Cluster Models of Decagonal Tilings and Quasicrystals

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Abstract

Two different relaxed versions of Gummelt's aperiodic cluster covering rules are considered. These relaxed covering rules produce certain random tiling type structures, which are precisely characterized, along with their relationships to various other random tiling ensembles. One variant of the relaxed covering rules allows for a natural realization in terms of a vertex cluster. It is shown with Monte Carlo simulations that the structures with maximal density of this cluster are the same as those produced by the corresponding covering rules. The entropy density of this covering ensemble is determined by Monte Carlo simulations, using entropic sampling techniques. Perfectly ordered structures, like those produced by Gummelt's perfect covering rules, can be obtained in our model if a coupling between neighboring clusters is introduced. This coupling can order the random tiling type structures to perfectly ordered quasicrystals.

Keywords: Quasicrystals; Clusters;

1. Introduction

Many quasicrystals are completely covered by overlapping copies of a single cluster. Two clusters which overlap must agree in the overlap region, which restricts the possible relative positions and orientations of the clusters. Cluster overlaps therefore create order, in favorable cases even perfect quasiperiodic order. This observation can be used to formulate several variants of an ordering principle for quasicrystals (for a review, see [1]). A perfect quasicrystal can be obtained either by requiring that a given cluster completely covers the structure, or that the cluster has maximal density in the structure, or that it covers the structure with maximal density. With such ordering principles, perfect quasiperiodic order could be obtained for decagonal [2, 3, 4], octagonal [5], and dodecagonal [6] quasicrystals. The same ordering principles can also be used to obtain supertile random tiling structures, which are preferred by the ordering principles whenever the chosen cluster is not selective enough, in the sense that it allows too many different overlaps [1]. This happens, in particular, if the cluster is too small to restrict the number of overlaps [7], or if it is too symmetric. In this respect, it is interesting to note that *asymmetric* clusters seem to be preferred by the electronic structure in decagonal quasicrystals [8].

In this paper, we shall concentrate on cluster models for decagonal tilings and quasicrystals, making use of the ordering principles mentioned above. For decagonal structures, Gummelt's aperiodic decagon [2, 3] provides a striking example where perfect quasiperiodic order can be obtained by a simple cluster covering principle. This example was so convincing, that many researchers have tried to map their experimental structures to Gummelt's aperiodic decagon, even though the fit in the overlap was often not perfect (see, e.g. [8]), and the overlapping constraints not exactly equivalent. However, many experimental decagonal quasicrystal structures are not perfectly quasiperiodic, and it is therefore interesting to consider also overlap rules which are less restrictive than Gummelt's, and which do not enforce a perfectly ordered structure, but rather a (supertile) random tiling structure. The analysis of such relaxed overlap rules and their corresponding structures will be the main topic of this paper, which is organized as follows. In Section 2, two different relaxed versions of Gummelt's overlap rules are introduced, and the structures they produce are precisely characterized, along with their relationships to various known ran-

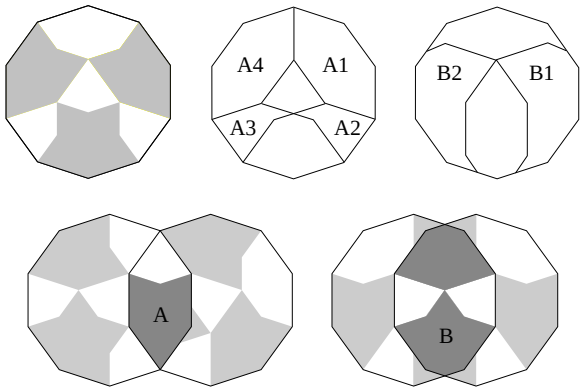

Fig. 1. Gummelt decagon (top left), with the allowed overlap zones for A- and B-overlaps (top middle and right), and representative A- and B-overlaps (bottom).

dom tiling ensembles. In Section 3, we introduce a vertex cluster whose structure imposes the previously discussed overlapping constraints in a natural way. It is shown by Monte Carlo simulations that the structures with maximal density of this cluster are the same as the structures produced by the overlap rules. In Section 4, the entropy density of the set of states with maximal cluster density is determined by entropic sampling techniques. In Section 5, a coupling between neighboring clusters is introduced, and it is shown that this coupling is capable of ordering the random tiling structures maximizing the cluster density to perfectly ordered structures. In Section 6, we finally conclude by discussing the relationship of our models with experimentally observed structures.

2. Cluster coverings for perfect and random Penrose tilings

It is well known that each covering of the plane by Gummelt's aperiodic decagon (Fig. 1) is equivalent to a perfect Penrose tiling [2, 3], if the covering has the property that, whenever two decagons overlap, their colorings agree in the entire overlap region. More precisely, the decagon centers of such a covering form the vertex set of a Penrose pentagon tiling (PPT), and every PPT can be obtained in this way from exactly one covering satisfying the Gummelt overlap rules. This is a one-to-one correspondence between PPTs and Gummelt coverings, which moreover is local. As the Gummelt decagon represents a cluster in the corresponding quasicrystal, we will often use the term cluster for the covering decagon.

In order to allow for partially disordered coverings, Gummelt and Bandt [9] have proposed to relax the overlap rules to some extent. To understand the type of relaxation, recall that if the perfect rules are obeyed, a decagon may have small A-overlaps with neighbor decagons in four possible directions, and

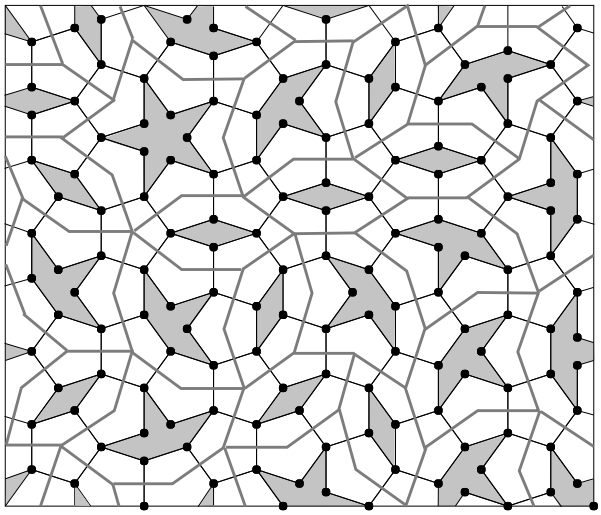

Fig. 2. Random Penrose pentagon tiling. Spiky tiles (shaded in gray) are surrounded by pentagons. This tiling is equivalent to a HBS tiling (drawn in gray), whose tile edges connect centers of neighboring pentagons.

bigger B-overlaps with neighbor decagons in two possible directions (Fig. 1). The coloring in the overlap region has an orientation, which must be respected. All possible overlaps are therefore *oriented*. As a relaxation of the perfect rule, Gummelt and Bandt have now proposed [9] to abandon this orientation constraint, and to retain only the condition which specifies the possible *overlap zones*, without orientation inside. This overlap rule will be referred to as the fully relaxed rule.

There is a natural intermediate rule between the perfect rule and the fully relaxed rule. In this rule, which will be just called the *relaxed rule*, the orientation condition is abandoned only for the small A-overlaps, but is retained for the larger B-overlaps. This intermediate rule and the resulting structures will be the main topic of this paper.

Gummelt and Bandt have shown [9] that every covering satisfying the fully relaxed rule has the property that its set of cluster centers forms the vertex set of a random PPT having the additional property that all the spiky tiles (stars, ships, and rhombi, shaded in gray in Fig. 2) are completely surrounded by pentagons (in the following, when we say random PPT, we always mean one satisfying this extra condition; more general ones won't play any role here). Such a random PPT is equivalent to a random hexagon-boat-star (HBS) tiling, drawn in gray in Fig. 2. Since coverings satisfying the more restrictive relaxed rule satisfy also the fully relaxed rule, their set of cluster centers also forms the vertex set of a random PPT. Conversely, it is easy to see that every random PPT can arise both from relaxed and from fully relaxed coverings. The only differ-

ence between relaxed and fully relaxed coverings is the number of coverings associated with a given random PPT.

To see this, we note that the orientation of a cluster is already determined by the presence of two B-neighbors, or the presence of four A-neighbors. In Fig. 2 we see that A-neighbors are separated by an edge of a tile, or a long diagonal across a ship or a star, whereas B-neighbors are separated by a short diagonal of a rhombus, ship or star. The only vertices whose cluster orientation is not fixed by the tiling are the obtuse corners of the rhombi, where two cluster orientations are possible. For the fully relaxed rule, where no orientation conditions are to be observed, we therefore have altogether four choices per rhombus for the cluster orientations.

For the relaxed rule, which has to obey the orientation condition for the B-overlaps, one can easily verify that the orientation condition for B-overlaps across ships and stars is always satisfied. In order that the same is true for B-overlaps across rhombi, however, the orientation of the cluster on the obtuse rhombus corners can not be chosen independently. The orientation condition is satisfied only for two of the four possible combinations.

Summarizing, for a given random PPT we have four independent choices per rhombus for a fully relaxed cluster covering, and two choices per rhombus for a relaxed cluster covering. In the same way, one can quantify the relationship between the cluster coverings and certain variants of a random Penrose rhombus tiling. The random HBS tilings arise from random Penrose rhombus tilings which still satisfy the double arrow condition [10, 11]. Such random Penrose rhombus tilings are also called 4-level Penrose random tilings. The double arrowed edges of the 4-level random Penrose tilings are simply wiped out to obtain the HBS random tilings, which are also known as 2-level Penrose tilings [10]. The relationship between 4-level and 2-level Penrose random tilings is not one-to-one: whereas the subdivision of boats and stars is unique, there are two choices for the subdivision of each hexagon into rhombi, just as there are two possible cluster assignments on the obtuse rhombus corners in the PPT. Since rhombi in the PPT and hexagons in the HBS tiling are in one-to-one correspondence, this implies that the multiplicity of relaxed cluster coverings and 4-level Penrose random tilings related to a given random PPT is the same.

3. Cluster density maximization

In the last section we have considered cluster *coverings*, where our clusters have simply been decagons with certain overlap rules. Another variant of the ordering principle is *cluster density maximization*. The

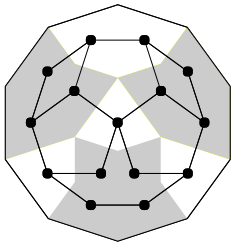

Fig. 3. Vertex cluster superimposed on Gummelt decagon. This cluster enforces the relaxed overlap rules.

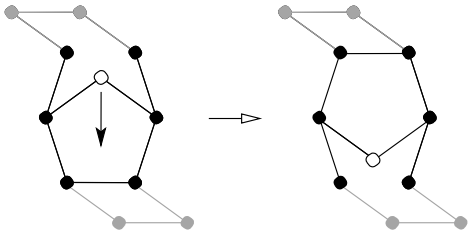

Fig. 4. Flip move for Monte Carlo simulation.

relaxed overlap rule allows for a very natural realization in terms of a vertex cluster in a random PPT (we still require that spiky tiles are completely surrounded by pentagons). This vertex cluster is shown in Fig. 3, superimposed on a Gummelt decagon. The tile edges are drawn only as a guide to the eye; they are not part of the cluster, only the vertex set counts. It is easy to see that the orientation of the A-overlaps of the Gummelt decagon can not be enforced by the vertex set of the cluster, whereas the orientation of the B-overlaps is enforced. The A-overlap consists of a rhombus, and the B-overlap of a hexagon with an extra vertex in asymmetric position inside.

With this cluster we can now build a statistical model of cluster density maximization. We consider the set of all random PPT, and assign a weight to each tiling, which is simply the number of vertex clusters it contains. With Monte Carlo simulations it is then possible to find the subensemble of those random PPTs which have maximal cluster density. For the simulations we will need a Monte Carlo dynamics which is ergodic in the ensemble of all random PPTs. We have found that the flip move shown in Fig. 4 has the required properties. By repeated flips it is possible to turn any random PPT into any other. One must be careful, however, not to execute any flips which would introduce new kinds of tiles. This can be avoided if some local constraints on the flips are obeyed.

With such a Monte Carlo model, the states of maximal cluster density have been determined with simulated annealing, using as energy function the negative of the number of clusters, thus mimicking the total cohesion energy of the clusters. It turns out that these states of maximal cluster density are pre-

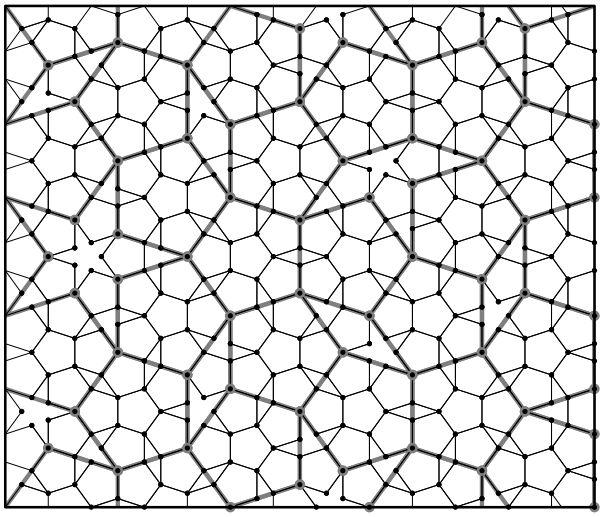

Fig. 5. Structure with maximal cluster density. The cluster centers form the vertices of a supertile random PPT.

cisely the supertile random PPTs, whose tiles have an edge length τ^2 times that of the small tiles. An example of such a supertile tiling is shown in Fig. 5. In view of the results of the previous section, this is of course not too surprising. The cluster centers sit on the vertices of the supertile tiling, covering all vertices of the small tiles. Since the vertex cluster is smaller than the Gummelt decagon, it does not cover the whole area, but only the vertices. There remain small pentagons uncovered, which sit at the center of the supertile pentagons. This does not affect the overlap constraints, however. Our results therefore imply that there is a one-to-one correspondence between cluster coverings satisfying the relaxed rule, and structures with maximal density of the vertex cluster. Although these two ordering principles are very similar, they are conceptually slightly different and must be distinguished.

4. Entropy density

With the cluster model of the previous section, it is also possible to measure the entropy density of the ensemble of structures with maximal cluster density, and thus the entropy density of the relaxed cluster covering ensemble. We have an energy model which assigns a cohesion energy to each cluster in the structure. In this model, the ground state, the state of maximal cluster density, consists of supertile random PPTs, with an extra weight of two per rhombus, because for each rhombus there are two choices of a cluster configuration with the same number of clusters. At infinite temperature, on the other hand, we have the full random PPT (with the small tiles). With entropic sampling techniques [12, 13], it is now possible to determine the entropy of the system as

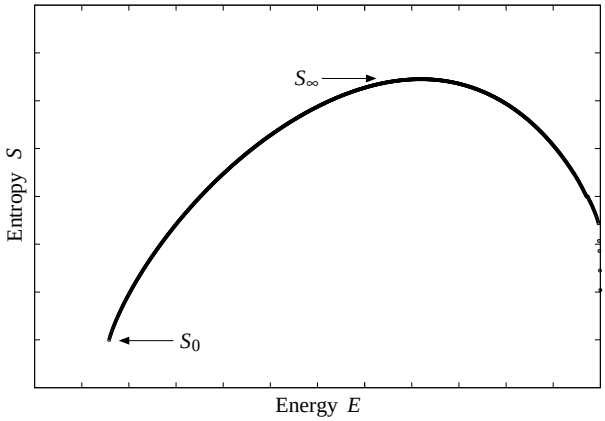

Fig. 6. Entropy as a function of energy for a selected approximant.

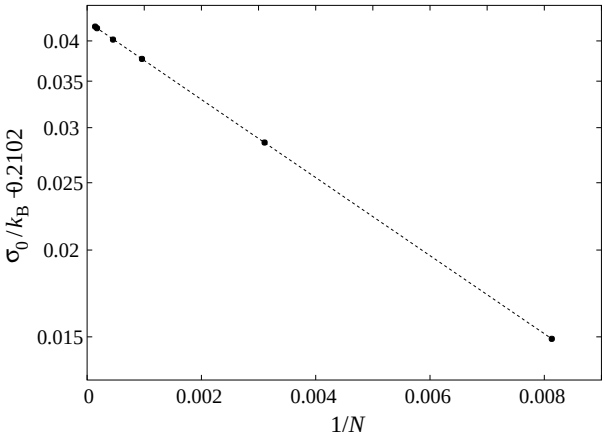

Fig. 7. Finite size scaling of the entropy density.

a function of energy, and in particular the difference in entropy between the ground state and the infinite tempererature state. An example of such an entropy function is shown in Fig. 6. The entropy of the ground state can be extracted in the following way. The entropies at zero and infinite temperature are both entropies of random PPT, once with and once without extra degeneracy for each rhombus. Moreover, the two random tilings are at different scale. Taking both differences into account, we arrive at the following relation for the two entropies densities:

$$\sigma_0 = \tau^{-4}(\sigma_\infty + \rho_{\mathrm{rh}} k_{\mathrm{B}} \ln 2) \qquad (1)$$

where ρ_{rh} is the measured rhombus density in the high temperature state. If we now write $\sigma_\infty = \sigma_0 + \Delta\sigma$, we obtain an equation for the ground state entropy denstiy σ_0, in which all other quantities are known. The ground state entropy density has been determined in this way for several periodic approximants. By finite size scaling, this can then be extrapolated to infinite system size. This is shown in Fig. 7, where one can see that the scaling works very nicely. At a scale where the supertile edges (which

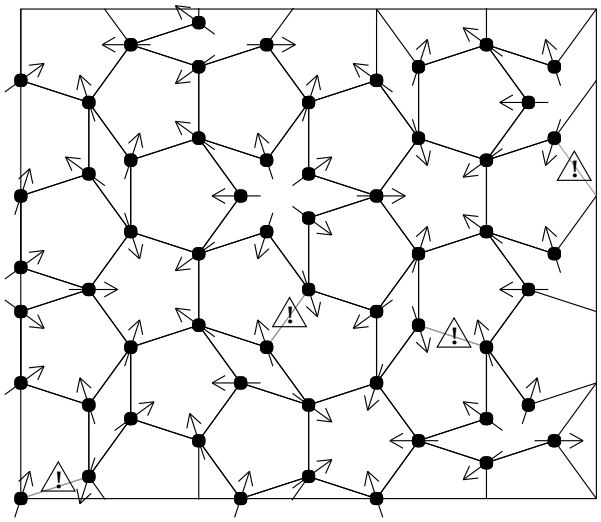

Fig. 8. Supertile tiling with cluster orientations indicated by arrows.

separate A-overlap neighbors in the cluster model) have unit length, we obtain a value of

$$\sigma_0/k_B = 0.253 \pm 0.001 \tag{2}$$

for the entropy density. This value can be compared with the value Tang and Jaric [11] have obtained for the entropy density of the 4-level random Penrose tiling. In Section 3 we have seen that 4-level random Penrose tilings are in one-to-one correspondence with relaxed cluster coverings. If the different scales of the two tilings are corrected for, Tang and Jaric obtained a value of

$$\sigma_0/k_B = 0.255 \pm 0.001 \tag{3}$$

which is compatible with ours.

5. Couplings between clusters

The only difference between the perfect and the relaxed overlap rule is that the latter does not require *oriented* A-overlaps. Since not all relaxed coverings are perfect, there must be A-overlaps in an relaxed covering which do not obey the orientation condition of the perfect rule. A closer analysis shows [9] that there is actually only one kind of disoriented A-overlap which can actually occur in a relaxed covering. If we represent the orientation of a cluster on a vertex of a supertile PPT by an arrow, we can represent the covering in a much more compact way, as shown in Fig. 8. The disoriented A-overlaps not permitted by the perfect rule are marked in Fig. 8. The corresponding tile edges have antiparallel arrows at their ends.

This representation suggests to introduce a coupling of neighboring clusters in such a way that overlaps which are not permitted by the perfect rules are energetically penalized. We expect such a coupling to be weak, because these kinds of defects can be detected only at larger scales. However, at low enough temperature it might still be able to order the supertile random tiling ground state of the relaxed cluster covering to a perfectly ordered structure. This suggests a scenario with two energy scales: the presence of each vertex cluster lowers the cohesion energy by a large amount, so that structures with maximal cluster denstity are strongly favored, even at relatively elevated temperature. The equilibrium structures at these temperatures are therefore relaxed cluster coverings. Additionally, there is a small coupling between neighboring clusters, which at low temperatures can order the supertile random tiling to a perfectly ordered tiling.

We have verified the feasibility of this model by Monte Carlo simulations. Our model considers only the subensemble of states with maximal cluster density. In other words, we simulate at the level of the supertile tiling. The cluster on each vertex is represented by an arrow, like in Fig. 8. This setup keeps the number of clusters constant, so that we cannot leave the states of maximal cluster density, which simplifies the simulation considerably. As flip move we can still use the one of Fig. 4, except that here we have to adjust the cluster orientations of the neighboring vertices of the flip to new values consistent with the new tile configuration. Additionally, we have to introduce a new type of flip, which only changes the cluster orientations on the obtuse corners of a rhombus, but keeps the tiling fixed.

With this model, we have verified that the coupling of the clusters can indeed order the model to a perfectly quasiperiodic structure. In other words, the ground state is a perfect quasicrystal, whereas the high temperature state is the supertile random tiling with the additional degeneracy of the cluster orientations on the obtuse corners of the rhombi, corresponding to the relaxed cluster coverings. In this respect, high temperature means high compared to the cluster coupling, but still low compared to the energy required to break up clusters.

Also in this model it is possible to measure the entropy density of the relaxed covering ensemble. In this case, the ground state is ordered and has zero entropy (at least in the thermodynamic limit), and the high temperature state is the one whose entropy we are interested in. We therefore need only measure the difference between the entropies of the high temperature state and the ground state, and extrapolate to the thermodynamic limit. It turns out, however, that finite size scaling for the present model does not work as well as for the model with a random tiling ground state. The results are therefore less precise, although they are consistent with the results reported in Section 4.We therefore won't present any

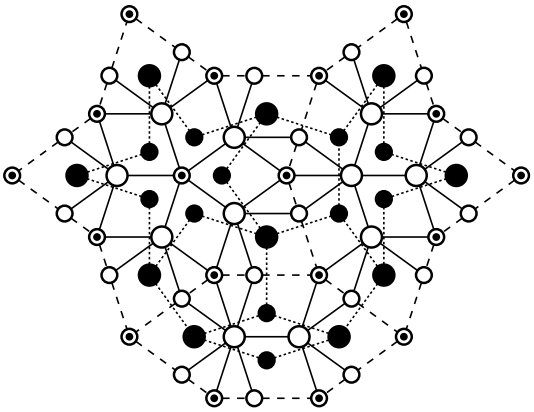

Fig. 9. Atomic cluster found by Henley and Roth [14] in a molecular dynamics simulation. In units of the period in z-direction, white atoms are at $z = 0$, black atoms at $z = \frac{1}{2}$, and dotted atoms at $z = \frac{1}{4}, \frac{3}{4}$.

details here.

6. Discussion and conclusion

After a short review of different ordering principles for quasicrystals, which all make use of cluster covering or cluster density maximization, we have presented several relaxed versions of these ordering principles, which produce as ground state a supertile random tiling structure. Such structures might therefore also be called random covering structures. Models of this kind can be very suitable for the explanation of experimentally observed quasicrystals, as the latter are often not perfectly quasiperiodic, but more of a random tiling nature. The relationship between the different random covering ensembles, and their relation to known random tiling ensembles, has precisely been established.

One of the relaxed overlap rules has a very natural realization in terms of a vertex cluster in the random PPT. Such PPT, and the closely related HBS tilings, are often observed in high rsolution electron microscopy images. The most striking resemblence, however, is with an atomic cluster found by Roth and Henley [14] in a molecular dynamics simulation of decagonal Frank-Kasper type quasicrystals. This cluster is shown in Fig. 9. The only discrepancy with our vertex cluster is at the center of a star, where we would have expected a single atom at $z = 0$, not two atoms at $z = \frac{1}{4}, \frac{3}{4}$ (Fig. 9).

The feasibility of all our models has been checked by Monte Carlo simulations. In particular, we have measured the entropy density of the random covering ensemble, and we have verified that the relaxed cluster coverings coincide with the states of maximal cluster density. Moreover, a coupling between neighboring clusters can order the random tiling type ground states to a perfectly ordered structure.

All our simulations have been in two dimensions. It is well known that for finite range interactions, quasicrystal structures in two dimensions cannot be stable at any positive temperature [15]. At positive temperature, they are always in a random tiling state, the phase transition from the ordered state to the random tiling state being at zero temperature. One could consider a periodic stacking of our two-dimensional models, however, with a suitable coupling between neighboring layers. Such layered *tiling* models have already been considered by Jeong and Steinhardt [16], but should be possible also in the cluster covering setting. It is expected that for such three-dimensional models, the phase transition from ordered to random tiling type structures will be at positive temperature. Preliminary results in this direction look promising.

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