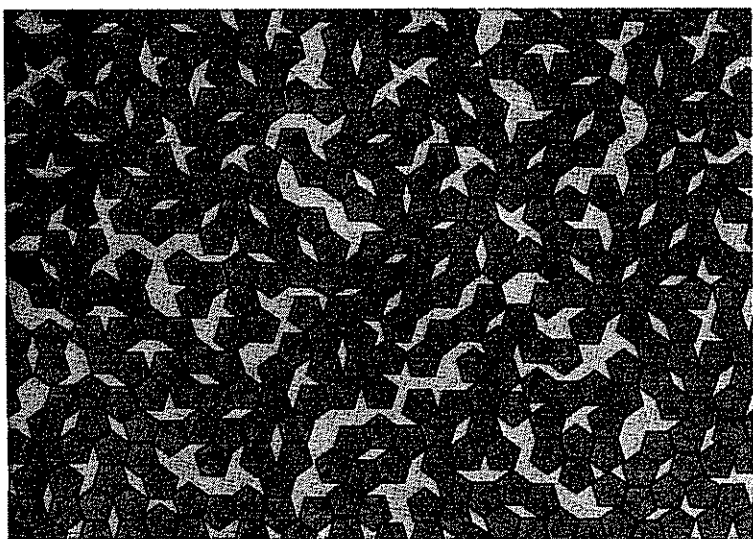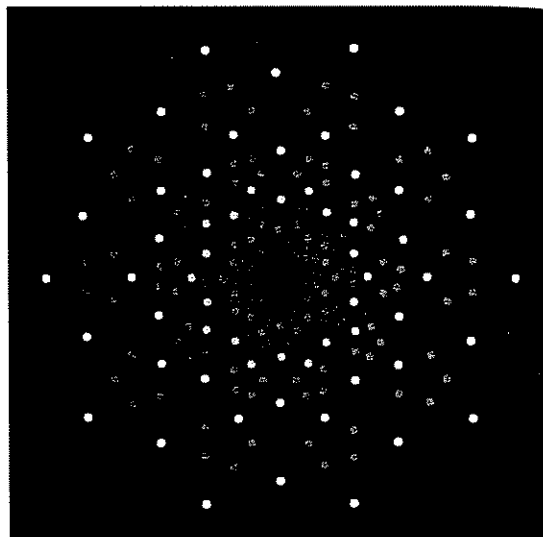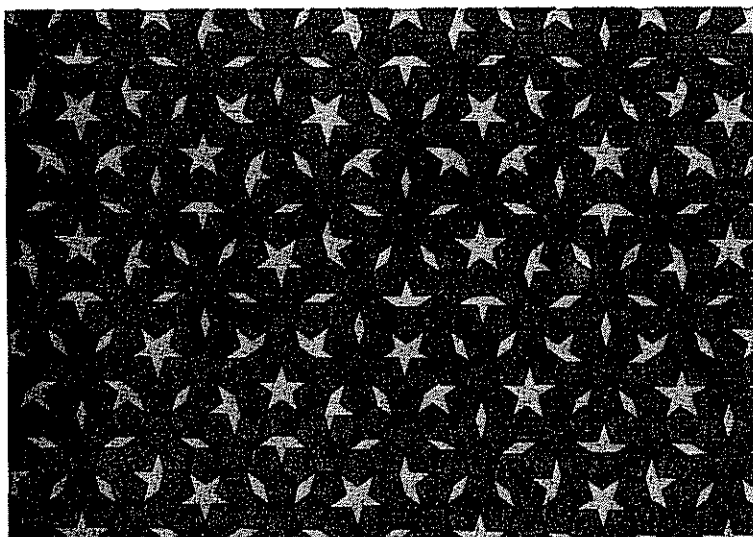
THREE MODELS have been proposed to account for the structure of quasicrystals and their diffraction patterns. The Penrose model is represented by the quasiperiodic lattice at the top left. The lattice is composed of pentagons, diamonds, stars and "boats" that have been assembled according to specific matching rules. The glass model yields the structure at the middle left. This structure is made of pentagons that have been stuck together, side by side, in a random way. The random-tiling model depicted at the bottom left is similar to the Penrose model, but it contains mistakes that are forbidden by the matching rules of the Penrose model. For each of these structures, one can calculate its diffraction pattern (*diagrams at right*), which can then be compared with experimental results. The Penrose and random-tiling model both yield sharp diffraction peaks, but the diffraction peaks from the random-tiling model (*bottom right*) are surrounded by faint halos.

cannot fit. These tears are, of course,
nt in the Penrose model. The net
t of the tears is that the glass mod-
el overestimates the degree of broad-
ening in diffraction patterns.

While the proponents of the Penrose
and glass models debated the impor-
tance of local growth and phason dis-
order, a third group of investigators de-
vised the random-tiling model, which
combines some of the best concepts
from the Penrose and the icosahedral
glass models. The random-tiling mod-
el suggests that the strict matching
rules of the Penrose model do not have
to be obeyed, as long as there are no
gaps left in the structure. Surprising-
ly, the random-tiling model predicts
perfectly sharp diffraction peaks, just
like its more ordered cousin, the Pen-
rose model.

The apparent advantage of the ran-
dom-tiling model is that it requires
only local growth rules. For instance,
Michael Widom, Katherine J. Strand-
burg and Robert H. Swendsen of Car-
negie-Mellon University demonstrated
that they could simulate the growth
of these defect-filled tilings by apply-
ing the same computer algorithms used
to simulate the growth of periodic crys-
Furthermore, they found that un-
certain circumstances the defect-
filled tiling was more thermodynamical-
ly stable than a normal crystal. These
investigators and Christopher L. Hen-
ley of Boston University demonstrat-
ed that the disorder associated with
errors in a perfect quasiperiodic struc-
ture can actually stabilize quasicrystal-
line order, at least with respect to some
competing crystalline phase. The rel-
ative importance of disorder increases
with temperature, so that the random-
tiling model predicts that quasicrystals
achieve the stable, equilibrium phase
only at elevated temperatures.

Over the past few years the three
competing models for the icosahedral
alloys have been refined to produce
closer agreement with experiments and,
in particular, diffraction data. For in-
stance, theorists introduced mecha-
nisms for producing disorder in ideal
quasiperiodic structures to mimic more
closely the broadened diffraction peaks.
They developed algorithms to grow
nearly perfect Penrose tilings by rules
that seemed more plausibly local. At
about the same time, Veit Elser, then at
AT&T Bell Laboratories, modified the
glass model by incorporating more
istic atomic motions during the
ulated growth of a quasicrystalline
grain; he found that the calculated dif-
fraction patterns in such a grain did
not exhibit excessive peak broadening
beyond the experimental results. All
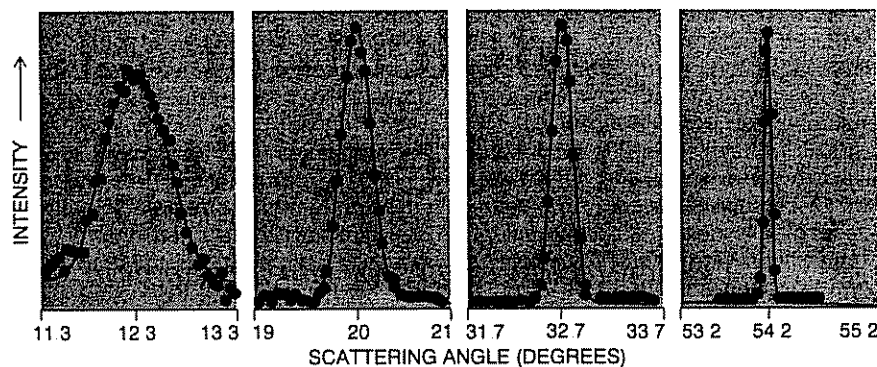
DIFFRACTION PROPERTIES of a quasicrystal composed of aluminum, copper and
lithium were revealed by bombarding the quasicrystal with a beam of X rays and
by measuring the intensity of the scattered X rays at various angles relative to the
beam. The graph shows that the diffraction peaks sharpened with increasing angle.
This sharpening is the signature of phason disorder, a kind of structural defect
found only in quasicrystals. The measurements were taken by Paul Heiney, Paul
Horn, Frank Gayle and their co-workers at the National Synchrotron Light Source.

these models were converging toward
a middle ground incorporating some
degree of phason disorder. To some ob-
servers, it seemed as though the differ-
ences between the models were dis-
solving into semantics.

A series of experiments performed
in 1989, however, essentially narrowed
the field of plausible models to two:
the Penrose and random-tiling models.
Physicists at Tohoku University in Ja-
pan discovered a new family of icosa-
hedral alloys, including aluminum-cop-
per-iron and aluminum-copper-rutheni-
um. Remarkably, when we took X-ray
diffraction patterns of these materials,
we found that the peak broadening as-
sociated with phason disorder—an ef-
fect evident in all previously studied
icosahedral alloys—was absent. Peter
Bancel of the IBM Thomas J. Watson Re-
search Center independently confirmed
these results and showed that as the
iron alloy was heated and cooled, pha-
son peak broadening could be en-
hanced or diminished.

I s quasicrystalline order produced
by the matching rules of the Pen-
rose model or by the freedom to
scramble local groups of atoms to al-
low limited phason disorder? In an at-
tempt to answer this question, sever-
al laboratories, including our own, are
continuing experiments to study the
conditions that control the perfection
of quasicrystals.

While some investigators have been
working to fathom the novel forms of
atomic order displayed by these ma-
terials, others have been developing ap-
plications. Yi He, S. Joseph Poon and
Gary J. Shiflet of the University of Vir-
ginia have used insights about quasi-
crystalline structure to synthesize me-

tallic glasses containing up to 90 per-
cent aluminum. They hope to exploit
the low density and unusually high
strength of these materials. Jean-Marie
Dubois and his colleagues at the Na-
tional School of Mining Engineering
in Nancy, France, have discovered that
certain quasicrystals produce excellent
low-friction coatings.

Quasicrystals continue to pose excit-
ing challenges for condensed matter
scientists. Nearly all the ideas that have
been developed to understand the elec-
tronic, thermal and mechanical proper-
ties of crystalline solids are based on
the simplifying framework of periodici-
ty. Now faced with quasiperiodic struc-
tures, we must seek more sophisticat-
ed levels of understanding.

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