Simple tiling model and phason kinetics for decagonal Al-Cu-Co

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For decagonal Al-Cu-Co, a tiling model is presented which is based on a deflated version of Burkov's model [S.E. Burkov, Phys. Rev. B **47**, 12 325 (1993)] and which instead of 34 atoms requires at most two atoms per basic tile. The model allows the realization of flips that involve the motion of only two atoms. Two such correlated flips in the deflated tiling mimic such a flip in an inflated tiling with a collective jump of ten atoms forming a ring. Thus for a real quasicrystal the kinetic features of phasons can be represented through a geometrical algorithm. Atomic surfaces and motion of the ten-rings agree with experimental observations. [S0163-1829(96)50426-1]

Even before quasicrystals had been discovered in 1984,[1] structural models were available in the form of aperiodic tilings. Prototype for decagonal quasicrystals was the planar Penrose tiling,[2] consisting of acute and obtuse rhombs. Later, a decagonal partner tiling was established, the Tübingen triangle tiling. Its cells are isosceles golden triangles, an acute one of angle $2\pi/10$, and an obtuse one of angle $6\pi/10$.[3] Tilings allow one to depict a fundamental property of quasicrystalline systems directly, namely the phason distortion. It appears in the form of ''simpleton flips'' of vertices, where a simpleton is the vertex configuration that has the smallest number of neighbors in the Penrose pattern within a hexagonal subpattern, in the triangle tiling within a rhombohedral one. Through consecutive flips it is possible to transform a quasiperiodic tiling into a rational approximant,[4] to perform a random tiling transition,[5] or to transport vertices through arbitrary distances in a mode of self-diffusion.[6–8]

Imposing a tiling structure to a point atom model has been possible in the form of the planar binary tiling, containing two species of atoms.[9] One type of binary tiling arises from the Tübingen triangle tiling, if small atoms are placed to the vertices, and large ones into the acute triangles.[10] Due to steric constraints it is possible to draw tiles within a binary tiling by connecting vertices of differing atoms, even after Monte Carlo flips or molecular dynamics motions have occurred. Binary tilings have been applied to test the entropic stabilization hypothesis for quasicrystals,[12,11] to calculate phasonic elastic constants,[13] or to observe motion of dislocations.[10]

Up to date, however, it has not been possible to arrange the atoms of *real* quasicrystalline compounds into cells of a quasiperiodic tiling in a fully satisfying way. First, the lack of translational symmetry has prevented a reliable structure analysis of most of the over 200 existing quasicrystals, and Per Bak's outcry ''Where are the atoms?'' (Ref. 14) is still being echoed in the literature. The most detailed structure determination has been performed for the decagonal *T* phases Al-Cu-Co and Al-Ni-Co. It is possible to grow these phases from the melt up to centimeter-sized, thermodynamically stable single crystals. They have been studied by high resolution electron microscopy[15] and scanning tunneling microscopy.[16] Steurer[17,18] has evaluated the intensity of 3000 sharp Bragg peaks of Al-Cu-Co, has established an electron density profile by Patterson analysis and the positions for a patch of about 20000 atoms. Accordingly, Al-Cu-Co and the isostructural Al-Ni-Co consist of two plane decagonal quasicrystalline layers, which are alternatively stacked along a perpendicular tenfold screw axis with a stacking period of 4.18 Å .[19,20]

Motivated by Steurer's data Burkov has proposed a tiling model for Al-Cu-Co, first based on a cluster decoration of a Penrose tiling,[21] later on a decoration of the Tübingen triangle tiling.[22] A single tile, however, contains up to 34 atoms. A flip then requires correlated jumps of large clusters in combination with changes of the chemical species. Such processes as addressed, e.g., in Ref. 23 are very unlikely to occur.

Further detailed x-ray investigations of both *T* phases by Steurer and by Frey,[24] however, displayed an appreciable diffuse background and indicated a large amount of disorder. Also, Fettweis *et al.*[25] recently detected transitions from the quasicrystalline state to coherently twinned microcrystalline domains. Thus the structure does not resemble the deterministic model of Burkov by far and it appears necessary to find mechanisms of atomic motion by which disorder can be introduced.

In this article, we start from one of Burkov's

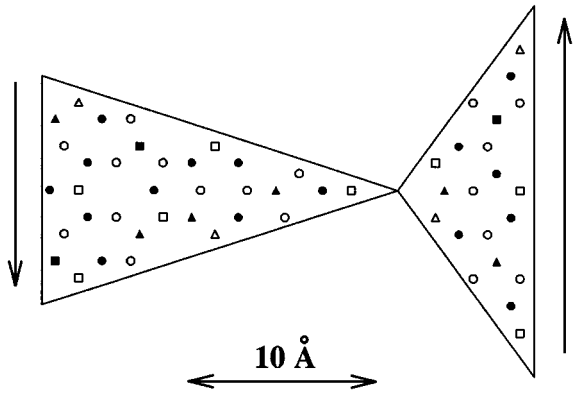

FIG. 1. The two decorated fundamental arrowed golden triangles. Symbols denote: □ Co, △ Cu, and ○ Al. Filled symbols mark layer one, empty symbols mark layer two.

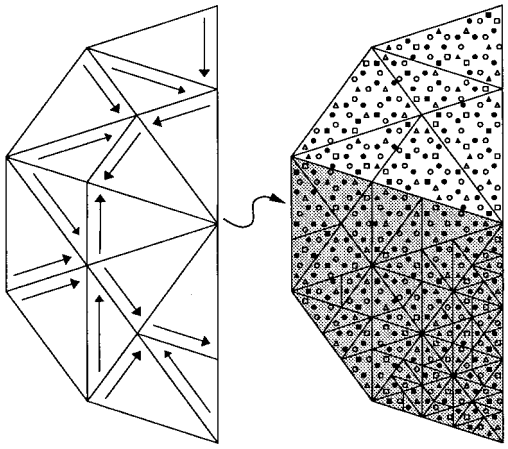

FIG. 2. Left: Patch of a triangle tiling. Right: Decoration of the patch and deflation in three parts (shaded).

decorations,[22] slightly modify it and prove that it is equivalent to a deflated triangle tiling consisting of two basic triangles, each containing at most two atoms. In the deflated tiling, simpleton flips are possible involving jumps of only two atoms by distances of a maximum of 2.75 Å . Two correlated jumps in adjacent layers imitate rigid motions of atomic ten-rings, which are characteristic features of the Al-Cu-Co structure. Thus we have shown that a tiling model and its phason kinetics can describe a real quasicrystal.

The Tübingen triangle tiling can be obtained by the projection method[26] or by a deflation procedure,[27] and it possesses matching rules expressed by arrows.

For Burkov's model two basic triangles must be decorated

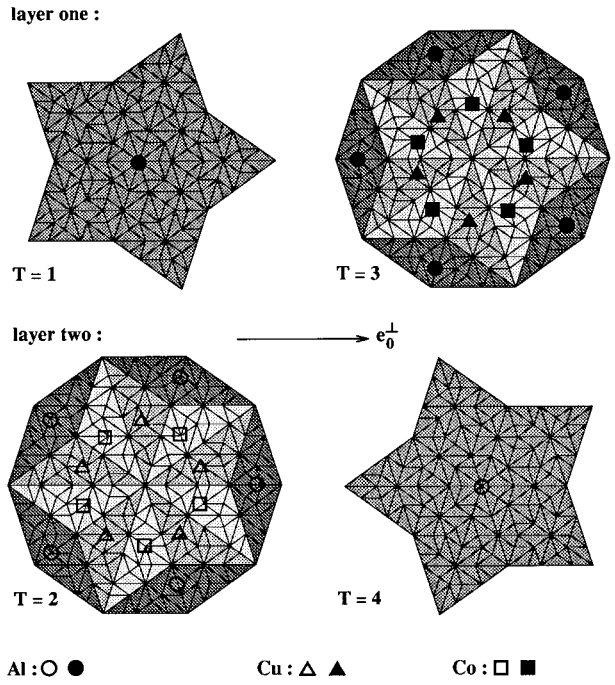

FIG. 3. (a)–(d) Atomic surfaces of the atoms for different translation classes T and the two layers.

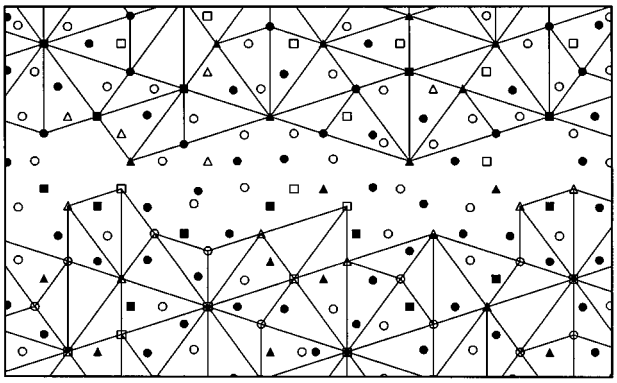

FIG. 4. Patch of a d-Al-Cu-Co quasicrystal. Upper part: Section of the binary tiling for layer one. Lower part: Section of the binary tiling for layer two.

(Fig. 1). The decoration contains atoms for both layers, 34 atoms for the acute and 21 atoms for the obtuse triangle. The rules to transfer the decoration to the other triangles of the tiling are the following: (i) Merely translated copies of one of the fundamental triangles show the translated decoration. (ii) If a triangle is rotated then the decoration is rotated with it, but if the rotating angle is an odd multiple of $2\pi/10$ additionally the layers must be exchanged. (iii) For a triangle resulting from a reflection of a fundamental triangle at the axis of symmetry also the decoration is reflected and then Cu and Co atoms are exchanged. The right side of Fig. 2 shows the decorated arrowed patch of the triangle tiling on the left side.

The peculiar decoration allows to express the coordinates of each atom as an integer linear combination of the vectors of a regular pentagonal star. Hence the atomic coordinates of the superposed two layers can be established by the method of atomic surfaces in five dimensions. The atomic surfaces are depicted in Fig. 3. They are identical to those proposed by Burkov.[22] [The corresponding decoration as calculated by us and displayed in Fig. 1 disagrees, however, with Fig. 2(a) in Burkov's article.[22]]

The atomic coordinate quintuples $\boldsymbol{n} \in \mathbb{Z}^5$ can be arranged into five translation classes $T = \Sigma_{i=1}^{5} n_i \bmod 5$. There are four atomic surfaces: two regular pentagonal stars and two decagons, each labeled by a different translation class $T \in \{1,2,3,4\}$ (Fig. 3). Each layer of the decagonal quasicrystal contains only atoms of two different translation classes (and hence surfaces). Layer one (marked with filled symbols) consists of Cu and Co atoms of translation class $T=3$ and Al atoms of $T=1,3$. Layer two (marked with empty symbols) contains Co and Cu atoms of $T=2$ and Al atoms of $T=2,4$. With the polar calculus[28] the composition of the model quasicrystal becomes 61.8% (τ^{-1}) Al atoms and

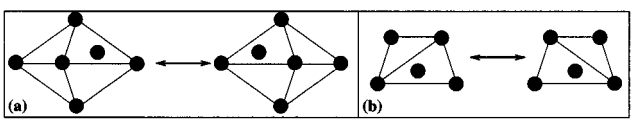

FIG. 5. (a) Simpleton flip. (b) Trapezoid flip.

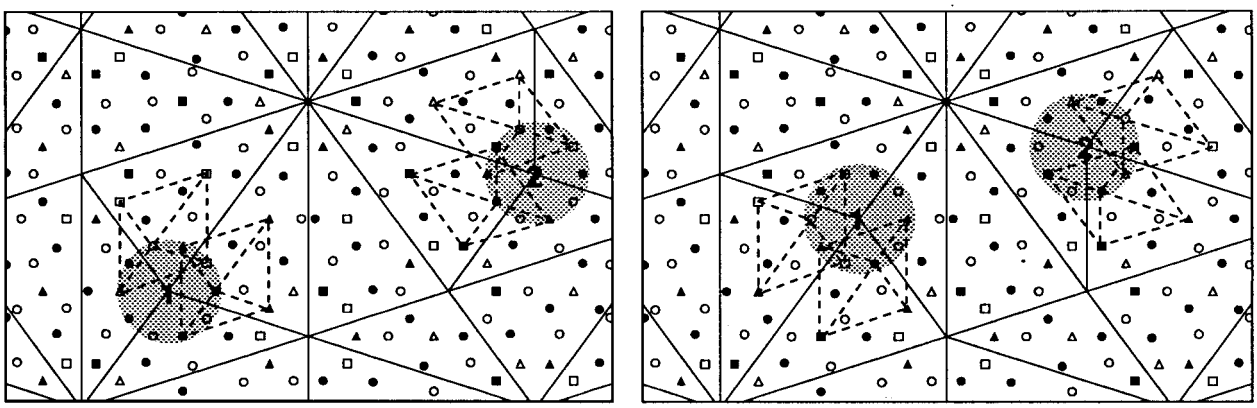

FIG. 6. Tiling of deflation step 1 (full lines) and some tiles of deflation step 3 (dashed lines). Left: Initial state. Right: Two double flips have occurred, each of them involving four atoms within the small tiling. They mimic the motion of entire atomic ten-rings, whose center perform a simpleton flip within the large tiling.

19.1% Co and Cu atoms. There is an amazing consequence of the symmetry of the atomic surfaces: One can deflate the tiling by factors of τ consecutively, obtaining smaller triangles with fewer atoms as shown in Fig. 2. At each deflation step two basic triangles can be chosen, from which the entire tiling is restored by Burkov's decoration rules (i) to (iii). In the first deflation step the vertices are centers of rings, consisting of ten atoms (Fig. 2). These rings are striking features of Al-Cu-Co in transmission electron microscopy exposures.[15]

The triangles of deflation step three (τ^3) are the smallest tiles for the model. There is an acute triangle containing one Al atom and one Cu or Co atom, depending on the arrow of the triangle, and an obtuse triangle hosting one Al atom.

The deflation steps are also visible in the atomic surfaces. As Fig. 3 shows, they are subdivided into a hierarchy of congruent triangles of ratios $1:\tau:\tau^2:\tau^3$. The smallest triangles are atomic surfaces of the undeflated tiling, and their number is that large, because for each atom in the decoration there is a translate of the atomic surface of its host tile. The deflation procedure is possible only due to the simplicity and symmetry of the atomic surfaces.

On the scale of the smallest triangles of the tiling simpleton flips appear now possible, as they require only hopping motions of few atoms along small distances. However, these triangles are arrowed, and the arrows determine whether the triangle contains a Cu or a Co atom. Flips may require an unphysical simultaneous change of the atomic species (''chemical phason'').

Reasonable flips, however, seem to be possible, if one considers the two layers separately and interprets the atomic positions of each of them with the help of a binary tiling.

The atomic surfaces of a binary tiling are a regular decagon [as in Fig. 3(c)] and two regular pentagonal stars [as in Figs. 3(a) and 3(d)].[10] The atoms corresponding to the decagon are placed on the vertices of a triangle tiling. Those belonging to a pentagonal star of type Fig. 3(a) are placed into the circumcenters of those acute triangles, which result from a basic acute triangle by a rotation of an odd multiple of angle $2\pi/10$. The atoms corresponding to the other pentagonal star [of type Fig. 3(d)] fill the centers of the remaining acute triangles.

Thus each of the layers forms a binary tiling with one atomic surface (in the form of a regular pentagonal star) missing. The vertices are occupied by all three types of atoms, half of the acute triangles are filled with Al atoms.

The triangle tilings are illustrated for both layers in Fig. 4. The upper part shows the triangle tiling describing layer one, the lower part the one for layer two.

Within this description we are able to perform phason flips involving only a few atoms. The simplest elementary flips are the simpleton flip and a following trapezoid flip.[29] The simpleton flip causes a jump of two atoms; the trapezoid flip rearranges the bonds and does not move any atom (Fig. 5). Depending on their type the atoms move over distances of 2.75 Å (atoms inside the acute triangle), 1.79 Å (vertex atom), or 0.87 Å (if the inside atom moves to a vertex and vice versa). At the beginning of the randomization process only Al atoms are able to jump in simpleton flips. Subsequent trapezoid flips bring Co and Cu atoms also into flip positions.

Thus we have presented a mechanism for an uncorrelated randomization of the layers on the highest level of deflation. Two correlated basic flips, one in each layer, however, can mimic simpleton flips of entire clusters, namely the characteristic ten rings, in the first level of deflation. We call such a process ''double flip.'' For a description we have to introduce the notion of ''atomic five-ring'' and of a ''regular simpleton flip.'' An atomic five-ring is a vertex configuration which contains a sequence of an obtuse, an acute, and again an obtuse triangle. A regular simpleton flip is one which leads to a vertex configuration with one five-ring more than initially. A double flip consists of two regular simpleton flips, one in each layer, which are separated by less than 6 Å , and which are followed by a trapezoid flip in each layer.

In Fig. 6 the double flip is depicted by dashed lines in the

small scale tiling. Although it requires the motion of four atoms only, it seems as if an entire ring of ten atoms has been displaced. The midpoint of the atomic ten-ring before and after the double flip describes a simpleton flip in a tiling of deflation step one. This larger scaled tiling is drawn in full lines.

The tiling and its kinetics are consistent with experimental results: Within the error bars the calculated shapes of the atomic surfaces agree with those found by Steurer.[30] Steurer also observes the rearrangements of decagonal rings of atoms,[31] which we interpret as double flips. The flips of the centers of these decagonal rings are simpleton flips in the triangle tiling of the first level of deflation.

Now a tiling model of Al-Cu-Co of a most simple decoration is available which in a straightforward geometrical way allows to construct transitions to an approximant phase or to a random tiling and to realize the phason induced self-diffusion.

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