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Substitution rules for icosahedral quasicrystals

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Original well-defined substitution algorithm on how to construct the icosahedral packings is proposed, simple inflation/deflation rules for icosahedral quasicrystals are described, and natural local matching rules are derived.

The discovering of quasicrystals by Shechtman\textsuperscript{1} showed that strict long-range order in solids do not require mandatory periodicity. Steinhardt \textit{et al.}\textsuperscript{2, 3} proposed the theoretical description of icosahedral quasicrystals having generated the packing with icosahedral symmetry by the grid projection technique from six-dimensional hypercubic lattice. The goal of the present paper is to describe icosahedral quasicrystalline packings without appealing to higher dimensions.

Three main special features of quasicrystals must be highlighted. The first is that they reveal strong long-range order that lacks any translational symmetry in the same way as, for example, the Penrose tiling does.\textsuperscript{4} According to widely held opinion, only groupoids represent the appropriate tool for studying long-range order in quasicrystals instead of symmetry groups. We will abandon this point of view and will operate in terms of usual group theory. Such approach leads to fractal tilings. The second main special feature is that quasicrystals exhibit self-similarity, which was usually considered to mean that the same patterns occur at larger and larger scales. We will consider self-similarities not as the repeating structural motifs in solids or substitution rules for tilings but as the true group automorphisms. Under such assumption, the composition of self-similarities with different fixed points takes on special significance. The third special feature shows that the diffraction patterns of quasicrystals reveal the discreteness, as has been first demonstrated by Mackay.\textsuperscript{5}

There is no unified viewpoint about which one of these properties would most uniquely characterize the aperiodic long-range order.\textsuperscript{6} For example, Dyson\textsuperscript{7} proposed the following definition: “A quasi-crystal is a distribution of discrete point masses whose Fourier transform is a distribution of discrete point frequencies.” Furthermore, he introduced an example of the artificial solid with distribution of atoms resembling the distribution of zeros of the Riemann zeta function, and showed that such solid would have an essentially discrete diffraction pattern. Therefore, according to the modern definition, aperiodic crystals are not identical with structures that were typically referred to as quasicrystals. We would like to keep the term \textit{quasicrystal} reserved for those aperiodic crystals that exhibit self-similarity, namely multiple self-similarity. We do not pretend to make a balanced review but rather refer the reader to more detailed descriptions of the modern viewpoints on the structure and properties of quasicrystals which may be found in literature.\textsuperscript{8–14}

In our study, we started with the idea of the fractal nature of Penrose tiling introduced by Bandt and Gummelt,\textsuperscript{15} who proposed to replace the basic tiles in the kites-and-darts tiling by corresponding tiles with fractal boundaries obtained iteratively by multiple application of the deflation procedure. Recently, we have considered the rhombus Penrose tiling under the assumption that both inflation and deflation procedures were applied an infinite number of times.\textsuperscript{16, 17} The resulting fractal tiling may serve as a corresponding ‘parent’ structure\textsuperscript{18} for the entire local isomorphism class. This ‘parent’ structure has maximum possible symmetry, which is revealed in ‘daughter’ structures as hidden or incomplete. For instance, one can reveal that the local environment of any arbitrarily chosen vertex ceases to change after several iterations coinciding with one of two possible self-similar arrangements, which are mutually inverse one in respect to another.

Goodman-Strauss\textsuperscript{19} proofed that every substitution tiling can be enforced with finite matching rules. In general, however, the connection between matching rules and self-similarity is far from being understood.\textsuperscript{20} Recent advances in the theoretical explanation of substitution tilings and their matching rules are surveyed in the literature.\textsuperscript{20–22} To our knowledge, no attempts have been undertaken to formulate the substitution rules for icosahedral packings, as well as no attempts have been made to apply the fractal approach and derive natural matching rules for the practically important case of icosahedral quasicrystals. Properly speaking, there exist only few papers concerning the possible fractal nature of actual self-similar tilings. Bandt and Gummelt\textsuperscript{15} were right when they wrote that some readers would find inconvenient to work with such tiles. On the other hand, there is no necessary need for inconvenient tiles when simply holding in mind that every convenient tile contains a fractal dust of quasi-lattice sites and taking into account that the true symmetry of the fractal tiling could be higher than the apparent symmetry of that one represented by finite-sized tiles.

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Consider the icosahedral tilings within the frameworks of the fractal approach. The specific problem is how to fill the three-dimensional space with so-called ‘golden’ zonohedra. Of course, the skeptical reader may claim that this problem has already been solved by Steinhardt et al.\textsuperscript{2,3} This assertion is not too far from the truth. The icosahedral packings have indeed been described as a result of the unit cell decoration by intersecting Ammann planes with the detailed subtiling specification. Unfortunately, because each cell was divided into very small pieces and the specification of the Ammann plane decoration of the unit cells was complicated, these small pieces of the cells would be very difficult to track in integral structure. The detailed method, by which the positions of the planes in the zonohedra were obtained, as well as the correct deflation of each cell, was difficult to understand without access to some 3D models. The complicated matching rules, the constant need to recall higher-dimensional representation in order to verify the local environment of a given cell, and the need to cross-check the specification reference table in order to perform decoration correctly – all of these complications hinder the accurate analysis and practical use of the algorithm. Moreover, we have a firm conviction that a three-dimensional description of a three-dimensional object is always preferable.

On the other hand, the higher-dimensional approach\textsuperscript{2,3,23-26} represents today the only successful way in solving the quasicrystals puzzle, so it would be silly to criticize it. We don’t mean to look for disadvantages, but rather to highlight the inherent difficulties due to the lack of any alternative.

Recently, we derived an elegant procedure enabling us to construct the icosahedral packings. As usual, four types of unit cells appear – triacontahedron, rhombic isosahedron, rhombic dodecahedron, and prolate rhombohedron. When formulating the deflation rules, we advise not to divide the unit cells into very little pieces but instead to inflate four original unit cells and deflate the enlarged cells back to their own original copies. In this case, the original unit cells should be increased by a factor of $\tau^3$, where $\tau$ is the golden mean, so that all vertices of the inflated unit cells coincide with some of vertices of the original quasilattice built by the conventional rules.

The inflation/deflation rules for the golden zonohedra are illustrated in Fig. 1. The golden zonohedra are inflated by a factor of $\tau^3$ and are uniquely decorated by the polyhedra of the original size. Some of the small units partially exceed the bounds of the inflated cells. On the one hand, they are simultaneously owned by all adjacent cells in the final tiling. On the other hand, they link the adjacent cells together, providing the integrity of the tiling as a whole. The inflated cells, in turn, have to be assembled face-to-face into larger cells that share their own reduced copies on the boundaries as well as on the edges. For example, two adjacent cells may share reduced rhombohedra, which are divided exactly in half. Another example may be when five adjacent inflated rhombohedra are assembled together around the common edge sharing either the rhombic triacontahedron or the rhombic isosahedron, so that the shared polyhedra are divided exactly in five equal parts, etc.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{Fig1}
\caption{Inflation/deflation rules for golden zonohedra. Four types of zonohedra serving as unit cells for icosahedral quasicrystals are inflated by a factor of $\tau^3$ and uniquely decorated by the tiles of the original size. (a), (b), (c), (d) Inflation/deflation rules for the prolate rhombohedron, rhombic dodecahedron, rhombic icosahedron, and rhombic triacontahedron, respectively.}
\end{figure}
Fig. 2 Deflation of the rhombic triacontahedron. (a) – (i) Consecutive steps of the deflation. We start with a triacontahedron at the center (a) and place thirty rhombic dodecahedra on the 2-fold axes (b), twenty rhombohedra on the 3-fold axes (c), twelve clusters of ten rhombohedra on the 5-fold axes (d), thirty triacontahedra on the 2-fold axes (e), twelve rhombic icosahedra on the 5-fold axes (f), and twenty clusters of ten rhombohedra on the 3-fold axes (g). Next, the rhombic icosahedra on the 5-fold axes are capped by twelve clusters of five rhombohedra (h), and finally, sixty rhombic icosahedra are placed in the middle of each edge of the inflated cell (i).

Fig. 3 Deflation of the rhombohedron. (a) – (h) Consecutive steps of the deflation. We start with a triacontahedron (a). Next, we place three rhombic dodecahedra (b), four rhombohedra (c), three triacontahedra (d), and one rhombohedron (e). Next, the unfinished cell is turned upside down (f). Then, twenty five rhombohedra are placed as indicated (g), and finally, nine rhombic icosahedra are placed in the middle of the corresponding edges of the inflated cell (h).

Fig. 2 represents the deflation rule for the rhombic triacontahedron. We start with a triacontahedron at the center and place corresponding zonohedra shell-by-shell as depicted in the figure.

The deflation rules for the remaining three zonohedra may be derived from the corresponding deflation rule for the triacontahedron. We have made an assumption that, for the infinitely fragmented fractal tiling, there must be an equivalent representation by overlapping spheres and then the shared regions must have identical decompositions. Thus, when the image of one of the inflated zonohedra is properly superimposed onto an existing inflated and deflated rhombic icosahedron, the desired deflation rules may be simply obtained by the duplication of intersecting areas.

The deflation rule for the rhombohedron is shown in Fig. 3. Let's take a look at the opposite vertices on the 3-fold axis. They are obviously not equivalent. One vertex is accompanied with rhombic icosahedra in the second shell, whereas the other vertex is accompanied with triacontahedron. These opposite vertices exemplify two alternative vertex types in the corresponding fractal tiling.
The deflation rules for the rhombic dodecahedron and for the rhombic icosahedron are depicted in Figs. 4 and 5, respectively. The decoration procedures are exactly analogous to those that are described above. As previously, we start again with a triacontahedron. Then, we add accordingly the shells of rhombic dodecahedra, rhombohedra, triacontahedra, rhombic icosahedra, rhombohedra, and rhombic icosahedra. Next, the unfinished cells are turned upside down, and finally, the additional rhombohedra and rhombic icosahedra are placed as indicated in the figures. Note again that the opposite sides of the rhombic dodecahedron are also not equivalent, as well as those of the rhombic icosahedron. For example, one side of the rhombic icosahedron inherits the outer surface of the inflated triacontahedron and is characterized by rhombic icosahedra, which are placed in the middle of each edge. The opposite side is characterized by the ring of ten triacontahedra arranged face-to-face around the 5-fold axis. The upper and down vertices on the 5-fold axis of the rhombic icosahedron also exemplify the same two alternative vertex types in the corresponding fractal tiling.
The described inflation/deflation procedures obey the composition/decomposition requirements. The cells, which match together, decompose into the smaller ones that naturally also match together. This rule makes it possible to construct the tiling of larger and larger sizes, which would eventually cover the whole space by repeatedly applying the inflation step with subsequent deflation step, so that the size of the original tiles remains unchanged. In that case the inflation/deflation rules described above may be considered as substitution rules for icosahedral quasicrystals.

Assume that there is a tiling which covers the whole space. Perform the deflation procedure on all cells once, and then superimpose the obtained tiling over the initial one. The alternating order of the reduced copies of the initial unit cells, on the common faces and along common edges, uniquely predefines the natural local matching rules. The matching rules include requirements for vertices, edges, and faces (see Fig. 6).

Assume that we have generated the tiling covering the entire space by repeatedly applying the inflation and deflation procedures to arbitrary unit cell. Choose an arbitrary vertex and apply successively the deflation and inflation procedures on all cells in its local environment, but now in the reverse order. Then, the both operations should be applied successively several times. Since we discuss an infinite tiling, the tiling itself in this case remains unchanged. This will only lead to an increase of the scale of consideration every time by a factor of $\tau^3$, as if we examine details of the picture through a very large magnifying glass. One can reveal that the local environment of any arbitrarily chosen vertex ceases to change after sufficient iterations. The regions initially surrounding the chosen site, in the limiting case, become removed infinitely far away. Any vertex becomes the center of the star, which is made of twenty rhombohedra. Therefore, in the corresponding fractal tiling, there are only two types of inequivalent sites that correspond to the centers of stars surrounded either by twelve rhombic icosahedra or by twelve triacontahedra in the second shell.

Two vertex types are referred to as the $A$- and $B$-types, respectively. The inflation/deflation procedure never permutes the vertices of different types into each other. In the whole tiling, there exist no edges connecting equivalent vertices. Only alternate vertices may be connected by edges. Further, there are exactly two types of edges. The first type edge decomposes after deflation into the reduced copy of the second type edge, the rhombic icosahedron, and the reduced copy of the first type edge itself. The second type edge decomposes into its own reduced copy and triacontahedron. We have marked the second type edge by an arrow indicating the position of the triacontahedron after deflation. Further, there are exactly three types of inequivalent faces. The Steinhardt’s designations for faces are specifically depicted in Fig. 6 for further understanding.

There exist exactly three packings with a single center of icosahedral point symmetry in the 3D Euclidean space. The deflation rule for the triacontahedron defines the first one. Indeed, let’s take the initial triacontahedron (Fig. 2a) and apply the inflation/deflation steps. We get the triacontahedron enlarged by a factor of $\tau^3$ and decorated as shown in the Fig. 2i. Next, we apply the inflation/deflation procedure to the enlarged and decorated triacontahedron just obtained. Let’s recall that the central triacontahedron (Fig. 2a) is surrounded by the shell of thirty rhombic dodecahedra (Fig. 2b) followed by rhombohedra (Fig. 2c,d), triacontahedra (Fig. 2e), rhombic icosahedra (Fig. 2f), and so forth. Thus, in the inflated packing, the central decorated triacontahedron (Fig. 2i) should be surrounded by thirty decorated rhombic dodecahedra (Fig. 4j) followed by decorated rhombohedra (Fig. 3h), decorated triacontahedra (Fig. 2i), decorated rhombic icosahedra (Fig. 5j), and so forth. When applied iteratively, the above procedure generates the unique well-defined algorithm on how to fill the entire space with golden icosahedra. When starting from single triacontahedron, the algorithm results in the desired first packing with a single center of icosahedral point symmetry.

The two remaining packings may be derived by the same manner when assembling twenty rhombohedra either around the $A$- or $B$-type vertices with subsequent performing the inflation/deflation procedures for rhombohedra (see Figs. 7 and 8, respectively).

The animated scheme of cluster assembling is available in an electronic supplement to the article – better viewed in full-screen presentation mode to assemble and disassemble the clusters step-by-step.
Consider the infinitely fragmented fractal tiling that is built around the \textit{A}-type vertex. For this purpose, we assemble twenty rhombohedra around the \textit{A}-type vertex and perform the inflation/deflation procedure infinitely many times, so that the tiling covers eventually the entire space. Then, we deflate every cell infinitely many times, too. The resulting fractal tiling reveals the true self-similarity, because it coincides with itself as a whole when mapping the entire space onto itself by single affine transformation. Consider now the fractal tiling that is built around the \textit{B}-type vertex. It also reveals self-similarity in the group-theoretical sense. The question arises on how to combine self-similarities with different fixed points. The affine transformation has the only singular fixed point. It cannot move. The solution is indeed so simple that it seems almost absurd – it is necessary to consider another topological space instead of the Euclidean one.

For example, when the infinitely fragmented fractal Penrose tiling is the subject of interest,\textsuperscript{16,17} one should replace the Euclidean plane with the extended complex plane. What is the secret of the trick? Imagine the Penrose tiling covering the entire plane and draw two axes indicating the real and imaginary parts of the complex variable. It seems like nothing has changed. The Penrose tiling itself looks like before. But for now there may exist two different centers of global fivefold symmetry in the plane – one at the origin and another at the infinitely distant point. Recall that the complex plane may be brought into correspondence with a sphere by stereographic projection, so that the origin corresponds to the south pole whereas the infinitely distant point corresponds to the north pole. The logarithmic spirals, which represent in the plane the geodesic lines for the rotational homothety, may be brought into correspondence with loxodromes on the sphere. The projective mappings of the Riemann sphere onto itself may be converted into transformations of the tangent complex plane.
Fig. 8 Building the tiling with complete icosahedral point symmetry around the B–type vertex. (a) – (j) Consecutive steps of the building procedure.

Now the problem of combining self-similarities with different fixed points is no longer the case. The first self-similarity operation that is acting on the fixed point of the second self-similarity operation produces the infinite set of singular points when it is performed repeatedly and *vice versa*. The resulting infinitely fragmented fractal tiling may be described as usual in terms of the group theory, in contradistinction to the quasicrystalline plane tilings from which they are derived. The corresponding group generators may be expressed in terms of Möbius transformations, where the symmetry group of the resulting fractal would exemplify the symmetry of a certain discrete subgroup of the continuous group of linear fractional transformations.

The detailed explanation may be found elsewhere.\textsuperscript{16,17} The essence of the problem can be clarified by the quote from the monograph by Mumford *et al.*,\textsuperscript{27} who noted that this kind of symmetry is characteristic for “...a family of unusually symmetrical shapes, which arise when two spiral motions of a very special kind are allowed to interact. These shapes display intricate ‘fractal’ complexity on every scale from very large to very small. Their visualization forms part of a century old dream conceived by the great German geometer Felix Klein. Sometimes the interaction of the two spiral motions is quite regular and harmonious, sometimes it is total disorder and sometimes – and this is the most intriguing case – it has layer upon layer of structure teetering on the very brink of chaos.” Simply put, these groups describe the interaction of spiral motions on the plane or, in other words, the multiple self-similarity.

According to the known property of linear fractional transformations, two-dimensional self-similar fractal tilings must exhibit the circular property. Any of their symmetry operations maps the given circle again into a circle. In the same way, the symmetry operations of three-dimensional self-similar tilings should map the given sphere again into a sphere. Generally, we have no right to cast aside such symmetries that map the exterior of a given sphere into the interior of its image – the generalized self-inversions.
Therefore, the problem of the quasicrystalline order is closely related to the pure mathematical problem that was raised by Coxeter, namely: “...how Euclidean geometry, in which lines and planes play a fundamental role, can be extended to \textit{inversive} geometry, in which this role is taken over by circles and spheres.” Of course, the polygonal and polyhedral quasicrystalline tilings are inconsistent with reflections in spherical mirrors, but their infinitely fragmented fractal ‘parents’ are apparently fully compatible with curvilinear symmetry, although it may seem surprising or hard to believe.

So, we have described the original well-defined substitution algorithm on how to simply construct the icosahedral packing firstly invented by Steinhardt \textit{et al}.\textsuperscript{2,3} It is necessarily to highlight that the golden zonohedra may be assembled without addressing to higher dimensions, and the local matching rules may be formulated without addressing to Ammann planes. Our description remains principally tree-dimensional, though we initially used the results of the higher-dimensional approach to formulate the first inflation/deflation rule. It is important that the three-dimensional structure is described by the three-dimensional algorithm. There is no need in the embedding space and strip-projecting.

An additional explanation is necessary. On the one hand, our tiling shown in the Fig. 2 exactly corresponds to that one described by Socolar and Steinhardt\textsuperscript{1} (see Fig. 9 in their paper). On the other hand, there is an essential difference. Socolar and Steinhardt\textsuperscript{1} provided further subdivision of initial zonohedra into smaller, less regular parts, so that the additional vertices appear inside zonohedra. We do not initially decorate zonohedra when considering tilings with finite-sized tiles. Danzer\textsuperscript{29} proposed the \textit{ABCK}-tiling by using four types of tetrahedra as prototiles. Both the Socolar-Steinhardt and the Danzer tilings are equivalent and characterized by the inflation factor of $\tau$. The detailed description may be found in literature.\textsuperscript{29-34} In contradistinction, our tiling has the inflation factor of $\tau^3$ with vertices probably forming a subset of those of the \textit{ABCK}-tiling. This becomes evident by examining more closely the inflation/deflation rules for edges. After scaling by a factor of $\tau^3$, the second type edge decomposes into its own reduced copy and triacontahedron. Thus, after scaling by a factor of $\tau$, one of the vertices unavoidably falls inside the triacontahedron. Therefore, further investigations are needed to establish the exact correspondence between our substitutional algorithm and the higher-dimensional approach.

Now the question arises on how to fill the initial four types of quasi-unit cells with specific atoms in order to obtain the solids with the given local atomic arrangement? The chains of atomic clusters in the shape of triacontahedra often occur in icosahedral quasicrystals.\textsuperscript{35,36} For example, the qualitative resemblance with an actual structure of the $\text{Al}_{27}\text{Cu}_{48}\text{Li}_2$ icosahedral phase seems clear at the first glance (compare Figs. 2e and 7c above with Fig. 20 in the paper by Audier and Guio\textsuperscript{15}). For other structural types, the icosahedra and interpenetrating icosahedra chains are more common. The question is whether such structures may be also generated by the unique substitutional algorithm starting from the single triacontahedron? Suppose, we have properly placed the Mackay-, Bergman-, or Tsai-type clusters\textsuperscript{11,37} within the initially empty triacontahedron. Its vertices are initially not occupied by any atoms, too. After that, we have to fill the remaining three types of zonohedra with atoms in a definite way in order to obtain the consistent structure compatible with the global symmetry. In general, this problem remains open.

Next question is whether the above results may be helpful in the practical refinement of the specific structures. At the first step of structure analysis, the corresponding crystalline approximants are often used instead of quasicrystals.\textsuperscript{38,39} Recall that the approximant is a compound whose composition and structural units are very similar to those of a quasicrystal, but are nevertheless a crystal. The proposed deflation rules may help in deriving suitable models with giant supercells. On the other hand, we have to issue a warning – our results clearly indicate that one can encounter a principal difficulties when trying to use the $\tau^i$ times enlarged golden rhombohedron (Fig. 3g) immediately as the unit cell of the corresponding approximant and vice versa. Indeed, in order to form the crystalline lattice, the rhombohedra in the structure of corresponding crystalline approximant must be translated and placed side-by-side. The opposite sides of such rhombohedra must be equivalent to enable the mere possibility of arranging them together by the crystalline manner. In contrast, in the quasicrystalline packings, the opposite sides of rhombohedra must be inequivalent to ensure the quasicrystalline order.

The significance of the proposed substitution rules can be illustrated as follows. For example, Abe \textit{et al}\textsuperscript{40} postulated that “quasicrystals cannot be defined as packing of identical unit cells,” so that their structures can be effectively viewed only in terms of packing by overlapping clusters that are the most stable, energetically favored atomic configurations. Bourdillon\textsuperscript{41} has the opinion that “…mathematically, it is as easy to multiply cells as to multiply dimensions, but neither contributes to understanding why and how quasicrystals form” referring the multiple cells approach to as “multiple cells higgledy-piggledy.” We hope that substitution rules proposed by us may bring a new perspective to the multiple cell approach and turn it into a much more effective tool in structure analysis.

Our results may be useful not only when describing quasicrystals, but also in the structure prediction and analysis of hollow cage and core-shell clusters, cluster aggregates, inorganic fullerenes and fullerene-like nanoparticles, as well as in designing appropriate molecular building blocks for artificial molecular quasicrystals.\textsuperscript{42-44} As a conclusion, we offer clear substitution rules for icosahedral packings that make it possible to fill the entire space with golden zonohedra in a strictly regular manner without addressing to higher dimensions. Natural local matching rules do not contradict the cut-and-project scheme. We have especially depicted the Steinhardt’s designations for faces to highlight the consistency of our approach. It is neither better nor worse than the higher-dimensional approach just another way to describe such a complicated phenomenon as quasicrystals.

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