Quasicrystalline Ground States Without Matching Rules

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Abstract

A simple, local cluster interaction is presented, which has as (only) ground states perfectly quasicrystalline tilings from a single local isomorphism class. Since these tilings do *not* allow for any perfect matching rules, it is thereby shown that the class of structures which are the ground state of some finite range interaction is considerably larger than previously anticipated. Cluster interactions having a quasicrystalline ground state turn out to be simple and robust, and therefore provide an attractive explanation for the existence of quasicrystals. A simplified version of our cluster interaction is found to have super-tile random tiling ground states. Due to the big size of the super-tiles, these random tilings still look very perfect on a local scale.

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I. INTRODUCTION

It is generally believed that the existence of local matching rules for a quasiperiodic tiling, and the existence of finite range interactions having as ground state a quasicrystalline structure locally decorating that tiling, are closely related to each other. More precisely, it can be argued[1, 2] that whenever a quasicrystalline structure and a quasiperiodic tiling with *perfect matching rules*[3] are *locally derivable*[4] from each other, there exist finite range interactions having this structure as their ground state. Perfect matching rules enforce tilings (or other discrete structures) from a single local isomorphism (LI) class, consisting, roughly speaking, of tilings locally indistinguishable from each other. Perfect matching rules thus are the strongest possible matching rules. This strong requirement of perfect matching rules was not considered a problem, since in most cases one is anyway interested in having interactions whose set of ground states consists of structures from a single LI class.

There are, however, interesting structures whose LI class does not allow for any local, perfect matching rules. Notable examples are certain eight- or twelve-fold symmetric quasiperiodic tilings[2]. In these latter cases the problem is usually "solved" by adding further, non-local information to the tiling, in the form of a (non-local) decoration[5]. However, this cannot really be considered a solution of the problem, since the structure with decoration certainly is not the same structure any more – the decorated structure is not even in the same local derivability class. The stabilization of the undecorated structure by finite range interactions therefore still remains as an open problem.

Some of the tilings without perfect matching rules still allow for so-called *strong matching rules*[3], which enforce perfectly ordered tilings, but allow for tilings from more than one LI class, usually from a whole 1-parameter family of LI classes. An important observation now is that structures from different LI classes, for generic interactions at least, will always have different energies. Therefore, if we have any interaction which favours a set of strong, but non-perfect matching rules, it will, of course, penalize tilings not satisfying the matching rules by giving them a high energy, but it will also differentiate between the different LI classes allowed by the matching rules, by giving them different energies. The problem then is to find out which LI class among those allowed by the matching rules has the lowest energy, and thus is the ground state of the interaction. Conversely, if a specific LI class is given, chosen among those allowed by the matching rules, the question is whether this LI class is the ground state of a suitable local interaction, which should preferably be simple and of short range. The latter seems possible if the LI class in question is somehow uniquely distinguished among the other ones allowed by the matching rules. For instance, it might have a higher symmetry than all the other LI classes. For a generic choice of an LI class, however, the existence of such an interaction is questionable. Still, we can conclude that under certain additional conditions it should be possible to build local interactions having a ground state which does not admit perfect matching rules.

In the following, these ideas will be illustrated by explicitly working out a particular example, the undecorated octagonal Ammann-Beenker tiling[6, 7]. In Section II a simple cluster interaction will be presented, which has as its only ground states perfectly quasiperiodic octagonal tilings. In Section III it is shown that a simplified version of this interaction, which is not able to stabilize the perfect octagonal tiling, still leads to super-tile random tiling ground states. Finally, in Section IV we summarize our results and discuss the prospects of applying these concepts to other systems.

II. A CLUSTER INTERACTION STABILIZING THE UNDECORATED OCTAGONAL TILING

It is well known that octagonal Ammann-Beenker tiling [6, 7], which is composed of squares and 45° rhombi, does not allow for perfect matching rules. This can be seen as follows. Any square-rhombus tiling which is consistently arrowable by Beenker arrows[7] on the edges can uniquely (and locally) be deflated an arbitrary number of times. This is the case, in particular, for the (periodic) tiling consisting of squares only. Starting with this square tiling, after two deflations we arrive at a tiling, all of whose vertex neighborhoods are allowed vertex neighborhoods from the perfect octagonal tiling. At the scale of vertex neighborhoods, this tiling thus cannot be distinguished from a perfect octagonal tiling, and with each further deflation, the scale at which the tiling is indistinguishable from the perfect octagonal tiling is increased by another factor $\sigma = 1 + \sqrt{2}$. Since all these tilings are periodic, one can find among them a counterexample to any attempt to characterize the octagonal tiling by an atlas[8] of allowed neighborhoods of maximal size R, which proves that the undecorated octagonal tiling does not admit perfect matching rules.

The octagonal tiling does support strong matching rules, however. Such strong matching rules are given by the *alternation condition*[9], which had originally been introduced as so-called weak matching rules for general 2D rhombus tilings, but failed to work in the octagonal case, because it is not able to exclude periodic approximants. The alternation condition requires that along any lane of tiles the rhombi have to point to alternating sides of the lane. An example of such a lane of tiles is shown in Fig. 1. It has been shown[10] that the alternation condition for the square-rhombus tiling, which is equivalent to the matching rules imposed by the Beenker arrows[7], enforces tilings which are perfectly quasiperiodic and (at least) four-fold symmetric, or which are periodic approximants[17] to such tilings, with square unit cell. The tilings compatible with the alternation condition consist of all cut- and projection tilings whose cut space is rotated with respect to the octagonal cut space by a Schur rotation[11] maintaining one of the two D_4 subgroups of the octagonal D_8 symmetry group (see also [12]). The space onto which the tiling is projected is always the same, in order to maintain the shape of the tiles. The D_4 symmetry group that is preserved contains those mirror lines which contain the tile edges. Maintaining the other mirror lines, which are contained in the other D_4 subgroup, does not lead to tilings satisfying the alternation condition. Within the family of tilings allowed by the alternation condition, there is a single LI class with full octagonal symmetry. All other tilings have only four-fold symmetry, some of which are even periodic.

Our strategy now is to choose a simple interaction which strongly favours the alternation condition, and at the same time prefers the perfect octagonal tiling against all other tilings allowed by the alternation condition. Since in the octagonal tiling there are never more than two squares between two consecutive rhombi pointing to opposite sides of a lane, we shall actually choose an interaction which favours structures having this more restrictive property too. To construct this interaction, we first have to take a closer look at the structure of the perfect octagonal tiling. It is easily verified that if between two consecutive rhombi there is no square in the same lane, then the two rhombi are part of a hexagon. Every such hexagon is contained in an octagon cluster, shown in Fig. 2. The same holds true for every pair of consecutive rhombi with one square in between. A pair of consecutive rhombi with two squares in between is always contained in a "ship" cluster also, shown in Fig. 2. Since in the octagonal tiling there are never more than two squares between two rhombi, every instance of the alternation condition being satisfied (i.e., two consecutive rhombi pointing to alternating sides of the lane) is contained in at least one of the two clusters shown in Fig. 2. For this reason, we shall give these two cluster low, negative energy, and only these. All other clusters are given zero energy. It will then be shown that if the ratio $\mu = E_{\rm oct}/E_{\rm ship}$ of these two cluster energies is chosen properly, tilings having minimal energy will satisfy the alternation condition and will be eight-fold symmetric, i.e., they are perfect octagonal tilings.

Interactions which give low energy only to a few clusters most important for the structure, and zero energy to all other clusters, have recently been proposed by Jeong and Steinhardt[13]. Such interactions do not try to exclude unwanted configurations by explicitly giving them high energy. Rather, these interactions minimize the energy of the structure by maximizing the density of the low energy clusters. This is achieved by frequent overlaps of such clusters, which leads to correlations. If the low energy clusters are chosen properly, perfectly ordered structures may emerge as the ground state, even though the interaction does not explicitly penalize bad, defective local configurations. Such interactions appear to be very robust, i.e., they need no excessive fine tuning of parameters in order to work, and they seem relatively easy to realize in terms of atomic pair interactions.

Here we should keep in mind that in complex structures such as quasicrystals we cannot expect that all pairs of neighboring atoms are at their ideal distances, where the corresponding pair potential is minimal. For geometrical reasons there will always be some bonds which are somewhat frustrated, and a compromise between the competing interactions has to be found for the ground state structure. However, there may be some finite clusters for which all interatomic distances fit almost perfectly to the corresponding pair potentials. Such clusters therefore will have a particularly low energy, and it is advantageous to pack them as densely as possible, with large overlaps. In our tiling model, these low energy clusters are represented by low energy tile clusters, which of course should be thought of as being decorated with atoms.

By construction, the interaction we have chosen strongly favours the alternation condition. Still, we have to demonstrate that it also favours the octagonal tiling among all tilings satisfying the alternation condition. For this we need to calculate the densities of the octagon and ship clusters as a function of the Schur rotation angle φ, which we set equal to zero for the octagonal tiling. These densities are easily obtained from the areas of the subregions of the (deformed) acceptance domains corresponding to these two clusters (see[12]). To leading order, both densities vary quadratically as a function of φ, one with a maximum and one with a minimum at $\varphi = 0$. The exact result is:

$$\begin{aligned} d_{\mathrm{oct}}(\varphi) &= \sigma^{-1}(1 - \sigma^2 \tan^2 \varphi) \qquad (|\tan\varphi| \leq \sigma^{-2}) \\ d_{\mathrm{ship}}(\varphi) &= \frac{\sigma^{-2}}{2}(1 + \sigma^4 \tan^2 \varphi) \qquad (|\tan\varphi| \leq \sigma^{-4}) \end{aligned} \tag{1}$$

where again we have set $\sigma = \sqrt{2} + 1$. The range of validity of (1) is determined by the domains of stability of the topology of the corresponding subwindows. It should be noted that these results are exact even for periodic approximants, for which the calculation of the areas of the subwindows does not necessarily lead to the correct result. In this case, however, there is only one possible unit cell content for each approximant[12], for which (1) can be checked explicitly.

From (1) it is now clear that the octagonal tiling has minimal energy among all tilings satisfying the alternation condition, at least among those with moderate phason strain, provided the ratio of cluster energies $\mu \equiv E_{\mathrm{oct}}/E_{\mathrm{ship}}$ satisfies the inequality $2\mu > \sigma$. Would this be an equality, a periodic approximant with square unit cell of edge length σ would be energetically competitive. Note that for that approximant (1b) holds even though its phason strain is outside the domain of validity of (1b). It is also clear that the ratio μ must be bounded from above, too, since with a cluster energy $E_{\mathrm{ship}} = 0$ the structure could be disordered by partially flipping certain worms without any energy cost (see [14]). In order to determine the correct interval of admissible values for the ratio μ of the cluster energies, we have numerically calculated the densities of octagon and ship clusters for a large number of various kinds of periodic approximants to the octagonal tiling, concentrating on approximants which are not permitted by the alternation condition, or which are outside the domain of validity of (1). From these densities we can derive that the octagonal tiling has lowest energy whenever the ratio of the cluster energies satisfies the inequalities

$$1+\sqrt{2} < \frac{2E_{\rm oct}}{E_{\rm ship}} < (1+\sqrt{2})^5 \qquad (2)$$

The structure with highest relative octagon density is found to be a periodic approximant with rhombic unit cell of edge length σ^2, which leads to the upper bound in (2). Therefore, at both borders of the (huge) window in (2) there is a periodic approximant which becomes energetically competitive.

While the above results provide ample evidence that under the condition (2) the octagonal tiling is preferred against periodic approximants, and probably also against non-periodic structures with a linear phason strain, they do not provide any information about other tilings made from the same tiles. In particular, these results cannot exclude that rearrangements of tiles not occurring in perfect approximants could further lower the energy. In order to exclude such possibilities, we have used Monte Carlo simulations, similar to the ones used in [13], to find the real ground state. A large number of periodic approximants of different shapes and sizes of up to a few thousand tiles have been slowly cooled from infinite to zero temperature. In all cases where the inequalities (2) were satisfied, and cooling was slow enough, the correct, perfectly ordered ground state has been found. As this ground state could be recovered from a completely disordered state, this provides strong evidence that the octagonal LI class of tilings indeed is the ground state of our cluster interaction.

III. AN INTERACTION WITH SUPER-TILE RANDOM TILING GROUND STATES

It is easy to see that favouring only one of the two clusters is not enough to obtain an octagonal ground state. As mentioned above, when only octagon clusters are given low energy, the overall ground state is a periodic approximant with rhombic unit cell of edge length σ^2. Still, it is interesting to consider such a system at fixed stoichiometry, i.e., at fixed concentration for each kind of tile, which also fixes an average phason strain for the tiling. An important observation now is that a perfect octagonal tiling can always be composed to a tiling of big squares and rhombi, both with edge length σ^2, which is nothing but its second inflation (Fig. 3). The big square and rhombic super-tiles are all decorated in the same way, and it is easily verified that rearranging these super-tiles does not change the number of octagons present in the structure (see Fig. 4). In fact, any random tiling with these super-tiles, and with the same density of squares and rhombi, will have the same density of octagons as the perfect octagonal tiling, and therefore will be energetically degenerate with it. In particular, this is the case for square-rhombus super-tile random tilings with zero average phason strain. The same reasoning can be applied to tilings with any other given, fixed phason strain. Note, however, that in the case of a periodic approximant there might be no super-tile tiling compatible with that periodicity, so that part of the periodicity has to be given up in the super-tile tiling, although the phason strain of the approximant is maintained. For each given phason strain, there is a whole ensemble of super-tile random tilings which are energetically degenerate.

In order to find the true set of ground states for this system, the Monte Carlo methods used in [13] have been applied also in this case. Our simulations, which we performed for many different periodic approximants compatible with a super-tile tiling, have shown that the square-rhombus super-tile random tilings indeed belong to the ground state, which therefore is heavily degenerate. An example of such a random tiling, obtained in one of the simulations, is shown in Fig. 4. There are, however, other ground state structures as well, which are not square-rhombus super-tile tilings. One of these is shown in Fig. 5. These other ground state structures can be described as super-tile tilings with isosceles triangles and darts as super-tiles. Note that any square-rhombus super-tile tiling can be decomposed into these smaller super-tiles, too: rhombi are divided into two triangles, and squares into two triangles and one dart. As we have found no other ground states, we conclude that the ensemble of ground state configurations at fixed phason strain consists of triangle and dart super-tile random tilings. Other examples of interactions having super-tile random tiling ground states have previously been found by Jeong and Steinhardt[13].

An interesting aspect of these super-tile random tiling ground states is that they look very perfect at a local scale. The reason is that, on the one hand, the decorations of the super-tiles are legal configurations which frequently occur also in the perfect tiling, and on the other hand, due to the big size of the super-tiles, the effective phason stiffness is strongly enhanced[15], compared to a random tiling will small tiles. Such states are therefore hard to distinguish from perfectly ordered states, and might be perfectly acceptable as models for the structure of even very well-ordered quasicrystals.

It is instructive to see what happens to the super-tile random tilings if the ship clusters are again included in the set of low energy clusters. We note that a ship cluster is located on all of the super-tile edges in Fig. 3, and most of the super-tile edges in Fig. 4. More precisely, there is a ship cluster on all those edges where the alternation condition is satisfied for the super-tiles. In other words: the ship clusters hook the square and rhombus super-tiles together in such a way that the alternation condition is satisfied at the super-tile level. For each violation of the alternation condition there is (at least) one edge where the two halfs of the ship cluster on both sides of the edge do not combine to a complete ship cluster. The decoration of the big squares and rhombi makes sure that on parallel edges of a square, ship clusters are oriented alike, whereas on parallel edges of a rhombus they have opposite orientation. The orientation of the ship clusters therefore works in the same way as the Beenker arrows on tile edges. Whenever there is a mismatch of the arrow directions of the two tiles adjacent to an edge, the two half-ships do not combine to a complete ship cluster. In a similar way, the ship clusters also make sure that triangles and darts are arranged in such a way that they can be composed to squares and rhombi. In every triangle-dart configuration where this is not possible, there is a lower than maximal density of ship clusters. A configuration where this happens is shown in Fig. 5. By these mechanisms it becomes thus very transparent how the inclusion of the ship clusters in the interaction can order the super-tile random tilings to perfectly ordered tilings, in the case of zero phason strain even to perfect octagonal tilings.

IV. DISCUSSION AND CONCLUSION

In this paper we have studied in how far simple cluster interactions can stabilize a unique LI class of quasicrystalline ground states, even in cases where this LI class does not allow for perfect matching rules. As we have worked with a pure tiling model, which implies, in particular, that we have completely rigid clusters, we have implicitly assumed that the interactions responsible for the formation of the tiles and the clusters are much stronger than the coupling between the clusters. It seems that this assumption is not completely unreasonable, as it is well known that certain clusters tend to form already in the melt, shortly before solidification, and it is certainly compatible with our other assumption that these clusters have much lower energy than all the other clusters.

Having made this reservation, the following conclusions, which are drawn from our (mostly numerical) results, appear to be relevant for a better understanding of quasicrystal formation:

i) Local isomorphism classes of tilings, or other discrete structures, which do not admit any local matching rules still can be the (complete) set of ground states of very simple, local interactions. In the example presented here, this is possible because there are local matching rules which enforce at least a family of LI classes of tilings which are already *perfectly ordered*. Within such a family of LI classes, however, only those LI classes can be selected by a local interaction, which are somehow distinguished from the other members of the family. In the present case, the LI class of octagonal tilings has a higher symmetry than all the other tilings in the family.

ii) There are very simple, local interactions having a quasicrystalline ground state, and these interactions seem to be *very robust*. No fine-tuning of any parameters was necessary. Our example shows that an interaction having a quasicrystalline ground state does not need to favour *all* allowed clusters up to a given size against *all* forbidden ones, nor does it need to include *all* these clusters in the interaction. It is sufficient to favour just the most important clusters, and disregard all the other ones, whether they allowed or forbidden. By giving a number of important clusters a lower energy than all the other clusters, a *quantitative element* is introduced in the interaction, which is not present in a pure matching rule interaction, where all allowed clusters have the same energy. In this respect, such a cluster interaction is more realistic.

iii) Interactions not capable of enforcing a completely ordered ground state may still have super-tile random tiling ground states. Even though such structures are not perfectly ordered, they still may look very perfect on a local scale. Since such ground states can be obtained with even simpler interactions than perfectly quasiperiodic ground states, they represent attractive models which can describe the structure of even well-ordered quasicrystals in a perfectly acceptable way.

We have obtained these results with the example of the octagonal Ammann-Beenker tiling, but it should be noted that there are other examples which are expected to be completely analogous. In particular, many (undecorated) dodecagonal tilings are suffering from the same deficiencies as the octagonal tiling[2], in that they do not allow for perfect matching rules, although one can expect matching rules to exist which enforce tilings in a family of LI classes with (at least) six-fold symmetry. We expect that also in these cases a simple cluster interaction is capable of stabilizing a single LI class of perfectly dodecagonal tilings. For the dodecagonal tiling introduced by Socolar[16], consisting of hexagons, squares and 30°-rhombi, the analogy seems to extend even to the matching rules. With the methods used in [10] it should be possible to prove that the alternation condition enforces a family of perfectly ordered tilings also in that case.

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[17] the periodicity of some of the "periodic" approximant tilings compatible with the alternation condition may be slightly broken, due some singular worms which are flipped (see [10])

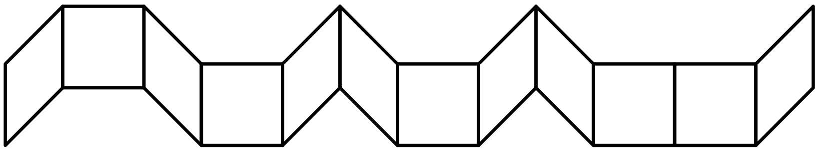

FIG. 1: A lane of tiles satisfying the alternation condition.

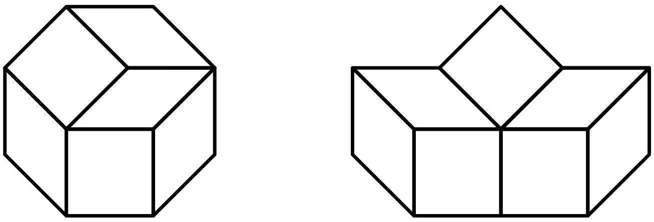

FIG. 2: The octagon cluster (left) and the ship cluster (right).

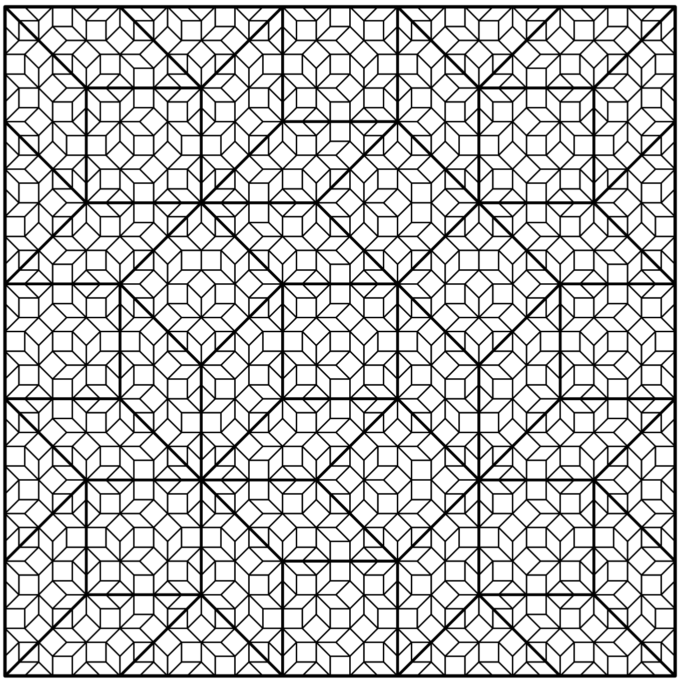

FIG. 3: Part of a perfect octagonal tiling, composed to square and rhombus super-tiles.

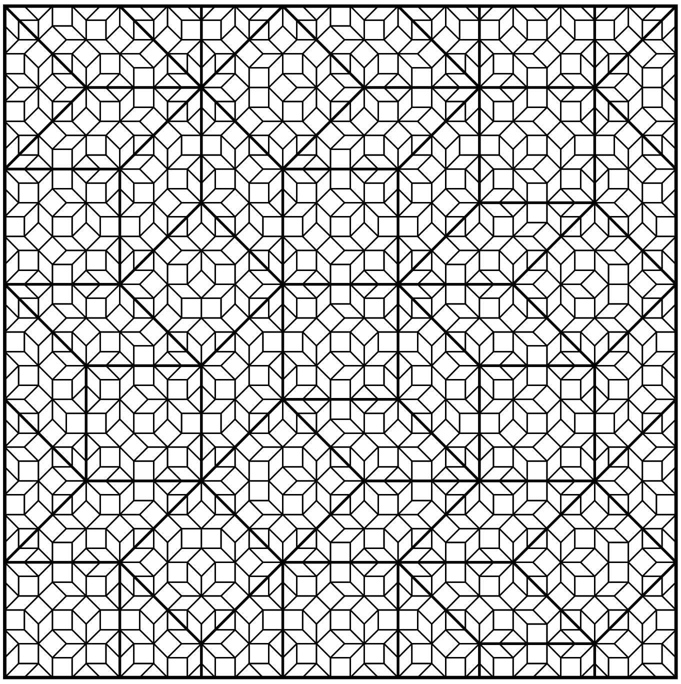

FIG. 4: Square-rhombus super-tile random tiling. Note that the super-tiles do not satisfy the alternation condition everywhere. This configuration was obtained by slow cooling in a Monte Carlo simulation, with an interaction favouring only octagon clusters.

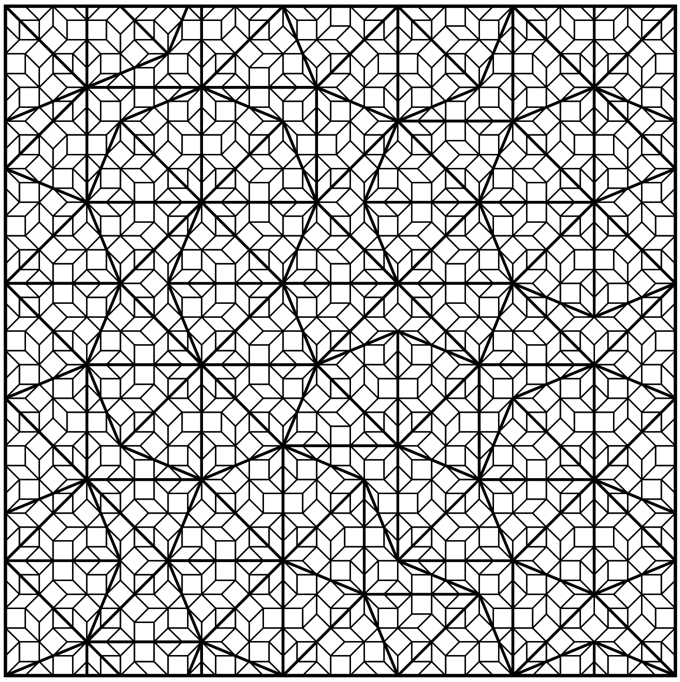

FIG. 5: Triangle-dart super-tile random tiling, obtained in a Monte Carlo simulation under the same conditions as the tiling of Fig. 4. Note that a region near the bottom of the figure cannot be composed to square and rhombus super-tiles.