CLUSTER COVERINGS: A POWERFUL ORDERING PRINCIPLE FOR QUASICRYSTALS

FRANZ GÄHLER
Institut für Theoretische und Angewandte Physik,
Universität Stuttgart, D-70550 Stuttgart, Germany
e-mail: gaehler@itap.physik.uni-stuttgart.de

Cluster density maximization and cluster covering have emerged as ordering principles for quasicrystalline structures. The concepts behind these ordering principles are reviewed and illustrated with two examples, Gummelt's aperiodic decagon, and a cluster model for octagonal Mn-Si-Al quasicrystals.

1 Introduction

The formation and stabilization of quasicrystals is still not very well understood. Various approaches have been used to explain how the observed quasicrystal structures could arise as states of minimal free energy. Some authors 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,[3,4,5] 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 povide 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 the all the disallowed ones. The ground state should then be a structure satisfying the matching rules, i.e., a perfect quasiperiodic tiling. However, favouring *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 this matching rule approach.

This approach neglects the fact, however, that not all local neighborhoods are equally important for the structure. A finer distinction than *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 favourable configurations. The system will therefore try to maximize their density. This means that clusters will have to overlap, which restricts their relative positions and orientations. The constraints imposed by the possible cluster overlaps therefore create order.

There are several ways to turn these ideas into an *ordering principle* for quasicrystals:

- Find simple clusters such that the maximization of their density leads to an ordered quasicrystal.
- Find simple clusters such that every structure covered by them is an ordered quasicrystal.
- Combine the two previous strategies: to obtain an ordered quasicrystal, 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, there will be considerable double counting of the configurational energy in a simple cluster maximization approach. Therefore, simply adding up the cohesion energies of overlapping clusters is not entirely suitable. The second approach seems better suited in this respect. It basically requires that every atom is contained in some energetically favourable 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 these 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 two examples from the literature, Gummelt's aperiodic decagon, and an octagonal cluster model introduced recently.[6]

2 Gummelt's aperiodic decagon

Whereas the earlier papers on cluster maximization did not attract too much attention, the discovery of an aperiodic decagon by Petra Gummelt[3,4] convinced many people that the cluster approach might be worth to consider more seriously. The aperiodic decagon (Fig. 1) has a colouring which restricts the possible overlaps to two kinds. Gummelt could prove[4] 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.

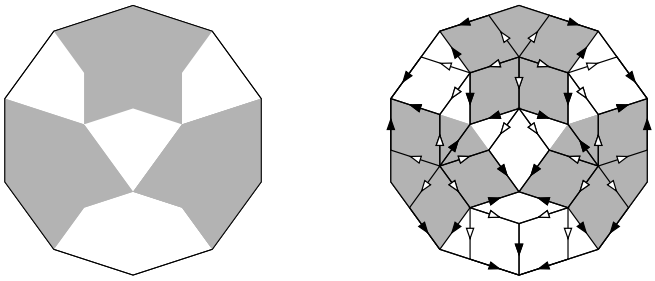

Figure 1. Gummelt's aperiodic decgon. On the right, an equivalent patch of a Penrose tiling is superimposed. Decagon coloring and tiling patch impose the same overlapping constraints.

There are several models of decagonal quasicrystals[7,8,9,10] which in one way or another have been described as cluster coverings, with a cluster which is a decoration of Gummelt's aperiodic decagon. With respect to the classification given in the introduction, these cluster models are all of the covering type, 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

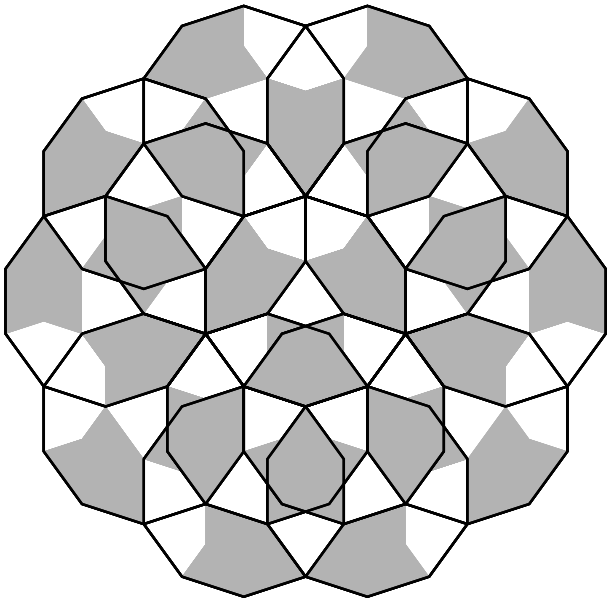

Figure 2. A patch of Gummelt's decagon covering. The coloring allows two kinds of decagon overlaps.

density of Gummelt's aperiodic decagon. This holds true, in particular, also for tilings which are not covered by decagons. Therefore, the cluster models based on Gummelt's aperiodic decagon can also be obtained by simple cluster density maximization. For these models, there is no distinction between the two approaches.

3 An octagonal cluster model

In many respects, the octagonal Ammann-Beenker tiling[11,12] 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.[13,12,14] They are expressed in terms of a complicated, 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

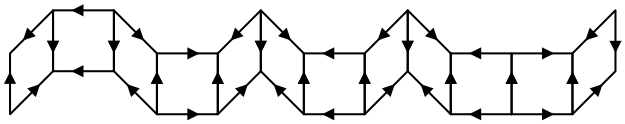

Figure 3. The alternation condition requires that along a lane of tiles the two kinds of rhombi alternate. It can be enforced by an arrowing of the tiles.

these matching rules. This is the alternation condition,[15] 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.[16] 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.

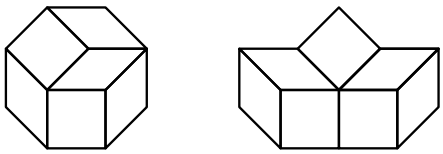

Figure 4. Two clusters to be energetically preferred, the octagon and the ship.

The alternation condition can therefore be used to obtain the Ammann-Beenker tiling with a cluster maximization principle. The idea is to favour clusters which favour 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 for the clusters, 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 this phenomenon is very rubust.[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.

Later on it was realized[17] that if the octagon cluster is *arrowed* (Fig. 5),

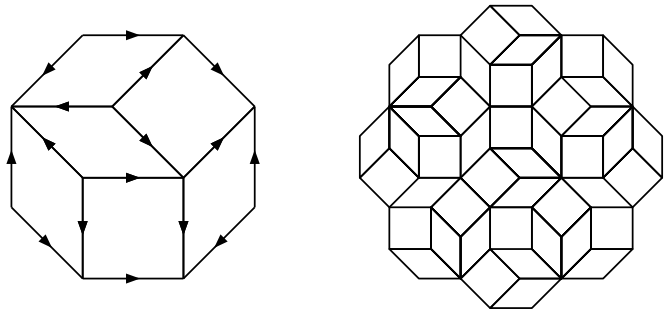

Figure 5. Arrowed octagon cluster, and inflated unarrowed octagon cluster. Both impose the same overlapping constraints.

the situation improves considerably. A tiling completely covered by the arrowed octagon must necessarily satisfy the alternation condition, and among these the Ammann-Beenker tiling has the highest octagon density. Such a cluster model is therefore of the third type of the classification given in the introduction. Under the assumption that tilings that are not completely covered by the cluster can not have a higher cluster density than the maximal density for tilings that are covered, the arrowed octagon can be used in a cluster maximization principle to enforce the octagonal Ammann-Beenker tiling. Although there is no proof for 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. One should keep in mind, however, that real quasicrystals are not tilings, but are, at best, decorations of a tiling. The tiling therefore 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, Hovmöller and Zhou,[18] 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

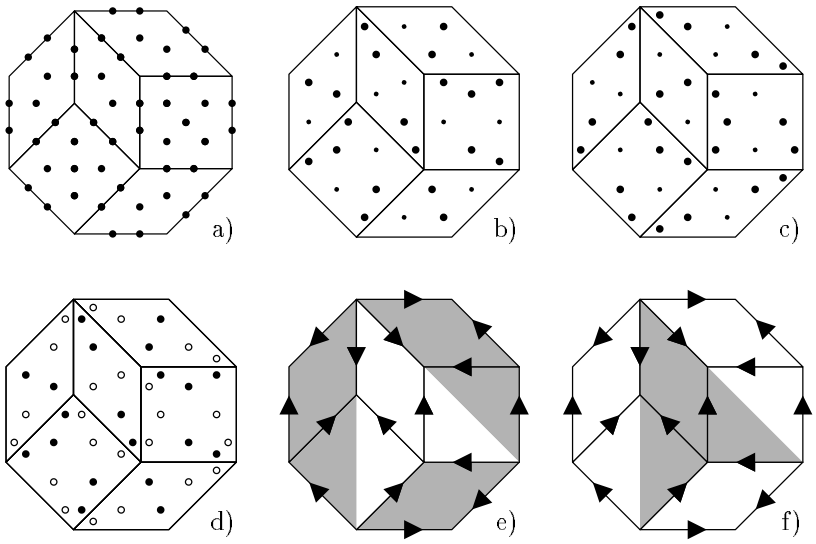

Figure 6. Decoration of a small octagon patch. a) layer A, b) layer B, c) layer B', and d) layers B and B' together. Large dots denote Mn atoms, small dots Si or Al atoms. In d), only Mn atoms are shown; atoms from the B layer are shown as full dots, and atoms from the B' layer as open dots. In e) and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

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 difference is that the decorations of the layers B and B' are exchanged. One kind of octagon is decorated with a stacking $\ldots ABAB' \ldots$, the other with a stacking $\ldots AB'AB \ldots$. 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 colouring, as shown in Figs. 6e,f. Tiles which differ in colour, but have otherwise the same decoration, correspond to prisms shifted by half

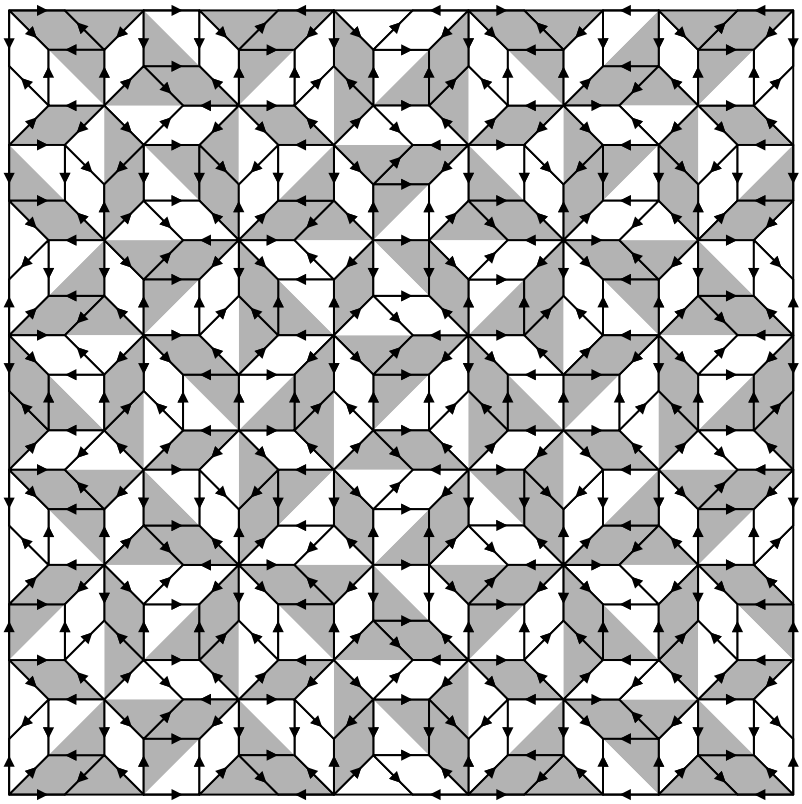

Figure 7. Colored and arrowed Ammann-Beenker tiling.

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 coloured 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 colour. If an even number of tile edges separates the two tiles, they have like colours. 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 colour change. This results in the octagonal centered lattice.

4 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 examples for which one has succeded in enforcing a perfectly ordered quasicrystal. While several authors have built their cluster models upon Gummelt's aperiodic decagon, these models can not be regarded as being essentially different. The question therefore arises whether and how this can be generalized to other cases. In a recent paper,[19] Kramer has argued that the existence of a cluster covering is quite typical, at least for canonical projection tilings. He showed what covering clusters one can expect, and at what positions they are to be found in the tiling. The examples shown in this paper are, in fact, exactly of the type discussed by Kramer.[19] In the general case, however, it is still a big step from finding a covering cluster (or two or three, which cover together) to finding clusters which enforce a perfectly ordered tiling. Some experimentation with the Tübingen Triangle Tiling, which is covered by two kinds of pentagons,[19] incdicates that these clusters cannot enforce an ordered tiling, even if they are decorated.

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