Matching Rules and Growth Rules for Pentagonal Quasicrystal Tilings

Kevin Ingersent and Paul J. Steinhardt
Department of Physics, University of Pennsylvania, Philadelphia, Pennsylvania 19104
(Received 18 September 1989)

We show that finite-range matching rules and growth rules exist for an infinite set of 2D pentagonal quasicrystal tilings *other than* the original Penrose tilings. There is a natural ordering of these tilings based on the range required for the rules. The results have implications for the determination of the atomic structure of icosahedral alloys.

PACS numbers: 61.50.Em, 61.55.Hg, 64.70.Pf

In order to reconstruct the atomic structure of a quasicrystal using the tiling description, it is necessary to determine the symmetry, the atomic decoration of unit cells, and the unit-cell packing arrangement. Whereas only one perfect packing (up to translations) is possible for periodic crystals, there are an uncountably infinite number of distinct, perfect quasicrystal packings —geometrically distinguishable arrangements of unit cells—for any given symmetry. The distinct packings are said to belong to different "local isomorphism" (LI) classes.[1] A key problem is that there is no method for determining both the correct LI class and the unit-cell decoration simultaneously from diffraction data.[1,2] Most reported structural models employ *ad hoc* assumptions about the proper LI class, but this is clearly an unsatisfactory approach.

In this paper, we consider three criteria that may be used to identify which LI classes are physically realizable in solids. We show that only a discrete subset of the continuous spectrum of LI classes satisfies the criteria, thereby simplifying the determination of quasicrystal structure. The first criterion is that the packing be *restorable*—it can be uniquely specified given only a set of rules which fix the allowed unit-cell clusters smaller than some bound.[3] These rules are longer-range generalizations of the nearest-neighbor matching rules of the original Penrose tiling (PT).[4,5] The second criterion is that the packing be *growable*—a perfect packing may be grown from a seed cluster of unit cells using a set of local aggregation rules.[6] The third criterion is that the packing be *rapidly growable*—a perfect, or at least near-perfect, packing may be grown using local aggregation rules at a rate comparable to crystal growth. The notion is that the local rules might be physically manifested through finite-range atomic interactions. Restorability seems necessary if the LI class is to be an energetically stable state; growability and rapid growability seem necessary for the state to be physically accessible.

We apply these criteria to 2D pentagonal quasicrystal tilings, specifically, the continuous spectrum of LI classes obtained by direct projection from a 5D hypercubic lattice.[7,8] These direct projection tilings (DPT's) include the original PT, the only LI class previously known to be restorable,[3] growable,[6] and rapidly growable.[6] Is the PT the sole LI class with these properties? If so, our criteria would suggest that it is the only DPT that can be realized physically. If the analog were true for 3D icosahedral symmetry, it would eliminate the LI-class issue in the analysis of atomic structure. Alas, the answer is not quite so simple, but is perhaps more remarkable. We will show that only a countably infinite subset (measure zero) of the LI classes satisfies the three criteria. The subset can be ordered according to the range of the restorability and growability rules. The promising result is that the range is physically plausible in only a handful of cases, reducing the physically relevant LI classes from a continuous spectrum to a tractable few.

Restorability.—DPT's are obtained by projecting a subset of points in a 5D hypercubic lattice into a 2D "parallel space" $R^2_\parallel$ orthogonal to the direction $\boldsymbol{\delta}=(1,1,1,1,1)$, and drawing tile edges between the projections of neighboring lattice points.[7,8] The subset consists of those points which project into a finite acceptance volume, K, in the complementary 3D "perpendicular space," $R^3_\perp$. The LI class is fixed by γ, the position of K along δ.[2] Since shifts by $\Delta\gamma=\pm 1,2,\ldots$ project into the same LI class, γ is defined modulo one. A rotation by π transforms a γ tiling to a $\bar{\gamma}\equiv 1-\gamma$ tiling.

The fat and skinny rhombi used to construct all DPT's can also be joined to form random or periodic tilings. A first step towards restorability is local rules which constrain the tiles to form DPT's only. We adopt the generalized PT matching rules of Kléman and Pavlovitch (KP): Decorate tile edges with arrows in a prescribed pattern resulting in two types each of fat and skinny tiles (Fig. 1).[9] The conjecture, plausible from our studies, is that any plane-filling tiling with matching arrows along all edges is a perfect DPT.

Restorability for an LI class γ requires that the KP rules be augmented to permit the formation of only that specific DPT. A concept useful for distinguishing LI classes is the "r map,"[3] the maximal configuration of tiles centered at a tile vertex which lies inside radius r (where the unit of length is the tile edge). Configurations differing only in their edge decorations are described by distinct r maps. Configurations which are

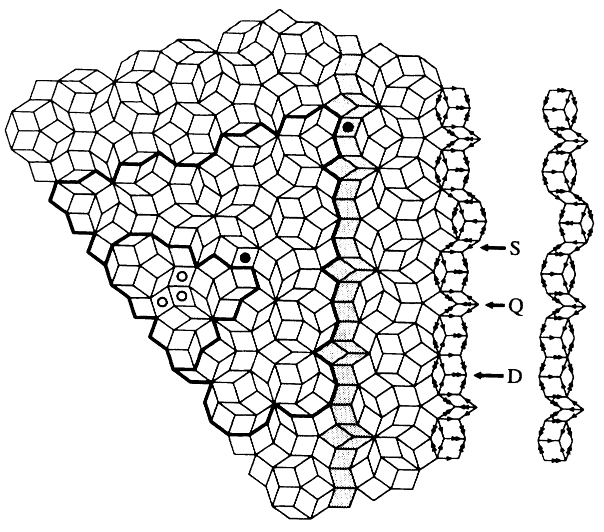

FIG. 1. A cluster obtained during growth of a perfect $\gamma=\{2\tau\}$ DPT from a seed cluster (tiles marked with an open circle), highlighting the dead surfaces encountered (heavy line). To grow beyond a dead surface, a local rule is invoked to add a corner tile (solid circle). Tiles are then added to forced sites until the next dead surface is reached. The first row of tiles forced beyond the dead surface is a worm row (e.g., the shaded tiles form a Q-type worm). The cluster has grown until it reaches a D-type worm (tiles marked with edge arrows) slaved to the shaded Q-type worm; i.e., the only correct choice is the D-type worm attached to the surface. The labels D, Q, and S indicate a D-type hexagon, a Q-type hexagon, and a single tile, respectively, which together form the building blocks for the worm. If the alternate D-type worm (at right) is substituted, the edge arrows match, but the combination of D- and Q-type worms is inconsistent with $\gamma=\{2\tau\}$.

identical up to rotation by $4\pi/5$ or $2\pi/5$ (but not $\pi/5$ or π, say) are described by the same r map and appear with equal density in the DPT. Each r map occurs with a density that varies continuously over some range of γ. The complete set of r maps for a given LI class is termed its "r atlas."[3] Restorability requires that there exists a finite r such that the r atlas is unique to the given LI class.

Our analysis is based on the direct projection technique.[7,8] Let $\{\epsilon_0,\ldots,\epsilon_4\}$ be the natural basis of R^5 and define the hypercubic lattice $\Lambda=Z^5$. $R^3_\perp$, with axes $(x^\perp,y^\perp,z^\perp)$, is embedded in R^5 in such a way that the vector $\boldsymbol{\delta}=(1,1,1,1,1)$ projects into $(0,0,5)$. Λ projects into a dense, uniform coverage of discrete planar "$z^\perp$ levels" in $R^3_\perp$, with $z^\perp$ an integer. Let the projections of the basis vector ϵ_i into $R^2_\parallel$ and $R^3_\perp$ be $\mathbf{e}_i^\parallel$ and $\mathbf{e}_i^\perp$, respectively. The acceptance volume K is obtained by projecting into $R^3_\perp$ a reference unit hypercube with edges oriented along the ϵ_i's. K is a rhombic icosahedron whose fivefold axis is five units long and aligned along $z^\perp$. The tiling is completely specified by $\boldsymbol{\alpha}$, the offset between the reference hypercube and the origin of Λ. The intersection of the $z^\perp$ levels with K is fixed by the component of $\boldsymbol{\alpha}$ parallel to $\boldsymbol{\delta}$; in fact, $\gamma=\{\boldsymbol{\alpha}\cdot\boldsymbol{\delta}\}$, where $\{x\}$ denotes the fractional part of x.

For fixed r, K can be partitioned into "r volumes": Points of Λ which map into the same r volume in K are projected in $R^2_\parallel$ as central vertices of the same r map with the same orientation. For an r map with N vertices, the associated r volume is the polyhedron obtained by the intersection of K with N translations of itself, each of the form $K-\sum_i k_i\mathbf{e}_i^\perp$, where $\sum_i k_i\mathbf{e}_i^\parallel$ is the displacement of one of the N vertices from the central vertex of the r map. Any region where two or more such polyhedra overlap is assigned to the r volume whose r map has the most vertices. An r map appears in a DPT with a density proportional to the intersection between its r volume and the $z^\perp$ levels. The density vanishes in the limit where the levels intersect an "extremum" of the r volume, i.e., the upper or lower boundary along $z^\perp$.

In order for a γ tiling to be restorable, there must exist "discriminants": r maps which distinguish the r atlas for γ from all others. Discriminants are characterized by two properties: (1) Their r volumes intersect $z^\perp$ levels at extrema, so that all discriminants are absent from the γ tiling, but one or more added by any shift in γ; (2) the rotation of any discriminant r map by π (by our definition, a different r map) must also be absent from the r atlas. The key to restorability, then, is interactions which forbid discriminant r maps. Condition (2) is necessary because any physically plausible interaction cannot make reference to absolute coordinates; that is, if the discriminant is forbidden, so is its rotation by π. By geometrical analysis of the r-volume construction, it can be shown that discriminants exist and, hence, *a DPT is restorable iff* $\gamma=\{n\tau\}$, *where n is an integer and* $\tau=\frac{1}{2}\times(\sqrt{5}+1)$. *For* γ*'s of this form, a sufficient interaction range to ensure restorability is*[10]

$$r=\mathcal{C}|n|+\mathcal{O}(1)\,, \tag{1}$$

where $\mathcal{C}$ *is a bounded geometrical factor.* In fact, a range $r=\frac{5}{5}|n|$ ensures that a $z^\perp$ level intersects an extremum lying at the lower boundary of one r volume and the upper boundary of another. The two associated r maps are potential discriminants for LI class γ. However, r must also be chosen so that the discriminants satisfy condition (2) above: For either r map, $\mathcal{R}$, its rotation by π, $\overline{\mathcal{R}}$, must be absent from the r atlas for γ. The r volumes $\mathcal{V}$ and $\overline{\mathcal{V}}$, corresponding to $\mathcal{R}$ and $\overline{\mathcal{R}}$, respectively, are related by inversion through the center of K; however, the $z^\perp$ levels are inversion symmetric only for $\gamma=0$ or $\frac{1}{2}$. If $\mathcal{V}$ intersects a $z^\perp$ level at an extremum, the distance between the analogous extremum of $\overline{\mathcal{V}}$ and the nearest $z^\perp$ level is

$$\Delta=2\min(\gamma,1-\gamma,|\gamma-\tfrac{1}{2}|)\geq(\tfrac{1}{4}\tau^6|n|)^{-1}\,,$$

whereas $\overline{\mathcal{V}}$ has height (along $z^\perp$) $h<\frac{5}{2}\tau^2r^{-1}$. Thus, if $h<\Delta$, $\overline{\mathcal{V}}$ cannot possibly intersect a $z^\perp$ level, and $\overline{\mathcal{R}}$ cannot be in the r atlas. This condition leads to the (conservative) bound $\mathcal{C}\leq\frac{5}{8}\tau^8\approx 30$.

Growability.— We can also show that all DPT's with $\gamma=\{n\tau\}$ can be grown *without defects* via local aggregation rules of the kind developed for the PT by Onoda *et al.*[6,11] Beginning from a seed cluster, the growth algorithm involves successively adding tiles at "forced-site" vertices where only one choice for adding tiles is locally consistent with the particular LI class. When the surface contains no more forced sites, the surface is said to be "dead." A single tile is then added at a corner site according to a subsidiary local rule. The added tile "revitalizes" the surface by creating new forced sites (see Fig. 1). This growth sequence can then iterate *ad infinitum.*

Dead surfaces are convex, polygonal configurations of tiles where each edge borders a "worm,"[6,12] a strip of connected tiles which can be replaced by a different worm strip so that the KP arrow rules are obeyed everywhere along the edge of the dead surface. Since two distinct worms can fit, the edge sites are, by definition, unforced. For PT's, each worm consists of a chain of hexagons made up of three tiles joined at a Q- or a D-type vertex[5] (see Fig. 1). There are two worm types, related by reflection about the worm axis. For DPT's with $\gamma=\{n\tau\}\neq 0$, each worm consists of a chain of hexagons interrupted by chains of single tiles. The four worm types can be paired into Q and D types. To replace one Q- (D-) type worm by the other, each Q- (D-) type hexagon along the worm is reflected about the worm axis; then any tile with edges normal to the axis is replaced by one of the same shape but the other arrow decoration (see Fig. 1). Dead-surface identification is complicated by the fact that each Q-type worm is "slaved" to a nearby D-type worm (and vice versa)—the choice of a Q-type worm normal to $\mathbf{e}_i^{\parallel}$, say, fixes a parallel D-type worm displaced by $[\frac{5}{2}n+\mathcal{O}(1)]\mathbf{e}_i^{\parallel}$. The growth rules must distinguish $\mathbf{e}_i^{\parallel}$ from $-\mathbf{e}_i^{\parallel}$ to determine whether a worm to be added to the surface is slaved to a worm inside or outside the cluster; as discussed after Eq. (1), a range $r=\mathcal{C}\,|n|$ is sufficient to resolve rotations by π. To force the pairing of worms, the growth rules must also span the distance from some central vertex to each of the slaved worms; $r=\frac{5}{4}\,|n|$ suffices for this. Thus, the range of the growth rules is also bounded by Eq. (1).

A complete growth algorithm, though, requires a local rule for revitalizing a dead surface. This depends critically on the existence of a local inflation operation[5,12] which transforms a DPT into a rescaled tiling in the same LI class. Here "local" means that there must be finite r_l such that all clusters belonging to a given r map with $r>r_l$ inflate in the same way; thus, inflation is uniquely defined for finite tilings of size $r>r_l$. Using this inflation, a sequence of successively larger, similarly shaped clusters can be constructed such that each is bounded by a dead surface and can be transformed into a predecessor by inflation. In fact, inflation guarantees that each cluster contains the preceding clusters within its borders. By inspection, one can determine the corner of a particular cluster to which a tile must be added so that additions at forced sites will produce the next larger cluster. A finite-range rule can be used to identify the appropriate corner from the local tile configuration.[6] Then, since the clusters are self-similar in shape, the same local rule can be used to grow a perfect cluster of arbitrary size.

Until now, local inflation operations were known only for PT's.[5,12] We have generalized the direct projection method[7] for obtaining these inflation rules to cover all $\gamma=\{n\tau\}$: Let Λ' be a sublattice of Λ whose basis is $\epsilon_i'=2\epsilon_i+\epsilon_{\langle i+1\rangle}+\epsilon_{\langle i+4\rangle}$, where $\langle k\rangle=k(\mathrm{mod}5)$. Let K' be the projection of a rescaled unit cell and $\boldsymbol{\alpha}'$ its offset with respect to the origin of Λ'. The tiling obtained by projecting Λ' using acceptance volume K' contains tiles with sides τ^2 longer than those in the original tiling. We seek $\boldsymbol{\alpha}'$ as a function of $\boldsymbol{\alpha}$ so that the two tilings belong to the same LI class and the inflation is local. Using an r-volume analysis similar to that outlined for the restorability problem, we have shown that these conditions can only be met for $\gamma=\{n\tau\}$, in which case

$$\boldsymbol{\alpha}'=\boldsymbol{\alpha}+\tfrac{1}{5}\,[3\gamma-5\lfloor\boldsymbol{\alpha}\cdot\boldsymbol{\delta}\rfloor-4N(n)]\boldsymbol{\delta}\,,$$

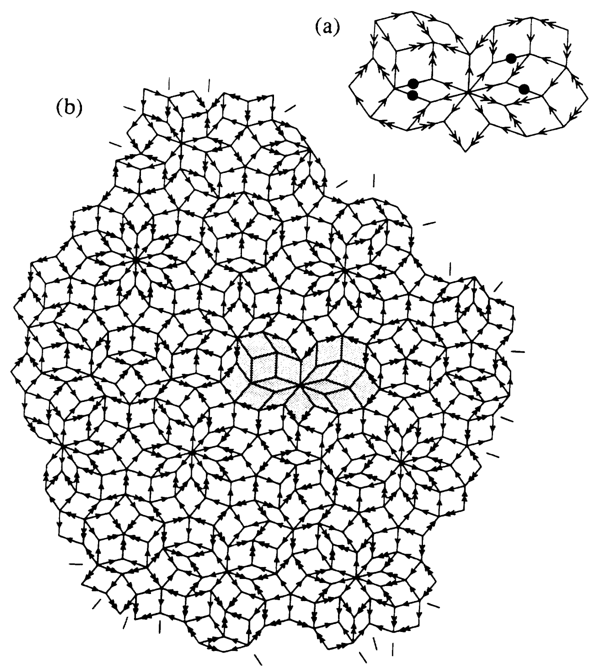

FIG. 2. (a) A decapod defect with charge 4 (see perimeter arrows) can be formed from a seed cluster with four edge mismatches (solid circles). (b) A cluster growing around the seed (shaded) in accordance with $\gamma=\{\tau\}$ growth rules. There always remain forced sites (tick marks) around the growing surface. By addition at only forced sites, a perfect DPT—apart from the pointlike seed defect—grows rapidly as a crystal.

where $N(0)=0$, and $N(n\neq 0)=2\lfloor n\tau\sqrt{5}\rfloor-3$; here $\lfloor x\rfloor = x-\{x\}$.

Rapid growability.—The growth algorithm described above could be physically realized in a stochastic growth process in which atoms or clusters impinge on a growing seed cluster. Local atomic interactions can be envisioned[6] which result in a large sticking probability, p_f, at forced sites, an exponentially small probability, p_c, at corner sites, and zero probability at all other sites. Such interactions would mimic the perfect growth rules until the cluster grew to a size of order p_f/p_c unit cells. However, for exponentially small p_c, the growth might be much slower than crystal growth.[11]

A simple corollary thereby assumes great significance: For any $\gamma=\{n\tau\}$, this hierarchy of sticking probabilities can be avoided if the seed contains a special topological defect known as a "decapod"[12] (see Fig. 2). A decapod can be assigned a "charge," q, by examining each edge on a closed path enclosing the defect: Add $+1$ (-1) if the arrow is along (opposite) the path.[6] Decapods can have $q=0,\pm 2,\pm 4,\ldots,\pm 10$ but a dead surface has $q=0,\pm 2$ only. Thus, a growing cluster containing a decapod with $|q|>2$ can never be enclosed by a dead surface, and, hence, there always exist forced sites. *A cluster containing a decapod with* $|q|>2$ *can grow into a perfect DPT (except for a single, pointlike defect) at a rate comparable to crystal growth.*

Hence, restorability and rapid growth rules exist only for DPT's with $\gamma=\{n\tau\}$, requiring an interaction range $\propto |n|$. Since ranges with only modest values of n seem physically plausible, determining the LI class for real quasicrystals has been reduced to a tractable problem.

We thank D. DiVincenzo and J. Socolar for many useful comments. This work is supported in part by the National Science Foundation, Materials Research Laboratories Program, under Grant No. DMR-8519059, and by the Monell Foundation through the Institute for Advanced Study, Princeton.

[1]D. Levine and P. J. Steinhardt, Phys. Rev. B **34**, 596 (1986).

[2]J. E. S. Socolar, T. Lubensky, and P. J. Steinhardt, Phys. Rev. B **34**, 3345 (1986).

[3]L. S. Levitov, Commun. Math. Phys. **119**, 627 (1988).

[4]R. Penrose, Bull. Inst. Math. Appl. **10**, 266 (1974).

[5]N. G. de Bruijn, K. Ned. Akad. Wet. Proc. Ser. A **84**, 39 (1981); **84**, 53 (1981).

[6]G. Y. Onoda *et al.*, Phys. Rev. Lett. **60**, 2653 (1988); **62**; 1210 (1989). The latter paper details how the growth rules can be mimicked by finite-range interactions in a stochastic process.

[7]A. Katz and M. Duneau, J. Phys. (Paris) **47**, 191 (1986).

[8]V. Elser, Acta Crystallogr. A **42**, 36 (1986).

[9]A. Pavlovitch and M. Kléman, J. Phys. A **20**, 687 (1987).

[10]Note that γ and $\bar{\gamma}$ require identical interactions.

[11]M. V. Jarić and M. Ronchetti, Phys. Rev. Lett. **62**, 1209 (1989).

[12]M. Gardner, Sci. Am. **236**, 110 (1977).