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Cluster coverings as an ordering principle for quasicrystals

Franz Gähler*

Institut für Theoretische und Angewandte Physik, Universität Stuttgart, D-70550 Stuttgart, Germany

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Abstract

Cluster density maximization and (maximal) cluster covering have emerged as ordering principles for quasicrystalline structures. The concepts behind these ordering principles are reviewed and illustrated with several examples. For two examples, Gummelt's aperiodic decagon model and a cluster model for octagonal Mn–Si–Al quasicrystals, these ordering principles can enforce perfectly ordered, quasiperiodic structures. For a further example, the Tübingen triangle tiling (TTT), the cluster covering principle fails to enforce quasiperiodicity, which sheds some light on the limitations of this approach.

Keywords: Quasicrystals; Clusters; Tilings; Coverings

1. Introduction

The formation and stabilization of quasicrystals is one of those problems which are still poorly understood. Different approaches by various authors have been used to explain how the observed quasicrystal structures could arise as states of minimal free energy. Some have concentrated on minimizing internal energy by using interactions mimicking matching rules for the underlying tilings, others have concentrated on the maximization of entropy in the random tiling approach. Both approaches have their problems and are not entirely convincing. In this situation, the observation that certain quasicrystal structures can be obtained by maximizing the density of a few well chosen clusters [1,2], or even by requiring a covering by overlapping copies of a single cluster [3–5], brought an entirely new aspect into the discussion. Since the internal structure of a covering cluster imposes constraints on the possible cluster overlaps, these overlap conditions impose also constraints on the possible structures that can be covered. For suitably chosen clusters, the overlaps can generate quasiperiodic order, and provide in this way a mechanism for the propagation of quasicrystalline order.

In a way, the overlapping constraints of the clusters are a particularly efficient kind of matching rules. Usually, matching rules for a quasiperiodic tiling are given by a list of all allowed local neighborhoods. They are translated into an energetic model for the tiling by giving all allowed local neighborhoods a lower energy than all the disallowed ones. The ground state should then be a structure satisfying the matching rules, i.e. a perfect quasiperiodic tiling. However, favoring *all* allowed local neighborhoods over *all* disallowed ones is, for a realistic atomic structure, a hopelessly complicated task. Such an approach is therefore not realistic, and this is the main problem of the matching rule approach.

This approach neglects the fact, however, that not all local neighborhoods are equally important for the structure. A finer distinction between *allowed* or *not allowed* is needed. Many quasiperiodic tilings have *characteristic clusters* of tiles that occur very frequently and may even cover the whole structure. Such clusters must therefore be very important for the structure, and must be the energetically most favorable configurations. The system will try to maximize their density. Clusters will have to overlap, which restricts their relative positions and orientations. The constraints imposed by the possible cluster overlaps therefore create order, even long-range order.

There are several ways to turn these ideas into an *ordering principle* for quasicrystals. Actually, one can distinguish three different, but closely related ordering principles:

1. *Cluster density maximization principle*: Find simple clusters such that the maximization of their density leads to a perfectly ordered quasicrystal.
2. *Cluster covering principle*: Find simple clusters such that every structure covered by them is an ordered quasicrystal.
3. *Maximal cluster covering principle*: Combine the two previous principles: to obtain an ordered quasicrystal,

* Tel.: +49-711-685-5260; fax: +49-711-685-5271.
E-mail address: gaehler@itap.physik.uni-stuttgart.de (F. Gähler).

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maximize the cluster density among all structures covered by the cluster.

Overlapping clusters share the atoms contained in their overlap. If there is much overlap, which seems necessary for the enforcement of perfect quasicrystalline order, this results in considerable double counting of the configurational energy in a simple cluster density maximization approach. Consequently, simply adding up the cohesion energies of overlapping clusters is not entirely suitable. The second approach seems better in this respect. It basically requires that every atom is contained in some energetically favorable local configuration. The third approach is a refinement of the second one: it requires a cluster *covering*, but prefers among those the ones with the higher cluster density. In a covering with higher cluster density, it is more likely that atoms are well in the interior of some cluster, which might be better than just on the cluster surface. Whatever variant of a cluster model is chosen, it can be regarded as a particularly efficient realization of a kind of matching rules. The primary advantage of these cluster models is that they concentrate on the local neighborhoods that are really important for the structure (the clusters), and neglect the rest, thereby avoiding unnecessary complexity.

Of course, depending on the chosen covering cluster, the class of covered structures can contain more than just perfectly ordered tilings. In fact, clusters which can enforce a perfectly ordered tiling will be rather rare. Much more typical will be clusters that can cover *super-tile random tilings* [1,2]. These are random tilings with big tiles (super-tiles), which are usually obtained by inflating the original tiles several times. Such super-tile random tilings look rather perfect on a local scale, but globally they are random tilings with a positive entropy density at zero temperature. In this way, also for the cluster models it is possible to obtain random tiling type models in a very natural way.

In the following, we shall illustrate these concepts with three examples from the literature: Gummelt's aperiodic decagon [3,4], an octagonal cluster model for Mn–Si–Al [6], and the Tübingen triangle tiling (TTT) [7].

2. Gummelt's aperiodic decagon

Whereas the earlier papers on cluster maximization did not attract much attention, the discovery of an aperiodic decagon by Gummelt [3,4] convinced many people that the cluster approach might be worth to be considered more seriously. The aperiodic decagon (Fig. 1) has a coloring which restricts the possible overlaps to two kinds. Gummelt [4] could prove that the only structures that can be covered by the aperiodic decagon are equivalent to perfect Penrose tilings. A larger patch of a covering is shown in Fig. 2.

There are several models of decagonal quasicrystals [8–11] which in one way or another have been described as cluster coverings, with a cluster which is a decoration of Gummelt's aperiodic decagon. Apart from minor details perhaps, all these models are, just like the different variants of the Penrose tiling, mutually locally derivable (MLD) from each other [12] (often, this property is also called local equivalence). For this reason, they cannot be regarded as being essentially different. With respect to the classification of ordering principles given in the introduction, all these cluster models are based on the cluster covering principle, and their ground state is a structure equivalent to a perfect Penrose tiling. Jeong and Steinhardt [5] could show, however, that among all tilings with the two Penrose rhombi, the Penrose tiling is the one with the highest density of Gummelt's aperiodic decagon cluster. This holds true, in particular, also for tilings which are not covered by decagons. Therefore, the cluster models based on Gummelt's aperiodic decagon are just as well characterized by the cluster density maximization principle, or even the maximal cluster covering principle. For these models, there is no distinction between the different approaches.

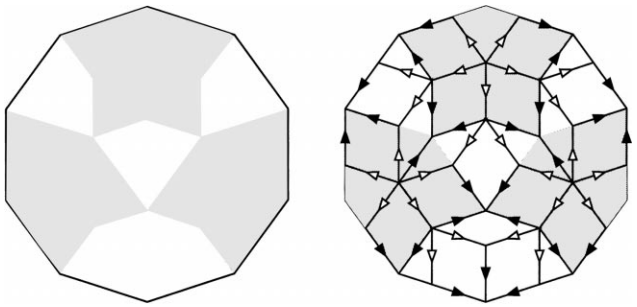

Fig. 1. Gummelt's aperiodic decagon. On the right, an equivalent patch of a Penrose tiling is superimposed. Decagon coloring and tiling patch impose the same overlapping constraints.

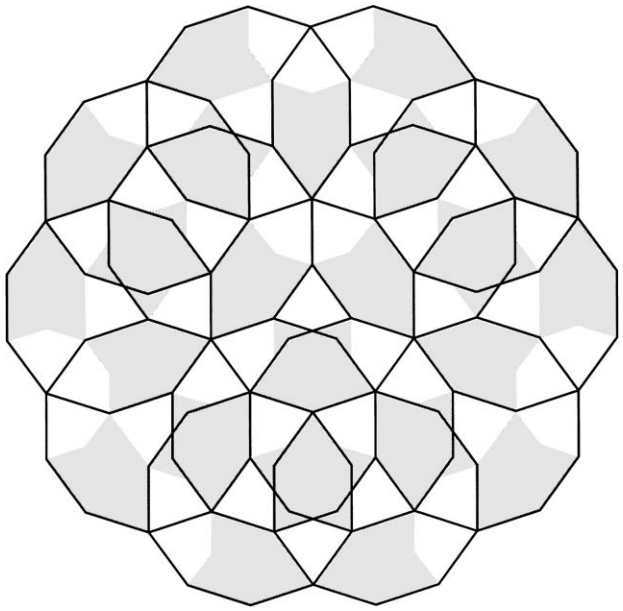

Fig. 2. A patch of Gummelt's decagon covering. The coloring allows two kinds of decagon overlaps.

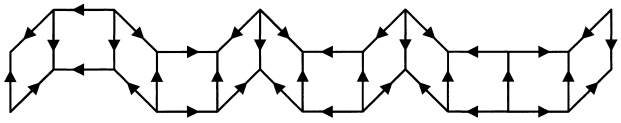

Fig. 3. The alternation condition requires that along a lane of tiles the two kinds of rhombi alternate. It can be enforced by an arrowing of the tiles.

3. An octagonal cluster model

In many respects, the octagonal Ammann–Beenker tiling [13,14] is among the simplest of all quasiperiodic tilings. To some extent this is true also for its cluster descriptions. We shall therefore use it as our second example to illustrate the general principles of cluster models. Matching rules enforcing a perfect octagonal tiling are rather complicated [14–16]. They are expressed in terms of a non-local decoration of the tiling. However, in order to make a cluster model work it is sufficient to build it upon a local subset of these matching rules. This is the alternation condition [17], which requires that along any lane of tiles the two types of rhombi have to alternate (Fig. 3). It is enforced by a suitable arrowing of the tiling edges. The alternation condition cannot enforce perfect octagonal tilings, but it does enforce *perfectly ordered*, quasiperiodic tilings, which are at least four-fold symmetric [18]. In fact, tilings satisfying the alternation condition are all members of a one-parameter family of four-fold symmetric, quasiperiodic tilings. The unique member of this family with even eight-fold symmetry is the Ammann–Beenker tiling.

The alternation condition can therefore be used to obtain the Ammann–Beenker tiling with a cluster maximization principle. The idea is to prefer clusters which favor the alternation condition. This is the case for the two clusters shown in Fig. 4, called the octagon and the ship. It has been shown numerically [2], that if these two clusters are energetically preferred, with suitable relative energies, the ground state is a perfectly ordered tiling with eight-fold symmetry. The relative weights of the two clusters have to be chosen such that among all tilings satisfying the alternation condition, the octagonal tiling has the lowest energy. This is the case for weights in a wide interval, so that the phenomenon is very robust [2]. If only octagon clusters are preferred, however, we arrive, at fixed stoichiometry, at a supertile random tiling structure [2]. The ship cluster therefore is really needed to enforce the alternation condition. It propagates the information on the orientation of rhombi across two adjacent squares.

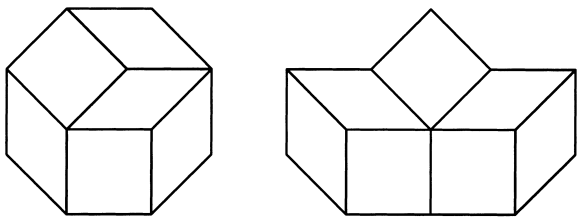

Fig. 4. Two clusters to be energetically preferred, the octagon and the ship.

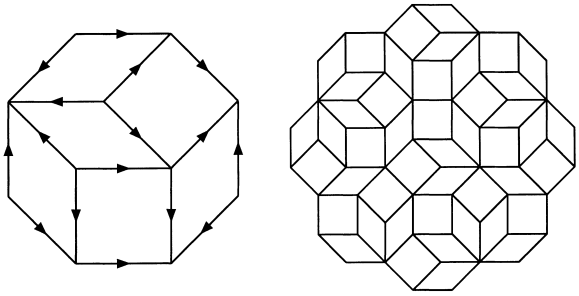

Fig. 5. Arrowed octagon cluster, and inflated unarrowed octagon cluster. Both impose the same overlapping constraints.

Later on it was realized [19] that if the octagon cluster is *arrowed* (Fig. 5), the situation improves considerably. A tiling completely covered by arrowed octagons must necessarily satisfy the alternation condition, and among these the Ammann–Beenker tiling has the highest octagon density. A cluster model can therefore be based on the maximal cluster covering principle. Under the (plausible) assumption that tilings that are not completely covered by the cluster cannot have a higher cluster density than the maximal density for tilings that are covered, the arrowed octagon can even be used in a cluster density maximization principle to enforce the octagonal Ammann–Beenker tiling. Although there is no proof of the above assumption, it appears very unlikely that a tiling which is not completely covered can have a higher cluster density. In any case, among all square rhombus tilings covered by arrowed octagons, the Ammann–Beenker tiling is the unique structure with the highest cluster density.

If undecorated clusters are preferred for some reason, one can inflate the octagon once to arrive at a larger cluster, which has exactly the same asymmetries as the arrowed octagon (Fig. 5). This larger, undecorated cluster therefore imposes the same overlapping constraints, and can be used in place of the arrowed octagon. In fact, arrowed and unarrowed tilings are in the same MLD class, or locally equivalent. The arrowing just reduces the size of the cluster that is needed. One should keep in mind, however, that real quasicrystals are not tilings, but are, at best, decorations of a tiling. To obtain a quasicrystal, the tiling has to be decorated with atoms, and it could also be this atomic decoration which introduces the necessary asymmetry and thus imposes the necessary overlapping constraints.

This is indeed the case for the quasicrystal structure of octagonal Mn–Si–Al described by Jiang et al. [20], as has been discussed in detail recently [6]. This quasicrystal is a layered structure $\ldots$ ABAB$'\ldots$, and can be regarded as a decoration of the Ammann–Beenker tiling. The decoration of the octagon motifs (which cover the whole structure) is shown in Fig. 6. Both the decoration of the edges in layer A and the decoration of the interiors of the squares in layers B and B$'$ show the same asymmetry as the arrowing. There are actually two possible decorations of an octagon, whose only

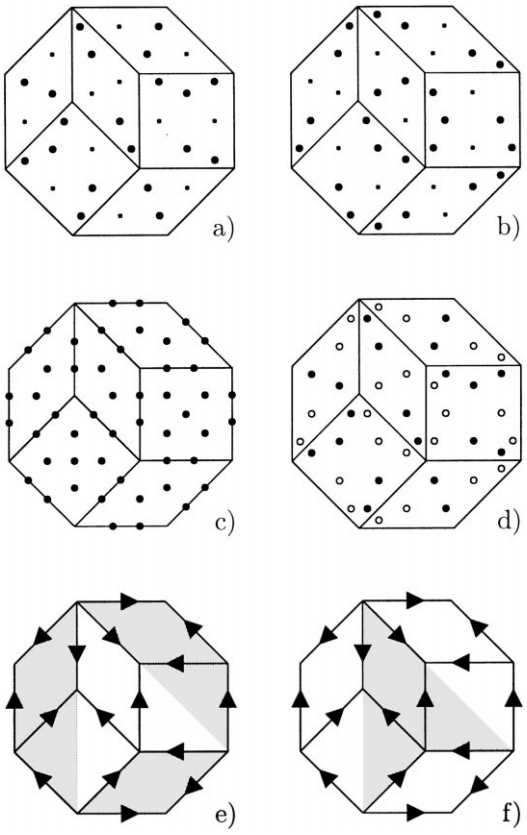

Fig. 6. Decoration of a small octagon patch. (a) Layer B, (b) layer B′, (c) layer A and (d) layers B and B′ together. Large dots denote Mn atoms, small dots Si or Al atoms. In (d), only Mn atoms are shown; atoms from the B layer are shown as full dots, and atoms from the B′ layer as open dots. In (e) and (f), abstract representations of the layer stackings ABAB′ and AB′AB are given, respectively.

difference is that the decorations of the layers B and B′ are exchanged. One kind of octagon is decorated with a stacking . . . ABAB′ . . . , the other with a stacking . . . AB′AB . . . Since each octagon actually represents an infinite prism with octagonal base, these two decorations correspond to prisms which are shifted by half a lattice period in *z*-direction with respect to one another, but are identical otherwise. We therefore have a covering by identical prisms, which, of course, can be chopped into identical, finite clusters.

It is most convenient to represent these prisms again by an abstract decoration of the octagon. The different vertical positions of the prisms are encoded by a coloring, as shown in Fig. 6e and f. Tiles which differ in color, but have otherwise the same decoration, correspond to prisms shifted by half a lattice period in *z*-direction. It is interesting to note that since the octagonal prisms occur at two different positions in *z*-direction, the Bravais lattice of this octagonal quasicrystal must be an octagonal centered one [6]. This can also be seen in the colored and arrowed tiling of Fig. 7: if a tile is a translate of another tile by an odd number of tile edges, it has the opposite color. If an even number of tile edges separates the two tiles, they have like colors. In order to obtain a lattice translation, a (horizontal) translation by an odd number of tile edges must be combined with a translation in *z*-direction by half a lattice period, in order to make up for the color change. This results in the octagonal centered lattice.

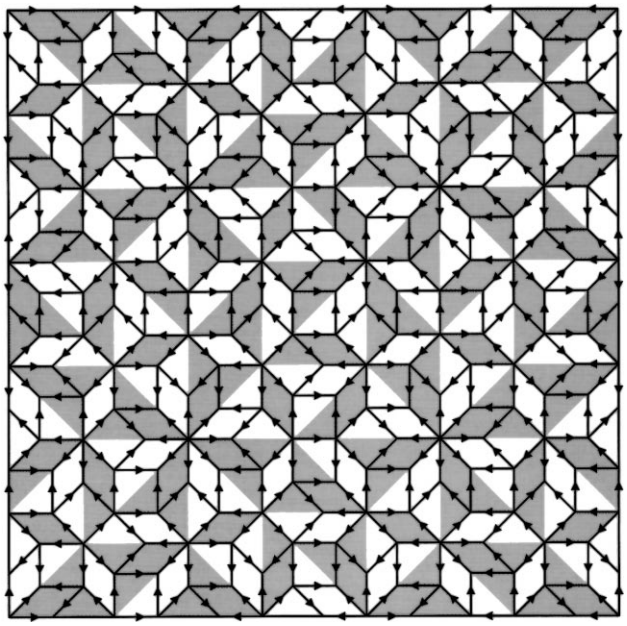

Fig. 7. Colored and arrowed Ammann–Beenker tiling.

4. How typical are covering clusters?

In the previous two sections we have seen two examples where the cluster covering principle works perfectly well to explain the stability of the corresponding quasicrystals. The question now arises whether these examples are rare exceptions, or whether such a phenomenon is, on the contrary, quite typical. One should distinguish between two different questions, however. One is the question of the mere existence of a covering cluster, and the other whether a cluster covering can enforce quasiperiodic order.

The notion of covering clusters is actually quite old. In 1991, Burkov [21] has given a realistic model of decagonal quasicrystals which is covered by copies of large, interpenetrating clusters. It is well known, however, that these clusters can cover also supertile random tilings with the same tiles, and thus cannot enforce quasiperiodicity. For icosahedral quasicrystals, the situation is somewhat more difficult. Duneau [22] found clusters which almost cover a quasicrystalline structure, but also here it is fairly clear that no quasiperiodic structure is *enforced*. In recent papers [7,23], Kramer has argued that the existence of covering clusters actually has to be expected for theoretical reasons, at least for the canonical projection tilings. There are two kinds of such projection tilings [24]. The first kind has vertices at projected lattice positions, with an acceptance domain which

is the projected Voronoi cell of the higher-dimensional lattice. The second kind has vertices at projected corners of the Voronoi cells, with acceptance domains which are the projected, dual Delaunay cells. According to Kramer's theory, for the first kind of canonical projection tilings we should expect covering clusters at projected corners of Voronoi cells, whose size is given by the corresponding projected Delaunay cell. Conversely, for the second kind of canonical projection tilings, covering clusters are expected at projected lattice points, with a size given by the projected Voronoi cell. In other words, covering clusters and vertices are, in a sense, dual to each other. To be precise, there does not seem to be a proof that Kramer's clusters indeed cover the whole tiling without gaps in the general case, but for many particular examples this has been verified. Indeed, the two examples discussed in the previous sections are exactly of this kind. Gummelt's decagon covering the Penrose (rhombus) tiling is centered at projected lattice points, whereas the vertices of the Penrose tiling are at projected corners of the Voronoi cells. In the case of the octagonal tiling, the vertices are at projeced lattice points, whereas the octagon cluster is centered at projected corners of the Voronoi cells. In both cases, the cluster size coincides with the size predicted by theory. Kramer has given as a further example the Tübingen-Triangle-Tiling (TTT) [7]. Here, the clusters are located at projected corners of the Voronoi cells, whose size correspond to projected Delaunay cells. There are four translation classes of cluster centers, and thus also four different clusters, two small and two big pentagons. Disregarding orientation, there are just two clusters, big and small pentagons (Fig. 8), which indeed cover the TTT.

Much more difficult is the question of enforcing a *quasiperiodic* structure just by the requirement of cluster covering, or at least by the requirement of a maximal cluster covering. Whereas for the octagonal tiling and for the Penrose tiling there is indeed such an enforcement, this does not seem to be the case for the TTT. It is very easy to see that the two pentagons can cover also periodic tilings, even ones with rather small unit cells. The situation improves to some extent if the pentagons are decorated. In order to formulate nearest neighbor matching rules for this tiling, there are two decorations necessary: edge decorations and vertex decorations [25]. The edge decoration can be applied

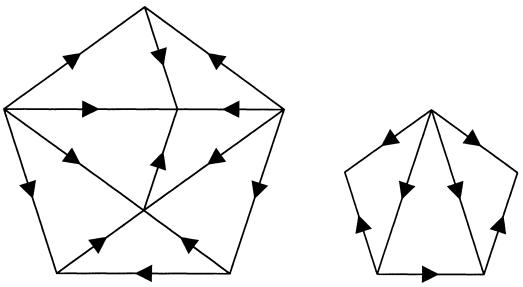

Fig. 8. Pentagon clusters covering the arrowed Tübingen triangle tiling. Both clusters, as well as their mirror images, occur in 10 orientations.

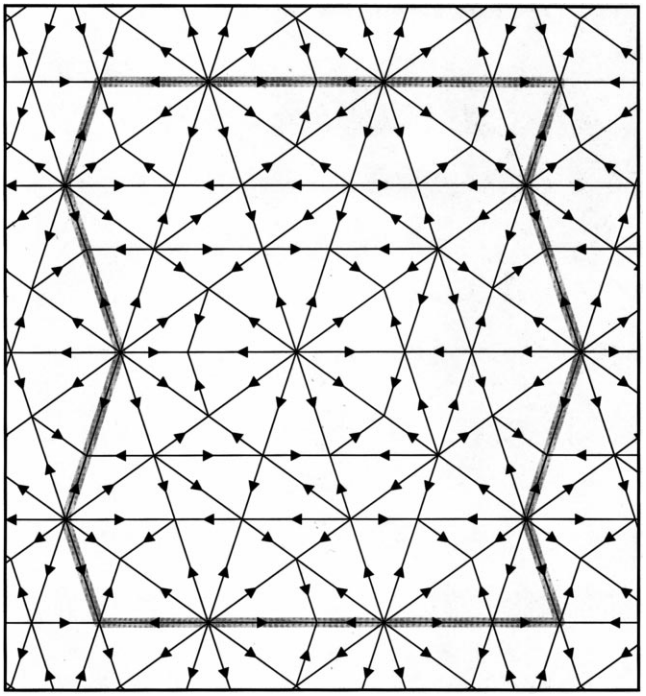

Fig. 9. Cluster covering of a periodic variant of the Tübingen triangle tiling. A unit cell is outlined in gray.

to the two pentagons in a consistent way: all pentagons of the same kind obtain exactly the same decoration. The edge decoration rules out covering of the periodic tilings with the smallest unit cells, but still admits a covering of a periodic tiling with a somewhat larger unit cell (Fig. 9). The edge decoration of the clusters alone can therefore not enforce quasiperiodicity. We should point out that passing to inflated clusters cannot improve the situation, because the edge decoration completely determines the inflated cluster. Inflation therefore cannot induce any further cluster asymmetry, and thus no further overlapping constraints. The situation would improve if also the vertex decoration is applied. This cannot be done in a satisfactory way, however, because each kind of cluster is decorated in many different ways, which means that much of the attractive simplicity of the model is lost.

5. Discussion and conclusion

In this paper we have discussed the general concepts upon which the different quasicrystal cluster models from the literature are built. These concepts have been illustrated with two instructive examples, Gummelt's aperiodic decagon, and the octagon cluster model. In fact, these are the only cluster models which are known to enforce a perfectly ordered quasicrystal. The many cluster models built upon Gummelt's aperiodic decagon are all in the same MLD or local equivalence class, and cannot be regarded as being essentially different. The question therefore arises whether and how this can be generalized to other cases. The analysis by Kramer

[7] suggests that the existence of a cluster covering is quite typical, at least for canonical projection tilings. The example of the TTT shows, however, that there is still a long way to go from the existence of a cluster covering to an enforcement of quasiperiodicity. Cluster coverings which can enforce the quasiperiodicity of a tiling or quasicrystal will be at least as rare as tilings with perfect matching rules, perhaps even rarer. One might hope, however, that in each MLD class of tilings with perfect matching rules, one can find a member with quasiperiodicity-enforcing covering clusters.

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