Dense quasiperiodic decagonal disc packing

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An inflation rule is given which proves the existence of infinite decagonal rectangle-triangle tilings. Such tilings correspond to the maximum possible density of a decagonal disc packing and can be used to model decagonal quasicrystals. A lower bound of 0.0336 per vertex is found for the entropy density of the corresponding random-tiling system. The atomic surfaces for a deterministic version of the packing are self-similar and disconnected. The relationship between rectangle-triangle tilings and real decagonal phases is discussed. It is shown that the large unit cell associated with observed microcrystalline phases has a natural explanation in the context of rectangle-triangle tilings.

I. INTRODUCTION

The problem of quasiperiodic disc or sphere packing is of great interest in the field of quasicrystals. For example, in a model for a quasicrystal where nonoverlapping clusters of atoms are energetically favored, the most stable structures are expected to correspond to maximum-density cluster packings. Such packings map naturally onto tilings, where the vertices of the tilings represent centers of clusters and the edges represent the linkages between neighboring clusters.

Thus the problem becomes one of maximizing the density of vertices on a tiling, subject to quasiperiodic symmetry and with edges belonging to a specified set of vectors. Perhaps the simplest such model for planar quasicrystals with n-fold symmetry (n = 8,10,12) is to limit the shortest edges to the set of n directions $\{2\pi i/n\}$. The solutions to the dodecagonal and octagonal disc packing problems are known: in the dodecagonal case, it is a tiling of equilateral triangles and squares;[1–3] in the octagonal case, it is a tiling of squares and various "hexagons."[4] In each case, there is a finite entropy density associated with various ways of arranging the same tiles under quasiperiodic symmetry.[4–8] Furthermore, deterministic versions of the packing yield atomic surfaces in the cut description[9] whose boundaries are *fractal*.[2–4,10,11]

Here we give an inflation rule which produces a tiling corresponding to a maximally dense decagonal disc packing. Furthermore, it has features in common with the octagonal and dodecagonal cases: there is a finite entropy density associated with different ways of arranging the tiles and, for deterministic inflation, the atomic surfaces that result are self-similar. The paper is arranged as follows. Section II describes the inflation rule and gives the calculation of the entropy associated with random inflation. In Sec. III, the connection is made between this model and real decagonal quasicrystals to conclude, in Sec. IV, that certain decagonal phases are unlikely to have simple atomic surface shapes in the cut description.

II. DECAGONAL RECTANGLE-TRIANGLE TILINGS BY INFLATION

This section begins with a summary of the notation used and a review of previous work on the decagonal disc packing problem. The following projections of the basis vectors in a four-dimensional representation are used:

$$\mathbf{e}_i^{\parallel} = a\,[\cos(2\pi i/5), \sin(2\pi i/5)], \tag{1}$$

$$\mathbf{e}_i^{\perp} = a\,[\cos(6\pi i/5), \sin(6\pi i/5)], \tag{2}$$

for $1 \le i \le 4$. The golden mean τ is defined as $(1+\sqrt{5})/2 \approx 1.618$. Associated with a vertex at position $\sum_{i=1}^{4} n_i \mathbf{e}_i^{\parallel}$ is its *perp space coordinate* $\mathbf{x}^{\perp} = \sum_{i=1}^{4} n_i \mathbf{e}_i^{\perp}$ and its *level* $\nu = (\sum_{i=1}^{4} n_i)_{\mathrm{mod}5}$. For convenience, levels 3 and 4 are alternatively called levels -2 and -1, respectively. The network formed by connecting points with $\nu = 0$ separated by distance $b = 2a\sin(\pi/5)$ is called the *zero-level network*. The set of $\{-\mathbf{x}^{\perp}\}$ for the level ν vertices delimits the level ν *atomic surface*. The decagonal disc packing problem is to maximize the density of vertices of a tiling subject to decagonal quasiperiodicity and with edges in the directions $\{\pm\mathbf{e}_i^{\parallel}\}$.

The standard Penrose rhombus tiling (2D PT) is illustrative. The edges all belong to the required set of near-neighbor vectors. However, the short diagonal of the thin rhombus, or "c bond,"[12] is of length $a/\tau < a$, and, thus, at least one vertex on each c bond must be removed to obtain a valid disc packing. Henley[12] eliminated some vertices of the 2D PT to obtain a packing fraction $f \approx 0.7386$. However, considering only tilings where the vertices form a subset of vertices of a tiling of the two Penrose rhombi, the highest density is achieved when the c bonds always form chains of even length m.[13] Then, the removal of $m/2$ vertices is sufficient to eliminate all short distances. The tiling thus formed will consist of fat Penrose rhombi and various hexagons. If edges of length b are drawn, the tiling will have only

two distinct tiles: a rectangle and an isosceles triangle (Fig. 1). Symmetry then fixes the packing fraction to be $(\pi\tau^3)/[8\sqrt{5}\cos(\pi/10)] \approx 0.7822$.[13] This packing fraction cannot be achieved by eliminating vertices from the 2D PT because it contains odd-length chains of c bonds.

The standard *binary decagonal tiling*[14,15] forms a better starting point. All of the vertices have $\nu = -1, 0$, or 1, thus it is a *three-level* tiling. Olamy and Kléman[16] found a three-level tiling, whose vertices are a subset of the binary tiling, corresponding to $f \approx 0.7627$. This tiling achieves the highest packing fraction of any known pentagonal structure with simple polygonal atomic surfaces. Mihalkovič[13] showed that, starting from the binary tiling, a packing fraction of about 0.7719 can be achieved by optimally eliminating vertices on the c bonds. However, the binary tiling still suffers from odd-length chains of c bonds. A maximally dense decagonal packing must therefore contain vertices not present in the binary tiling.

An inflation rule that leads to a three-level tiling with the maximum packing fraction is shown in Fig. 2. The linear scale of the inflation rule is $\tau^6 \approx 18$. There are four units involved in the inflation procedure: (1) the level 0 vertices (L vertices), (2) the level ± 1 vertices (S vertices), (3) the b edges on the zero-level network (b_{LL} edges), and (4) the other b edges (b_{SS} edges). As shown in Fig. 2(a), a D tile is drawn on each L vertex, a P tile on each S vertex, an X tile on each b_{LL} edge, and a Y tile on each b_{SS} edge. The interior of each tile is then filled as in Fig. 2(b) to yield the inflated tiling. The filling is independent of neighboring tiles; an inflation rule of smaller linear scale which has this property seems unlikely, but has not been ruled out. Note, however, that only the even powers of τ map the set of three levels onto itself.

There is still a good deal of choice possible in the inflation rule. Each small decagon containing ten triangles and five rectangles can be filled in two ways differing by a rotation of π; furthermore, the P, D, X, and Y tiles can all be filled in different symmetry-equivalent ways. The calculation of the entropy density due to such random choices in inflation is straightforward; the details (applied to the case of random Stampfli inflation[1]) have been given elsewhere.[7] The relevant parameters for the inflation rule presented here are given in Table I; these

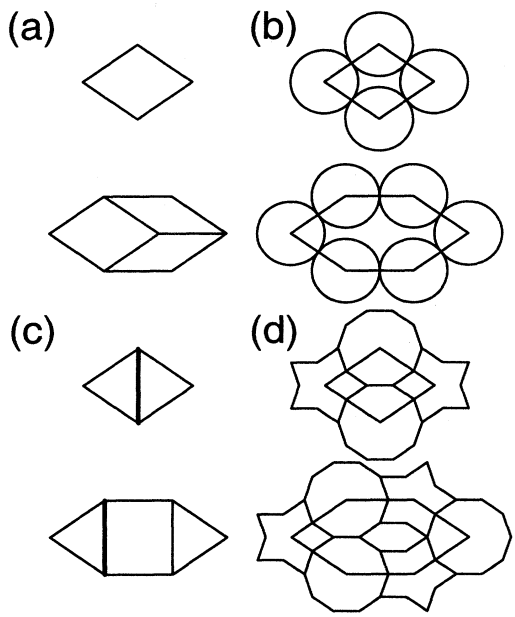

FIG. 1. Comparison of (a) two Penrose rhombus, (b) rhombus-hexagon (with disc packing shown), (c) rectangle-triangle and (d) Hiraga-Sun decagon-pentagonal star-hexagon tilings (Ref. 18). The zero-level network in (c) is highlighted. Longer "hexagons," containing adjacent rectangles, are not ruled out by packing considerations, but do not arise from the inflation rule in Fig. 2.

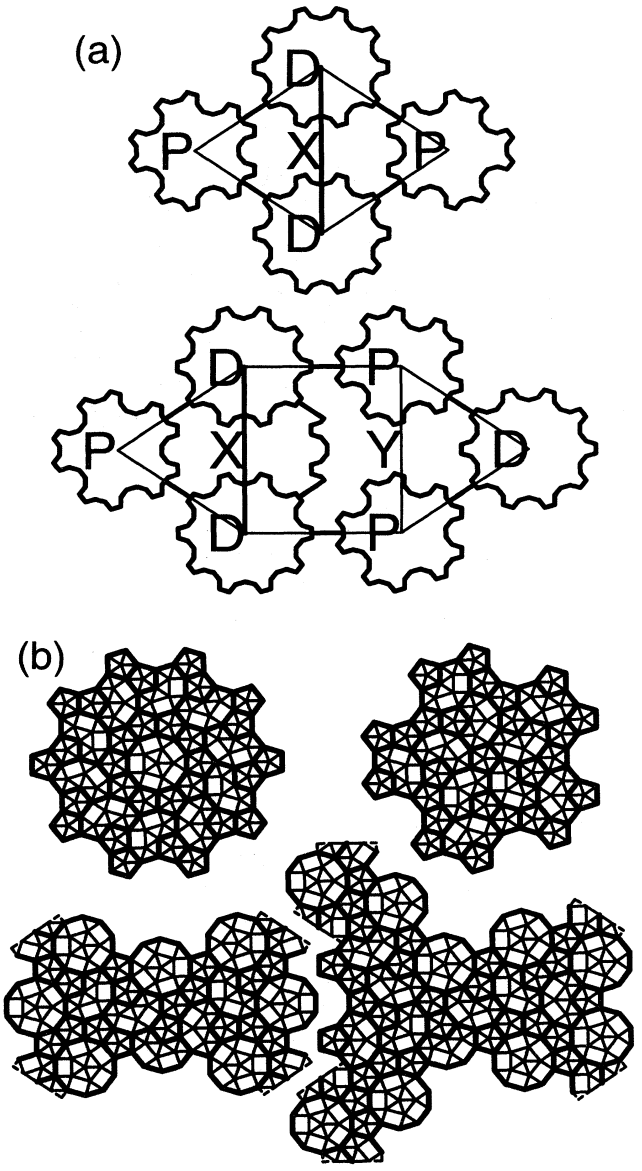

FIG. 2. The inflation rule: (a) division of rhombus and hexagon into D, P, X, and Y tiles, (b) filling of inflation tiles with rectangles and triangles. The zero-level network is highlighted.

TABLE I. Parameters for inflation.

Vertex or edge	No. per vertex	Inflaton tile	No. independent fillings
L vertex	$1-5^{-1/2}\approx 0.5528$	D	4
S vertex	$5^{-1/2}\approx 0.4472$	P	2
b_{LL} edge	$2\tau^{-2}\approx 0.7639$	X	512
b_{SS} edge	$\tau^{-3}\approx 0.2361$	Y	1024

parameters, combined with the inflation scale of τ^6, yield the random inflation entropy density per vertex:

$$\frac{S}{N}=\frac{(10-5^{-1/2})\tau^{-12}+2\tau^{-13}}{1-\tau^{-12}}\approx 0.03361. \qquad (3)$$

This sets a lower bound on the entropy density of the decagonal rectangle-triangle random tiling system.[17]

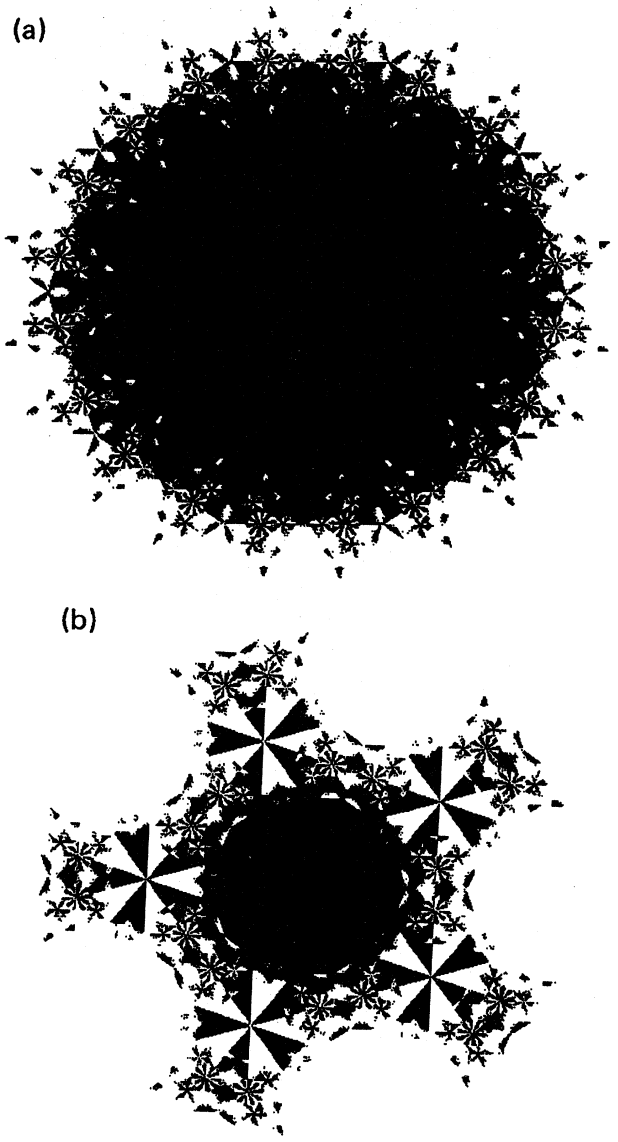

FIG. 3. Atomic surfaces for (a) level 0 vertices and (b) level 1 vertices. The atomic surface for level -1 vertices is the inverse of that for the level 1 vertices.

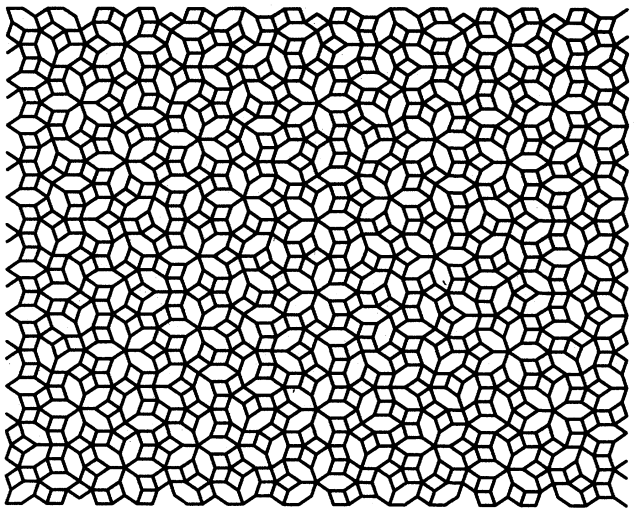

FIG. 4. Periodic approximant to decagonal rectangle-triangle tiling. For clarity, only the edges of length a are drawn.

To illustrate possible atomic surfaces for the model, the inflation rule was formulated in a deterministic way: each X and Y tile had its interior filled in a way that minimized perp-space variance $\sum(\mathbf{x}^{\perp})^2$, and the interiors of small decagons, D tiles and P tiles were filled in a deterministic way depending on the sector of perp space that the central vertex occupied. The result is shown in Fig. 3 for the level 0 and level 1 atomic surfaces. Note the complicated shapes containing self-similar features, "pinwheels,"[2,3] and disconnected pieces. A periodic approximant of the corresponding tiling, containing 1364 vertices, is shown in Fig. 4.

III. RECTANGLE-TRIANGLE TILINGS AND DECAGONAL PHASES

Oxborrow and Mihalkovič[17] have shown that the bright ring contrasts of certain high-resolution electron micrograph (HREM) images of decagonal Al-Pd-Mn (Refs. 18–21) can be interpreted as a zero-level network of linkage length $b\approx 20$ Å. More remarkably, when the edges of the corresponding rhombus tiling ($a\approx 17$ Å) are drawn, the chains of c bonds are almost exclusively of even length. Thus, these images can be interpreted as nearly defect-free rectangle-triangle tilings. Note that the zero-level vertices are occupied by one type of columnar cluster of decagonal symmetry and the $\nu=\pm 1$ vertices by a second type of columnar cluster of pentagonal symmetry.[19] Since the radii of the two types of cluster are approximantly equal, packing considerations still apply.

The smallest (three-level) periodic approximant possible in this model is a centered-orthorhombic phase of unit cell $\sqrt{5}\tau a$ by b which has recently been observed in Al-Pd-Mn (Refs. 21 and 22) [see Fig. 5(a)]. The rhombic unit cell associated with the Al-Cu-Co(-Si) (Refs. 23 and 24) and Al-Ni-Co (Ref. 25) "microcrystalline" phases, while

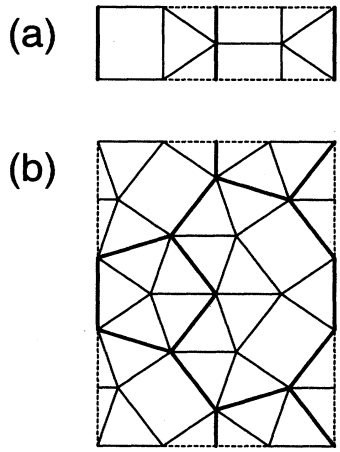

FIG. 5. Small rectangle-triangle approximants: (a) $\sqrt{5}\tau a$ by b, (b) $\sqrt{5}\tau a$ by $\tau^3 b$ (fundamental cell rhombic, edge $= \tau^2 b$ and $\gamma = 108°$).

seemingly very large (edge length $\tau^2 b \approx 51$ Å, $\gamma = 108°$), is actually the smallest known approximant containing a *connected* zero-level network [see Fig. 5(b)]. The microcrystalline phase might thus be explained as the result of an instability transition[26] of a quasicrystal to a dynamically accessible twinned approximant state, both based on clusters of approximately 20 Å diameter. Interestingly, an approximant phase with a 36° rhombic unit cell of edge $\tau^2 b$ is not tilable by rectangles and triangles (due to an odd number of thin 17 Å Penrose tiles per unit cell). This cell (which is rarely observed) would naturally be relatively unstable in the context of the rectangle-triangle model.

IV. CONCLUSION

This work (1) gives an inflation rule which proves the existence of infinite decagonal rectangle-triangle tilings, which have been shown to correspond to maximum density decagonal disc packings, (2) provides a lower bound on the entropy of this tiling system, and (3) reiterates the viewpoint that some real decagonal phases are based on tilings of the rectangle and triangle.

The atomic surfaces for deterministic versions of the inflation rule are self-similar and disconnected. It is unlikely that the exact set of atomic surfaces shown in Fig. 3 correspond to any physical atomic structure. Real structures are more likely be to based on *random*[17] rectangle-triangle tilings; randomization will cause a "blurring" of the atomic surfaces. In any case, decagonal structures based on the rectangle-triangle tiling should have atomic surfaces that cannot be described by simple shapes such as polygons.

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[1] P. Stampfli, Helv. Phys. Acta **59**, 1260 (1986).
[2] A. P. Smith, J. Non-Cryst. Solids **153&154**, 258 (1993).
[3] E. Cockayne, Phys. Rev. B **49**, 5896 (1994).
[4] E. Cockayne, J. Phys. A **27**, 6107 (1994).
[5] P. W. Leung, C. L. Henley, and G. V. Chester, Phys. Rev. B **39**, 446 (1989).
[6] M. Widom, Phys. Rev. Lett. **70**, 2094 (1993).
[7] M. Oxborrow and C. L. Henley, Phys. Rev. B **48**, 6966 (1993).
[8] P. A. Kalugin, J. Phys. A **27**, 3599 (1994).
[9] P. Bak, Scr. Metal. **20**, 1199 (1986).
[10] F. Gähler, Ph.D. thesis, Swiss Federal Institute of Technology, ETH, Zürich, 1988.
[11] M. Baake, R. Klitzing, and M. Schlottmann, Physica A **191**, 554 (1992).
[12] C. L. Henley, Phys. Rev. B **34**, 797 (1986).
[13] M. Mihalkovič, in *Aperiodic '94, an International Conference on Aperiodic Crystals*, edited by G. Chapuis (World Scientific, Singapore, in press).
[14] F. Lançon and L. Billard, J. Phys. (Paris) **49**, 249 (1988).
[15] M. Baake, P. Kramer, M. Schlottmann, and D. Zeodler, Int. J. Mod. Phys. B **4**, 2217 (1990).
[16] Z. Olamy and M. Kléman, J. Phys. (Paris) **50**, 19 (1989).
[17] M. Oxborrow and M. Mihalkovič, in *Aperiodic '94, an International Conference on Aperiodic Crystals*, edited by G. Chapuis (Ref. 13).
[18] K. Hiraga and W. Sun, Philos. Mag. Lett. **67**, 117 (1993).
[19] K. Hiraga, J. Non-Cryst. Solids **153&154**, 28 (1993).
[20] Figure 5 in C. Beeli and S. Horiuchi, Philos. Mag. B **70**, 215 (1994).
[21] Figures 17 and 18 in M. Duneau and M. Audier, in *Lectures on Quasicrystals*, edited by F. Hippert and D. Gratias (Les Editions de Physique, Les Ulis, France, 1994).
[22] C. Beeli, P. Stadelmann, R. Lück, and T. Gödecke (unpublished).
[23] M. Fettweis, P. Launois, F. Dénoyer, R. Reich, and M. Lambert, Phys. Rev. B **49**, 15 573 (1994).
[24] R. Wittmann, M. Fettweis, P. Launois, R. Reich, and F. Dénoyer, Philos. Mag. Lett. **71**, 147 (1995).
[25] M. Kalning, S. Kek, B. Burandt, W. Press, and W. Steurer, J. Phys. Condens. Matter **6**, 6177 (1994).
[26] C. L. Henley, in *Quasicrystals: The State of the Art*, edited by P. J. Steinhardt and D. P. DiVincenzo (World Scientific, Singapore, 1991).