Growing Perfect Quasicrystals

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We present an approach for aggregating Penrose tiles to form a defect-free, 2D pentagonal quasicrystal tiling. Contrary to conventional wisdom, defect-free quasiperiodic tilings can be constructed by use of local rules alone. The results provide new insights as to how materials with only short-range atomic interactions can grow large, nearly perfect quasicrystal grains.

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In this paper, we present a growth algorithm for aggregation of Penrose tiles[1] to form an infinite, defect-free perfect Penrose tiling (PPT). Only local (short-range) matching rules are used to determine how tiles are joined to a cluster, contradicting the standard lore[2] that long-range interactions are required to grow a PPT. Thus, a mechanism is suggested by which materials with only short-range atomic interactions can grow large, nearly perfect quasicrystal grains.

PPT's can be constructed from fat and skinny rhombi[1] (which are analogous to unit cells in a quasicrystal[3]) with edges marked with single and double arrows as in Fig. 1(a). The original Penrose matching rules constrain adjacent tiles to have matching arrow types and directions along their shared edge.[1,4] However, random addition of tiles at the surface of a cluster fails to produce a PPT even when Penrose's rules are obeyed. "Defects" —local-rule or geometric conflicts which deviate from the perfect quasiperiodic translational order of the PPT —occur after only a handful of tiles are added. The alignment of distant tiles is required to avoid defects,[5] and it was believed that only global (long-range) rules could ensure such alignment.[6] As a consequence, perfect quasicrystals have sometimes been viewed as physically unrealizable, a disappointing conclusion, if it were true, in view of the unique physical properties which such materials might have. Structural modeling for the icosahedral phases has turned towards random aggregation models,[7] and entropically stabilized random tilings.[8]

Here, we show that the PPT can be grown by use of only local rules. The rules are "vertex rules" which constrain the tiles sharing a common vertex to be one of the eight vertex configurations[4] found in a PPT (see Fig. 1). The constraints include but extend beyond Penrose's original edge rules. According to the vertex rules, a new tile can be added to an edge on the surface of a given cluster only if the configurations formed around each vertex of the new tile are consistent with at least one of the eight vertices. A vertex that is not yet completely surrounded is called a "forced vertex" if, for at least one of the free edges sharing the vertex, there is only one way of our adding a tile consistent with the vertex rules. The tiles determined in such cases are called "forced tiles."

To grow a PPT from a seed cluster, two additional rules must be followed. R1: *If one or more vertices are forced, randomly choose a forced vertex and add a forced tile to it.* Forced tiles are successively added in any order until the entire surface of the cluster is "dead," consisting only of unforced vertices. As discussed below, the dead surface forms a convex polygon (when viewed macroscopically) with at least two 108° corners. R2: *If there are no forced vertices, add a fat tile (consistent with the vertex rules) to either side of any 108° corner.*

The growth procedure has a natural physical interpre-

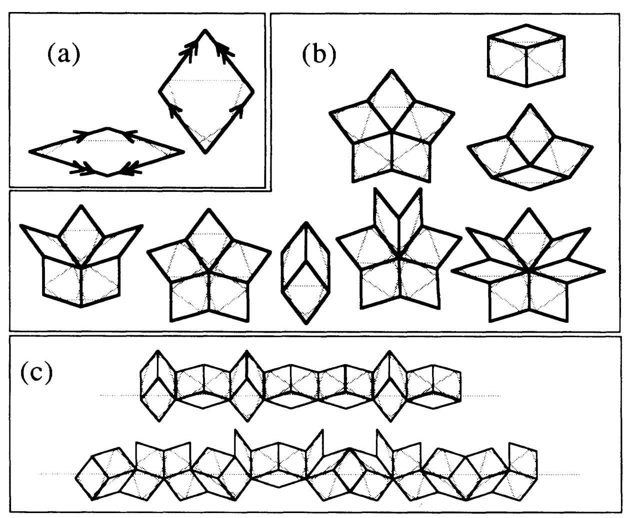

FIG. 1. (a) Fat and skinny Penrose (rhombic) tiles marked with Ammann lines. The edges are marked with double or single arrows (Ref. 4). (b) The eight possible vertices in a PPT. We require continuity of the Ammann lines across adjoining edges which assures that Penrose's edge rules are obeyed. (c) Segments of two rows in a PPT. Above is a "worm segment" and below is a generic row [see also Fig. 2(f)].

tation for real materials. The vertex rules can be interpreted as short-range interactions between atomic clusters that share a vertex. R1 could be approximated if forced vertices have a much larger sticking coefficient than unforced vertices. In sufficiently slow, diffusive growth, atoms would not stick to the surface until they hit a forced vertex. R2 could result if the sticking coefficient for the 108° corners is much less than those of the forced vertices, but much greater than those of the unforced vertices. Another rule we have explored is R2′: *If there are no forced vertices, add a fat or skinny tile (consistent with the vertex rules) to a randomly chosen unforced vertex.* Actually, this rule is sufficient to generate very large defect-free tilings ($\geq 2^{50}$ tiles). However, a rare combination of random selections leads to defects in the tiling, as discussed below.

We will next outline the proof, the details of which will be given in a later paper. As a preliminary, it is useful to decorate the tiles with the line segments shown in Fig. 1. In a PPT, the segments join to form five sets of infinite, parallel lines, known as Ammann lines, oriented along each of the pentagonal directions.[3,9] In each set, the spacings between lines form a Fibonacci sequence of long (L) and short (S) intervals. The tiles that intersect a given Ammann line form a "row." Each tile intersects one line in each pentagonal direction.

The vertex rules have several features that force a cluster to grow by the successive addition of rows of tiles. First, they ensure the continuity of the Ammann lines in the tiles around any vertex, which at the same time assures the correct alignment of distant tiles.[5] When a forced tile is added to an edge, any Ammann lines in the cluster that pierce the edge are extended through the new tile. The same tile may extend existing lines in some directions at the same time that it initiates new lines in other directions. Second, these rules ensure that a new row grows by a sequence of forced moves until it extends at least to the ends of the adjacent, parallel row.

Suppose row I lies along the surface of a cluster and a new tile is added to a vertex in the middle of it. Every Ammann line through the new tile also passes through row I and is fixed in the cluster, except for the one parallel to row I. The new tile initiates a new Ammann line and, hence a new row, row II, parallel to row I. On each side of the new tile is an incomplete vertex where the new tile and row I join. The tiles that complete the vertex are forced: Row I fixes four of the Ammann lines that pierce them and the new tile fixes the fifth. Whenever all five lines are fixed, only one tile choice can fit, and it is forced by the vertex rules. These tiles produce further forced vertices where they adjoin row I, etc., until row II reaches the ends of row I. At those points, the tiles in row I no longer fix the Ammann lines in four directions. Further tiles beyond the ends may be forced in some circumstances (see the discussion of corners below). The completed row II results in a new set of incomplete vertices on the boundary. If at least one of these vertices is forced, the associated forced tiles initiate a new row, row III, oriented parallel to rows I and II. The process repeats until a row is reached whose surface consists only of unforced vertices.

Next, we can show that a dead surface consists of straight sections joined at corners. In order for the sequence of forced moves to stop, every Ammann line must continue uninterrupted across the cluster, and no new Ammann lines must be forced beyond the surface. An unforced tile randomly added to the surface necessarily initiates a new Ammann line that does not yet appear as part of the existing cluster. This is not possible unless the new line does not intersect the surface; that is, the surface must be parallel to the new Ammann line and have only "microscopic" fluctuations (i.e., no larger than S), so that it is "macroscopically" flat. At corners, a flat surface in one direction ends and a flat surface in a new direction begins. An unforced tile at the corner can initiate two Ammann lines, one in each direction.

The configurations of tiles along a dead surface is very special. At any site along the surface, either a fat or a skinny tile can be added consistent with the vertex rules. If a skinny tile is added to a given edge, it initiates a new Ammann line parallel to the edge and fixes the spacing, L or S, between that line and the nearest parallel line in the cluster. This in turn forces a row of tiles along the new Ammann line. If a fat tile is placed along the same edge, the opposite spacing is fixed and a different row of tiles is created along the flat surface. The two rows are related by a "flip" reflection through a line parallel to the row. The only difference in the Ammann lines that pass through the two rows is the position of the new line parallel to the surface. Lines in the other four directions pass through the dead surface and, hence, are already fixed by tiles inside the cluster. The row of tiles that has these properties is already known, referred to as a "worm segment" in the literature.[10,11] A worm segment [Fig. 1(c)] has the unique property that both of its sides have the same geometric shape and arrow directions.

On a macroscopic scale, dead surfaces form a convex polygon whose sides lie at the borders of finite worm segments. A PPT has an infinite number of arbitrarily long, finite worm segments.[10,11] A tile added to any side of the dead surface forces an entire, finite worm row to grow along it, extending to the corners; it determines which way the worm is flipped; and it determines a finite number of additional rows parallel to the worm. Furthermore, it may force tiles at the corner, which, in turn, may begin to force tiles along the adjoining edge of the dead surface. Forced tiles continue to be added until a new dead surface is reached. On average, a cluster roughly doubles in size from one dead surface to the next (see Fig. 2). In some cases, the old dead surface lies totally within the new dead surface; in other cases, one or more edges of the old dead surface remain as part of the new dead surface.

The possible dead polygonal surfaces can be cataloged by our noting how worm segments join to form a closed surface in a PPT. In order to obtain a dead polygonal surface, dead surfaces that border the worm segments on the interior must meet to form "dead corners." There are two types of 72° dead corners (related by mirror symmetry) and two types of 108° dead corners. (Also, two 108° dead corners can merge to form a 36° dead corner, an uninteresting, degenerate case.) The only possible closed polygons with 72° and 108° corners have either (a) two 72° and two 108° corners; or (b) five 108° corners. The problem of cataloging all dead polygonal surfaces is greatly simplified by the observation that any dead surface remains dead under the inflation transformations introduced by Penrose.[1] That is, if the tiles enclosed by a given dead surface are inflated to produce fewer, larger tiles, the inflated tiles that lie totally within the original dead surface form a new dead surface of the same shape. A dead surface can be repeatedly inflated until the cluster consists of only a small number of tiles. The observation allows a complete catalog of dead polygonal surfaces to be constructed from an exhaustive search of clusters containing less than about 50 tiles.

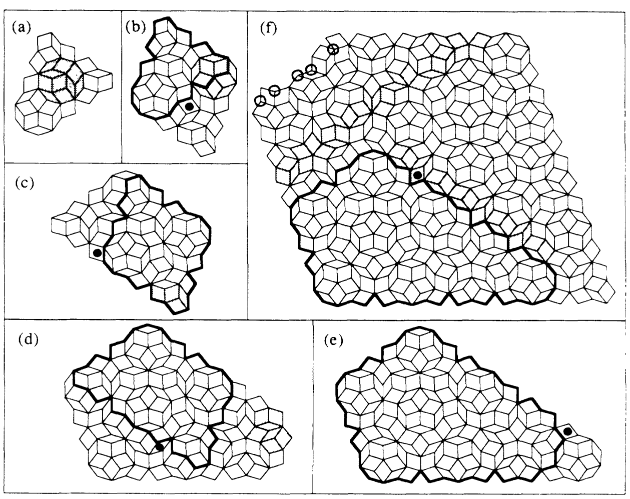

FIG. 2. A succession of clusters grown from a seed consisting of ten tiles [shaded in (a)]. In (a), forced tiles are added to the seed according to R1 until the first dead surface is reached. In succeeding panels, an unforced tile (black dot) is added to the dead surface in the preceding panel (heavy line); then, forced tiles are added until the next dead surface forms. Panels (c) and (e) illustrate how to add the unforced tiles according to rule R2; in (b), (d), and (f), rule R2′ is used. We refer to (d) as the "coffin" shape. In (f), the dead surface is not quite complete; forced vertices (encircled) remain along the top left surface. In different panels, tiles are shaded to illustrate configurations discussed in the text: the two types of 108° corners are shaded in (b) and (d); a 72° corner is shaded in (c); a generic row (upper left) and a worm segment (along the heavy line) are shaded in (f).

To establish the sufficiency of R2, we have also cataloged the "empires,"[10] the set of forced tiles following a random choice, as a function of dead surface polygonal shape, choice of random edge, and choice of worm orientation forced by the random choice. The catalog is simplified by the fact that, if two dead surfaces are similar (in the strict geometrical sense), then their empires are similar. In particular, to analyze a large tiling, one can inflate it many times to produce an arrangement with just a few tiles that can be easily analyzed.

The catalog shows that almost any choice of tile added to a dead surface forces tiles out to a larger dead surface containing a proper Penrose tiling. Thus, growth rule R2′ is almost sufficient to ensure a PPT. The exceptions are coffin-shaped dead surfaces [see Fig. 2(d)] or surfaces obtained by our adding to the coffin shape but leaving its short edge on the surface. If a random choice is made along the short edge, it can force a worm segment flipped one way or the other. However, only one choice can be combined with the rest of the tiles along the length of coffin to produce a PPT. The correct choice produces a legal Fibonacci sequence of Ammann lines parallel to the short edge, such as *LSLSLL*; the incorrect choice produces an illegal choice, such as *LSLSLS*. With use of R2′, the tiling typically contains 2^{50} tiles before such an error is encountered. [Unforced tiles are only added when the surface is dead. The probability of our obtaining an exceptional dead surface ($\approx \frac{1}{5}$), choosing an unforced vertex along the short edge ($< \frac{1}{5}$), and choosing the wrong unforced tile ($\frac{1}{2}$) combine roughly to $\frac{1}{50}$, and the tiling roughly doubles in size from one dead surface to the next.] The catalog reveals that growth rule R2, which forces tiles at the corners that bound the short edge, guarantees that the wrong choice is never made. This concludes the outline of the proof.

An important corollary is that there exists a class of pointlike defects which are ideal seeds for the growth of PPT's. If a cluster contains one of these defects, a dead surface is never obtained, and the tiles are forced throughout the plane. In terms of our physical analogy, a quasicrystal grain with one of these defects would always have sticky regions on its surface which promote perfect grain growth. The defects are a subclass of illegal Penrose tile arrangements known as "decapods."[10] One can assign a "charge" to a decapod defect using the Penrose arrow rules: Consider any closed path around the decapod running along the tile edges; add $+1$ if the arrow is along the path and -1 if the arrow is opposite the path. The net charge about any closed path in a PPT is zero. A dead surface can have charge $0, \pm 2$: (a) Any dead surface edge has charge zero; (b) all 108° corners have charge zero; and (c) 72° corners have charge ± 1. Recall that a dead surface can have at most two 72° corners. Decapod defects, though, can have charges ranging from -10 to 10. Thus, all decapod defects with charge greater than 2 in absolute value can never be enclosed by a dead surface. The other decapods have interesting properties that will be detailed in our later paper.

We have constructed a series of computer programs to test the prescription. The programs successfully produce undefected tilings with up to 10^6 tiles, limited only by central processing unit time. We are presently extending this analysis to the case of 3D icosahedral quasicrystals. There exist 3D tiles with many of the properties of the 2D Penrose tiles, including the generalization of Ammann lines to Ammann planes.[9] We expect most of the 2D analysis to be valid in 3D, but ensuring the construction of an infinite undefected tiling depends upon details involving edges and corners. Already, though, the present work dispels the myth that it is impossible to grow perfect quasicrystals by utilization of only short-range interactions.

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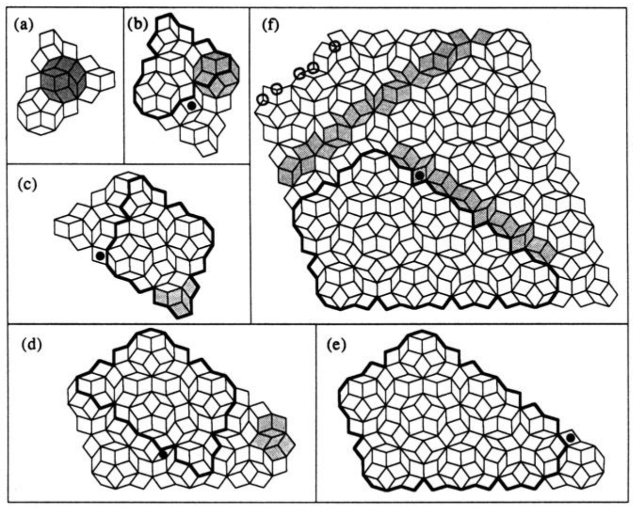

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