Growing Perfect Decagonal Quasicrystals by Local Rules

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A local growth algorithm for a decagonal quasicrystal is presented. We show that a perfect Penrose tiling (PPT) layer can be grown on a decapod tiling layer by a three dimensional (3D) local rule growth. Once a PPT layer begins to form on the upper layer, successive 2D PPT layers can be added on top resulting in a perfect decagonal quasicrystalline structure in bulk with a point defect only on the bottom surface layer. Our growth rule shows that an ideal quasicrystal structure can be constructed by a local growth algorithm in 3D, contrary to the necessity of nonlocal information for a 2D PPT growth.

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The announcement of the icosahedral phase of alloys in 1984 [1] posed many puzzles. The first question was what kind of arrangement of atoms could produce Bragg peaks with a rotational symmetry forbidden to crystals. The quasiperiodic translational order was proposed immediately as a candidate, and such materials began to be called quasiperiodic crystals or quasicrystals for short [2]. However, the appearance of quasicrystals brings new puzzles: why and how the atoms can arrange themselves to have such order, and especially, how quasicrystals can grow with perfect quasiperiodic order has been a dilemma since it seemingly requires nonlocal information while atomic interactions in metallic alloys are generally considered to be short ranged.

There are currently two alternative pictures to describe quasicrystals: energy-driven perfect quasiperiodic quasicrystals and entropy-driven random-tiling quasicrystals. Accordingly, two alternative scenarios for the growth [3] of quasicrystals exist: matching-rule based, energy-driven growth, and finite-temperature entropy-driven growth [4]. A major criticism for the former approach has been that no local growth rules can produce a perfect quasicrystalline structure in 2D [5,6]. Here, we show how to overcome this obstacle in a 3D quasicrystals.

Penrose tiling [7] has been a basic template for describing formation and structure of ideal quasicrystals. It can be constructed from fat and thin rhombi with arrowed edges shown in Fig. 1(a). The infinite tiling consistent with arrow-matching rules is the Penrose tiling but they do not guarantee the growth of a perfect Penrose tiling (PPT) from a finite seed. Successive ''legal'' (obeying the arrow-matching rules) additions of tiles to the surface of the already existing legal patch of tiles can produce defects. They usually occur after only a handful of tiles are added, and hence the arrow-matching rules cannot explain the long-range quasicrystalline order engendered by growth kinetics.

There has been a great amount of discussion and a number of debates on the possibility of a local growth algorithm for a PPT [3–6,8–10]. The debates partially emerge from a different assumption on the growth processes at the surface, uniform growth and preferential growth. In the former, growth occurs at any surface site with the same attaching probability, while it occurs with different attaching probabilities in the latter. In 1988, Penrose proved that a PPT cannot be grown by local rules with uniform growth by showing that ''deceptions'' are unavoidable [5] where a deception is a legal patch which cannot be found in a PPT [5,6]. In the same year, Onoda *et al.* introduced a preferential growth algorithm which can avoid deception by local rules called ''vertex rules'' [3]. However, vertex rule growth stops at a ''dead surface'' and nonlocal information or arbitrarily small growth rates are required to be an infinite PPT. Yet, their growth algorithm is believed to provide methods to grow the most ideal quasicrystalline structures with local information. If an initial seed contains a special kind of defect, called ''decapod'' [11], Onoda *et al.* showed that the seed can be grown to an almost PPT (whose only defect is the initial decapod defect) [3]. A point defect in a 2D tiling growth usually implies a line defect in a 3D decagonal tiling growth. If we apply a solid-on-solid type growth [12] so that a layer copies configuration of the one below, we get decagonal tiling consisting of identical layers with a decapod defect at the center of each layer. This line defect has been considered to be a minimum imperfection for the 3D decagonal quasicrystal structure from the local growth algorithm.

In this Letter, we consider the growth of decagonal quasicrystals and present a local growth algorithm for 3D decagonal tiling which consists of PPT layers except the bottom layer. We use the two well known results of the Onoda *et al.* study on planar decagonal quasicrystal growth [3]. (i) A local growth method around a ''cartwheel decapod'' leads to dead surfaces, from which further growth of a PPT requires nonlocal information. (ii) Infinite local growth is possible if it starts from an ''active decapod defect'' but the resultant tiling contains the defect and is

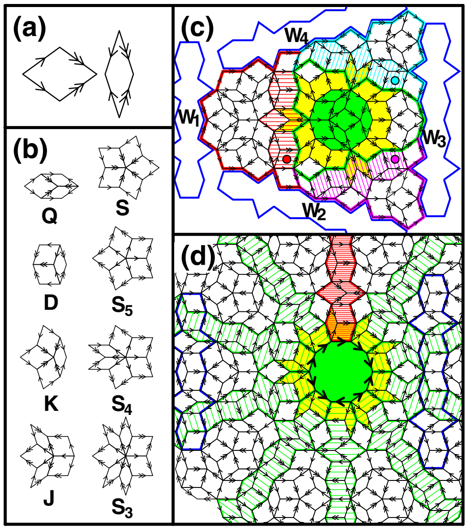

FIG. 1 (color online). (a) Fat and skinny Penrose tiles with arrows. (b) The eight ways of surrounding a vertex in a PPT. (c) Dead surfaces encountered when a tiling is grown by rule L from a cartwheel decagon. See text for details. (d) A decapod tiling. Ten semi-infinite worms meet at the decapod decagon at the center. From the yellow seed tiles, an infinite tiling (decapod tiling) can be grown by rule L.

not a PPT. By combining an active decapod defect in the bottom layer with a cartwheel decapod in the second layer and using the information of the underneath configuration, we make the growth continue beyond dead surfaces in the second, and subsequent layers. The bottom surface layer has a point defect, but we can consider the overall structure as that of a perfect decagonal quasicrystal since deviations from the bulk layer structure are natural for the surface layer even for ideal crystal materials.

Let us first discuss the growth rule in a 2D Penrose tiling. For the arrow-matching-rule growth, a deception can be made as few as three tiles [5]. Since the growth process does not allow tiles to be removed, a deception (which is not a part of a PPT) cannot grow to a PPT, and we need a growth rule which allows no deceptions of any size for a PPT growth. We can avoid three tile deceptions by introducing a more restricted growth rule which allows only correct (subset of a PPT) three tile patches. However, the new growth rule can make a deception in a larger scale, for example, a three tile deception of inflated [11] tiles. Since a deception can be made in all scales of multiply inflated tile sizes [11], it is unavoidable for a local growth rule. The absence of local rules for perfect tiling growth seems to be the case for general aperiodic tilings in 2D [6]. Based on this observation, Penrose even speculates that there may be a nonlocal quantum-mechanical ingredient to quasicrystal growth [5,13].

Onoda *et al.* proposed "vertex rules" which avoid an encounter of deceptions [3]. Here, a tile can be added only to a "forced edge" which admits only one way of adding a tile for its end vertices to be consistent with any of the eight PPT vertex configurations shown in Fig. 1(b). In this Letter, their vertex rules will be called "rule L" and used for the "lateral"-direction growth (for 3D decagonal tilings). The problem of rule L is that the growth stops at a finite size patch called a "dead surface" which consists of unforced edges.

There are special kinds of pointlike defects, called decapod defects [11], which can be an ideal seed to grow an almost PPT without encountering a dead surface [3]. A decapod is a decagon with single arrowed edges. Since there are 10 arrows, each of which can take two independent orientations, there are 2^{10} combinations of states. After eliminating rotations and reflections, we get 62 distinct decapods. We can tile inside the decagon legally for only one decapod, the cartwheel case and the rest of the 61 decapods are called decapod defects. One notable property of the decapods is that the outside of the decagon region can be legally tiled for all 62 cases. This can be easily understood from the fact that six semi-infinite worms and two infinite worms meet at the center cartwheel decagon [shown by green tiles in Fig. 1(c)] in a cartwheel tiling. If we remove the tiles in the center cartwheel decagon, two infinite worms become four semi-infinite worms, and we have ten semi-infinite worms which start at the perimeter of the center decagon. A decapod defect tiling is formed by flipping one or more of these ten semi-infinite worms. The arrows on the worm perimeter will still fit except a mismatch at the decapod decagon perimeter. Figure 1(d) shows an example obtained by flipping the worm denoted by red hatched tiles. Among 61 decapod defects, there are 51 "active" decapod defects which have at least three consecutive arrows of the same orientation on their decagon perimeter [14]. One can show that a patch containing an active decapod defect is never enclosed by a dead surface [15].

Our 3D growth rules are constructed by observing that a cartwheel PPT and a decapod tiling can be different only in ten semi-infinite worms. Consider two layer growth from a (two layer) seed that contains a cartwheel decagon [yellow tiles in Fig. 1(c)] and a decapod defect [yellow tiles in Fig. 1(d)] at the upper and the lower layers, respectively. If each layer grows with rule L independently, the growth of the upper layer would stop at the red-purple-blue dead surface while the lower layers grow indefinitely. Now, we introduce a vertical growth rule so that a tile can be added at the dead surface of the upper layer properly. Note that the basic tiles for 3D decagonal tiling are rhombus prisms which have top and bottom faces as well as side faces. By vertical growth, we mean attaching tiles on the surface layer such that the bottom faces of the attached tiles contact with the top faces of the surface layer tiles, while lateral growth means attaching tiles to side faces at the perimeters. We propose a vertical growth rule, "rule V," with which the lateral growth rule, rule L produces a PPT on a decapod tiling. If a tile in a flipped worm of the decapod tiling (the lower layer) is copied by a vertical growth, a defect on the upper layer is inevitable. Our rule V is designed to avoid such a case and allow tiles to attach vertically only on the "sticky" top faces, blue-circled fat tiles in S_3 and S_4 configurations shown in Fig. 2(a) [16]. They always form D hexagons indicated by dotted lines. Such D hexagons can lie only at the end of worms since the other (uncircled) tiles in S_3 or S_4 configurations prevent formation of hexagons next to the circled tiles. Therefore, the sticky sites can be located only outside or at the ends of the semi-infinite worms as illustrated in Figs. 2(b)–2(e). One can further show that sticky sites are strictly outside of the semi-infinite worm if it is flipped since the flipping makes the vertices at the end be illegal (and therefore they cannot be S_3 or S_4). Therefore, no sticky sites are in the flipped worm, and hence rule V does not introduce a defect or deception for the layer that grows on a decapod tiling.

Now we show that rule V is enough for the upper layer to grow beyond the dead surfaces when the growth starts from a proper seed. Our seed consists of two layer finite patches which include a cartwheel decagon and an active decapod decagon at the upper and the lower layers, respectively. Figures 2(b) and 2(c) show an example. The upper layer seed [Fig. 2(b)] consists of a cartwheel decagon and 10 D hexagons. It covers all ends of the semi-infinite worms in the lower decapod seed [Fig. 2(c)] which consists of a decapod decagon and 10 D hexagons [17]. Let us first consider the properties of dead surfaces which contain the upper layer seed. By applying inflations to a cartwheel

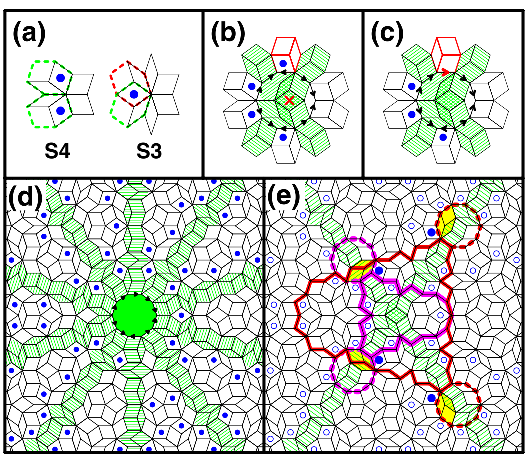

FIG. 2 (color online). (a) Sticky sites, on which a tile can be attached vertically, are indicated with blue circles. (b) Upper layer configuration of a (two layer) seed. It contains a cartwheel decagon and ten hexagons attached to the decagon. The tiles denoted by blue circles are sticky site and the tile denoted by X is the nucleate site. (c) Lower layer configuration of the (two layer) seed. It contains an active decapod decagon with five consecutive arrows of the same orientation. (d) Sticky sites on a decapod tiling. All sticky sites are outside of the ten semi-infinite worms. (e) Dead surfaces which contain the center cartwheel decagon in a cartwheel tiling. The two crossing infinite worms of the cartwheel tiling always pass the two 72° corners of dead surfaces.

tiling, one can show that the dead surfaces, which contain the center cartwheel decagon, have two 72° corners and each corner is passed by an infinite worm [green worms in Fig. 2(e)] of the cartwheel tiling [10,11]. The D hexagon at the 72° corner forces the next two hexagons just outside of the dead surface (in the infinite worm direction) to be D and Q. These two hexagons force a cartwheel decagon to form just outside of the corner as illustrated by (red and purple) dashed lines in Fig. 2(e). A Q decagon (denoted by yellow tiles) in the dashed cartwheel decagon forces a tile just outside of the 72° corner [denoted by the solid green circles in Fig. 2(e)] to be sticky. We call these sticky sites as "launching" sites. The exact position of a launching site depends on the orientation of the corner [10] but the patch can grow by rule L for both cases. The position of the launching site determines the orientation of the worm along the side lines of the dead surface making the edges at the dead surface become forced. Hence, the upper layer would grow to infinite by rule L if rule V guarantees tiles at the launching sites. This is the case when it grows on an active decapod tiling obtained by flipping a semi-infinite worm [18] of a cartwheel tiling as shown in Fig. 2(d). Since neither crossing infinite-worms are flipped [compare Figs. 2(d) and 2(e)], the underneath tiles of the launching sites will be always the sticky sites of the decapod tiling and tiles at the launching sites are guaranteed by rule V on the decapod tiling.

For the completeness of the 3D decagonal quasicrystal growth, we need to provide the rule for the nucleation of an island (seed) from the third layer. The physical process of the nucleation of an island on a PPT would be similar to that of a perfect crystal surface. High quality quasicrystals are grown when they grow slowly, or in other words, when the chemical potential of bulk quasicrystal is slightly less than that of the fluid phase. Therefore, adatoms or "adtiles" on a terrace would be unstable and probably diffuse on the terrace until they evaporate (i.e., go back to the fluid phase) or attach to preferential sites (forced or sticky sites) [12]. We believe that the chemical potentials of the forced sites are less than those of sticky sites and adtiles attach to forced sites for most cases. However, when the terrace forms a dead surface (or part of a dead surface), it is not easy for an adatom to find a forced site and it would attach on a sticky site, especially to a launching site whose chemical potential is expected to be lower than that of an isolated sticky site. Note that both forced and launching sites are at the perimeters of terraces and become irrelevant to adtiles on the middle of the terraces as they grow sufficiently large. It is then conceivable that two or more adtiles meet on a terrace and begin to form a new patch of the next layer before they arrive at the perimeter. With this physical process in mind, we allow a nucleation process in our growth algorithm. The nucleation of an island can happen in cooperation of a quite large cluster of tiles. We choose the "cartwheel seed," a cartwheel decagon and the 10 D hexagons arranged as Fig. 2(b), as such a cluster and introduce a nucleation site on it. The site X in the figure is called a ‘‘nucleate site’’ if its lateral neighboring tiles form a cartwheel seed and if it has a underneath tile [19]. When a nucleate site is selected, we create a cartwheel seed on it.

Let us summarize our growth mechanism for decagonal quasicrystals. It consists of the following three processes: lateral growth by rule L, vertical growth by rule V, and the island nucleation (seed formation) for the new layer. Algorithmically, it is realized by the following steps: (i) Start with a two layer seed whose upper and lower layers contain a cartwheel decagon and an active decapod decagon, respectively. (ii) Randomly choose a surface site. Check if it is a sticky, nucleate, or unsticky site when it is a top face. For a side face, check if it is a forced or unforced site. (iii) Perform the vertical growth, nucleation, or lateral growth if the chosen site is a sticky, nucleate, or forced site, respectively. Do nothing for an unsticky (top face) or unforced (side face) site.

For simplicity, we have chosen the unit attaching probability for all sticky, forced, and nucleate sites. In real material, they probably have different attaching probabilities due to differences in their chemical potentials and attaching kinetics. We think that the attaching probabilities are different even among the forced sites (and among the sticky sites) since they depend on local configurations. However, the nucleation probability would be much smaller than that of the attaching probability of a tile in any case since the former demands a cooperation of many tiles. The slow process of nucleation implies a layer by layer growth for a perfect decagonal quasicrystal [20]. It is beyond the scope of this Letter to predict the growth kinetics of real quasicrystals since it requires knowing atomic cluster structures corresponding to each type of tiles as well as the kinetic parameters of atomic attachment of real materials.

Our growth algorithm has a couple of limitations. First, it can produce only one kind of PPT, a cartwheel tiling. Second, the seed must include a decapod defect. However, a decapod defect may form under quite general conditions. It is believed that every possible hole surrounded by an arrow-matched Penrose tiling is equivalent to a decapod hole [11]. The bottom layer, which may grow under structurally different environment, is a natural place to have such defect. Our algorithm shows that PPT is possible from the second layer if the defect can be surrounded by legal tiles. Once a PPT layer begins to form on the second layer, our growth algorithm produces PPT layers easily from the third layer. We hope that the present work stimulates studies on 3D growth rules for real quasicrystals.

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[16] We introduce the concept of sticky sites as a mathematical device to avoid the mistakes of copying a tile in a flipped worm but it may mimic the real growth process of quasicrystals. Recently, Fournée *et al.* observed ‘‘traps’’ or sticky sites on which adatoms are easily captured for some quasicrystal surfaces [21].
[17] Covering of all ends of the semi-infinite worms further ensures that all sticky sites in the decapod tiling stay outside of the semi-infinite worms.
[18] From the Fig. 2(b), one can see that an active decapod tiling is obtained by flipping a semi-infinite worm in a cartwheel tiling while flipping a half of an infinite worm results in an inactive decapod tiling.
[19] The requirement of the underneath tiles is added to ensure that the nucleation happens from the third layer. In real growth, this requirement may not be needed. Nucleation is likely to happen only on the top layer (hence from the third layer) which can be large enough to wait the slow nucleation process.
[20] The sticky sites [Fig. 2(a)] are formed only on a compact cluster of tiles. Therefore, the vertical growth by rule V, which produces isolated tiles, cannot be continue more than one layer height without nucleation process.
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