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Ground States of Two-Dimensional Quasicrystals

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Hamiltonians for nonperiodic tilings are considered. It is shown that the quasicrystalline tiling obtained by the cut-and-strip method from a D-dimensional cubic lattice may be a ground state only if the tiling possesses a high orientational symmetry: the $(2, D)$-quasicrystal should have D-fold symmetry if D is even and $2D$-fold symmetry if D is odd. For interactions of a finite range the restrictions are stronger: only a (2, 5)-quasicrystal (Penrose tiling) may be a stable ground state.

KEY WORDS: Quasicrystal; Ground state; Tiling.

Recently a number of two-dimensional quasicrystalline alloys have been discovered. All of them have high planar symmetry. Three types of symmetry have been observed: eightfold,(1) ten fold,(2) and 12-fold orientational symmetry.(3) Nonsymmetrical quasicrystals are not known. HREM data show that these alloys are constructed of elementary cells of two or three types. For example, a decagonal quasicrystal is formed of two right-angle prisms having the two Penrose rhombi as the bases. Since cell arrangements in every layer are identical to each other, the compounds can be considered as crystals in the z direction and 2D quasicrystals in the xy plane. This z periodicity allows one to investigate tilings of the xy plane only. While it had been assumed that quasicrystals are metastable, the discovery of the thermodynamically equilibrium icosahedral phases in the Al–Li–Cu(4) and Al–Cu–Fe(10) alloys raised the question of whether a quasicrystal could be a ground state. The aim of this paper is to answer this question and to explain why nonsymmetrical quasicrystals are not observed.

HREM data are indicative of the fact that the ground-state problem can be decomposed into two independent problems: a decoration problem

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(how to arrange atoms in the cells) and a tiling problem (how to fill the plane by the given tiles of N types). I deal with the tiling problem only. It is useful to consider a tiling as a projection from the D-dimensional space.[5] I will use the following version of the generalized projection method[6]: tiles are parallelograms obtained by the projection of the two-dimensional facets of a unit D-dimensional cube onto a two-dimensional subspace $\mathbb{R}_t^2$ called a tiling plane. The tiling plane is the real physical xy plane. The slopes of $\mathbb{R}_t^2$ with respect to the basic vectors $\mathbf{e}_j$ of $\mathbb{R}^D$ are supposed to be fixed. Thus, there are $D(D-1)/2$ types of tiles. Their sides $\mathbf{e}_j^{(t)}$ are the projections of $\mathbf{e}_j$ onto $\mathbb{R}_t^2$; $\mathbf{e}_j^{(t)}$ will be called tiling vectors. The geometry of the tiles is therefore governed by the slopes of $\mathbb{R}_t^2$. Every tiling can be viewed as a projection of some two-dimensional lattice surface onto the tiling plane $\mathbb{R}_t^2$ (the vertices $\mathbf{X}$ of the lattice surface have integer coordinates: $\mathbf{X} \in \mathbb{Z}^D$; they are projected onto the tile vertices). A tiling associated with a lattice surface confined in the standard strip is called a quasicrystalline tiling. The strip is parallel to the grid plane $\mathbb{R}_g^2$, which does not necessarily coincide with the tiling plane $\mathbb{R}_t^2$. The projections of $\mathbf{e}_j$ onto $\mathbb{R}_g^2$ are called the grid vectors $\mathbf{e}_j^{(g)}$. The cross section of the strip is a projection of the unit cube onto $\mathbb{R}_*^{D-2}$, which is the subspace orthogonal to the grid plane $\mathbb{R}_g^2$.[6] This projection is a convex polytope called a "window." Consider the following model: D and $\mathbb{R}_t^2$ are fixed, tiles are given by the projection and are supposed to be undeformable. There is an interaction between tiles (it may be of finite or infinite range, and a pair interaction or not). The energy of an arbitrary tiling is the total energy of the interaction among all tiles, fluctuations being neglected. The tiling is called a ground state if it provides an absolute energy minimum. The (2, 3)-quasicrystals differ radically from higher dimensional ones. In this paper I deal with the case $D \geqslant 4$ only. The ground-state problem for $D = 3$ is solved in ref. 7.

Necessary conditions for a (2, D)-quasicrystal to be a ground state.

1. A (2, D)-quasicrystal may be a ground state only if the tiling space $\mathbb{R}_t^2$ is such that the tiling vectors $\mathbf{e}_j^{(t)}$ are

$$\mathbf{e}_j^{(t)} = (2/D)^{1/2}(\cos(\pi j/D); \sin(\pi j/D)), \quad j = 1, 2, \dots, D \qquad D \text{ odd}$$

$$\mathbf{e}_j^{(t)} = \begin{cases} a(\cos(\pi j/D); \sin(\pi j/D)), & j = 1, 3, 5, \dots, D-1 \\ b(\cos(\pi j/D); \sin(\pi j/D)), & j = 2, 4, 6, \dots, D \end{cases} \qquad D \text{ even} \tag{1}$$

where $a^2 + b^2 = 4/D$.

So, if D is odd, then the tiling vectors form a D-fold symmetric star, and the tiles are rhombi. The subspace $\mathbb{R}_t^2$ satisfying this condition is unique. If D is even, then there is one real number b/a parametrizing allowed subspaces $\mathbb{R}_t^2$. If $b/a = 1$, the tiling vectors form a symmetric star.

Condition 1 can be derived from the following speculation. The clusters shown in Fig. 1 can be found in a quasicrystalline tiling. Let the cluster in Fig. 1a be called a (+)-cluster, the cluster in Fig. 1b a (−)-cluster. The point shown by the solid circle is the projection onto $\mathbb{R}^2_\prime$ of the vertex $\mathbf{X} \in \mathbb{Z}^D$ of the lattice surface associated with the tiling. What is the projection of $\mathbf{X}$ onto $\mathbb{R}^{D-2}_*$? Exploiting the fact that only three edges (namely those parallel to $\mathbf{e}_1$, $\mathbf{e}_2$, $\mathbf{e}_3$) intersect in the point $\mathbf{X} \in \mathbb{Z}^D$, it is easy

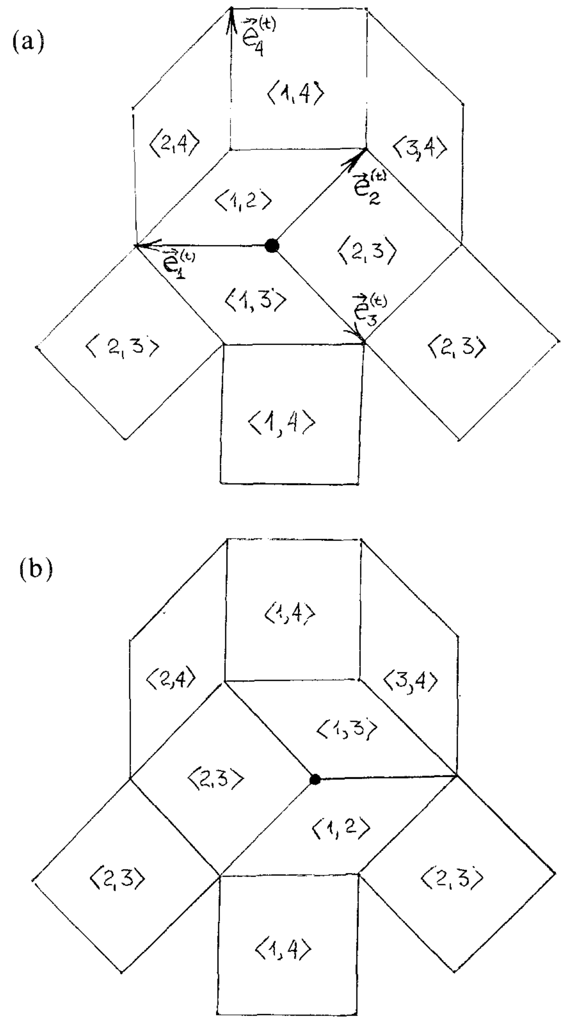

Fig. 1. The (+)- and (−)-clusters differing from one another by rearrangement of the three tiles ⟨1, 2⟩, ⟨1, 3⟩, ⟨2, 3⟩.

to show that $\mathbf{X}$ is located near the strip boundary. Hence $\mathbf{X}^*$, the projection of $\mathbf{X}$ onto $\mathbb{R}_*^{D-2}$, is located near the boundary of the window. The window is a parallelotope in a $(D-2)$-dimensional space; it is bounded by $(D-3)$-dimensional parallelotopes (facets). Each window facet is a projection of a $(D-3)$-dimensional facet of the unit cube onto $\mathbb{R}_*^{D-2}$; they can be labeled by the three integers $i, j, k = 1, 2, \ldots, D$: denote by G_{ijk}^+ and G_{ijk}^- two parallel window facets projected from two parallel $(D-3)$-dimensional facets of the unit cube that are perpendicular to $\mathbf{e}_i$, $\mathbf{e}_j$, $\mathbf{e}_k$. So, the projections of $\mathbf{X}$ in Fig. 1a onto $\mathbb{R}_*^{D-2}$ are situated near the facet G_{123}^+, and the projection of $\mathbf{X}$ in Fig. 1b near G_{123}^-.

If there is no symmetry between different tiling vectors, all the tiles based on different pairs $\langle \mathbf{e}_i^{(t)}, \mathbf{e}_j^{(t)} \rangle$ are physically different. There is no physical reason for the interaction energies between, say, a pair of tiles $\langle 1, 2\rangle$–$\langle 2, 3\rangle$ and $\langle 1, 3\rangle$–$\langle 2, 3\rangle$ to be equal if the tiles $\langle 1, 2\rangle$ and $\langle 1, 3\rangle$ are not identical. Thus, if there is no symmetry, the energies of these two clusters E_+ and E_- are not equal to each other. Let $E_+ > E_-$. Perturb the quasicrystalline tiling in the following manner: take all the $(+)$-clusters and permute three tiles, $\langle 1, 2\rangle$, $\langle 1, 3\rangle$, and $\langle 2, 3\rangle$, in every cluster to obtain the $(-)$-clusters (some authors call such local rearrangement "local phason" but I shall use the Russian term "perestroika"). The new tiling differs from the quasicrystalline one only by an infinite number of perestroikas. Every perestroika changes the coordinates of only one vertex, namely the central vertex in Fig. 1. This change is simply a translation by $\mathbf{e}_4 + \mathbf{e}_5 + \cdots \mathbf{e}_D$, so if the projection of this vertex onto $\mathbb{R}_*^{D-2}$ initially lies inside the window near the facet G_{123}^+, then after perestroika it will lie near the facet G_{123}^-, but outside the window. Bearing in mind that $E_+ > E_-$, we conclude that perestroikas in all $(+)$-clusters yield a tiling that is not associated with any strip and has an energy lower than the energy of the quasicrystalline tiling. If $E_+ > E_-$, then perform antiperestroikas in the $(-)$-clusters. So, the only chance for a quasicrystal to be a ground state is associated with the equality $E_+ = E_-$, i.e., with the symmetry between tiles and tiling vectors. Considering such equalities for all facets $G_{ijk}^{\pm}$, we obtain condition 1.

2. A $(2, D)$-quasicrystal may be a ground state only if the grid vectors $\mathbf{e}_j^{(g)}$ are given by Eq. (1). If D is odd, $\mathbb{R}_g^2$ must coincide with $\mathbb{R}_t^2$; if D is even, they need not coincide, since a parameter $a^{(t)}/b^{(t)}$ in Eq. (1) determining $\mathbf{e}_j^{(t)}$ need not be equal to $a^{(g)}/b^{(g)}$ for $\mathbf{e}_j^{(g)}$.

3. A $(2, D)$-quasicrystal may be a ground state only for those positions of the window in $\mathbb{R}_*^{D-2}$ that generate a tiling with $2D$-fold axis if D is odd and with D-fold axis if D is even.

Condition 3 is not trivial: for example, in the $D = 5$ case the shift of the window along the vector (1, 1, 1, 1, 1) breaks the decagonal symmetry. If a quasicrystal is a ground state, it is degenerate: if the shift of the window (phason shift) in $\mathbb{R}_*^{D-2}$ does not break the symmetry, the energy is not changed. Since not all shifts conserve the symmetry, the number of Goldstone modes is smaller than $D-2$. These phason modes do not affect diffraction patterns or electronic, thermodynamic, and other physical quantities. The question arises: is there any other degeneracy of the ground state except the phason shift? If the answer is negative, then only ideal, nonperturbed quasicrystalline tiling may be a ground state. If the answer is positive, then there is an alternative (below I exploit the one-to-one correspondence between tilings of $\mathbb{R}_t^2$ and lattice surfaces in $\mathbb{R}^D$):

(a) Perturbed surfaces that have the same energy as the ideal surface are confined in some strip parallel to the standard strip.

(b) There exist perturbed surfaces deviating from the strip by any large distance without affecting the energy.

In the former case only short-range noise (ripples on the ideal surface) does not change the energy. In the latter case with long-range perturbations, the strip can be distorted or bent without any change of the energy. This means that the quasicrystalline ground state is thermodynamically unstable (the reader desiring rigorous mathematics may consider case b as a definition of unstable ground states). The problem of an extra degeneracy is connected with Levitov's local rules approach.[8] An approximate qualitative correspondence is:

no extra degeneracy ↔ strong local rules exist

situation a ↔ no strong rules, but weak rules exist

situation b ↔ no weak rules

Combining Levitov's results[8] with conditions 1–3, we obtain:

4. In the case of a finite-range interaction a $(2, D)$-quasicrystal may be a stable ground state only if $D=5$ and conditions 1–3 are fulfilled.

I stress that condition 4 is only a necessary condition, in other words, it only means that if conditions 1–3 are not fulfilled, then the quasicrystalline tiling is not a ground state at all; if 1–3 are fulfilled and $D \neq 5$, then the quasicrystalline tiling may be an unstable ground state. I do not know sufficient conditions for $D=5$. Moreover, the question arises of whether there is any nonperiodic quasicrystalline ground state (either stable or unstable). If $D=3$, the answer is positive.[7] If $D=4$, the answer is also positive (see below). If $D \geqslant 5$, the answer is not known.

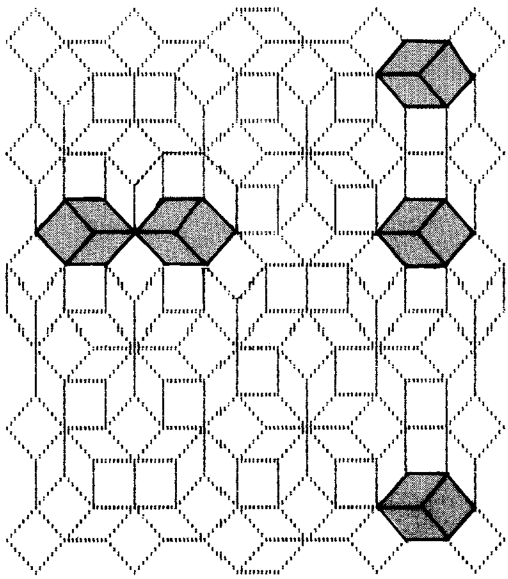

Fig. 2. Quasicrystalline tiling with eightfold symmetry ($D=4$, $a^{(t)}=b^{(t)}$, $a^{(g)}=b^{(g)}$). Clusters analogous to those in Fig. 1 are shaded.

Sufficient conditions for a (2, 4)-quasicrystal to be a ground state. Applying the necessary conditions 1–3 to the $D=4$ case, we see that tiles of three types are possible: a small square [side $b^{(t)}$, Eq. (1)], a large square (side $a^{(t)}$), and a parallelogram (sides $a^{(t)}$ and $b^{(t)}$, 45° angle). In the particular case $a^{(t)}=b^{(t)}$ there are two types of tiles: a square and a rhombus of 45° angle. The tiling itself should possess the fourfold symmetry ($a^{(g)} \neq b^{(g)}$) or the eightfold symmetry ($a^{(g)}=b^{(g)}$). Note that $a^{(t)}$ and $b^{(t)}$ in Eq. (1) determining the tiling vectors need not be equal to $a^{(g)}$ and $b^{(g)}$ determining the grid vectors. The window is an octagon; it is regular if $a^{(g)}=b^{(g)}$. In contrast to the case of larger D, any shift of the octagon in $\mathbb{R}^2_*$ does not break the symmetry. In this section I restrict consideration to the pair interaction between the tiles:

$$E=\frac{1}{2}\sum_{\mathbf{r},\mathbf{r}'} U_{\sigma(\mathbf{r})\sigma(\mathbf{r}')}(\mathbf{r}-\mathbf{r}') \tag{2}$$

the sum is taken over all pairs of tiles, $\mathbf{r}, \mathbf{r}' \in \mathbb{R}^2_t$ are the coordinates of the tile centers in the tiling plane, $\sigma(\mathbf{r})=\langle 1,2\rangle$, $\langle 1,3\rangle$,..., $\langle i,j\rangle$,..., $\langle 3,4\rangle$ label the types of tiles. Equation (2) contains six interatomic potentials $U_{\langle 12\rangle,\langle 34\rangle}(\mathbf{r})$, $U_{\langle 12\rangle,\langle 12\rangle}(\mathbf{r})$, etc. Their number is six instead of 21 due to a high tile symmetry. Consider the ground-state problem for the Hamiltonian (2). We should fix the sides of the tiles by fixing $b^{(t)}/a^{(t)}$ in Eq. (1). Then we should fix the grid space $\mathbb{R}_g^2$ by fixing $b^{(g)}/a^{(g)}$. Consider various two-dimensional lattice surfaces in $\mathbb{R}^4$ having the same average slopes as $\mathbb{R}_g^2$. The tiling obtained by the projection of the lattice surface providing an absolute minimum of the energy (3) is called a ground state at a fixed average slope. We claim that:

5. If some convexity conditions imposed on $U_{\langle ij\rangle,\langle kl\rangle}(\mathbf{r})$ (see below) are fulfilled, then the ground state at fixed $b^{(g)}/a^{(g)}$ is the tiling obtained by the projection of the strip. It is a quasicrystalline tiling if $2^{1/2}b^{(g)}/a^{(g)}$ is irrational, and it is periodic if $2^{1/2}b^{(g)}/a^{(g)}$ is rational. Any shift of the window in $\mathbb{R}_*^2$ does not change the energy, i.e., there are two phason Goldstone modes.

The convexity conditions are rather cumbersome, so I give here only one of them. Introduce a function $\varepsilon_1(\mathbf{X})$: $\mathbb{Z}^4 \to \mathbb{R}^1$,

$$\begin{aligned}
\varepsilon_1(\mathbf{X}) = {} & U_{23,23}(\mathbf{R}+\mathbf{e}_4^{(t)}) - 2U_{23,23}(\mathbf{R}) + U_{23,23}(\mathbf{R}-e_4^{(t)}) \\
& + U_{24,24}(\mathbf{R}+\mathbf{e}_3^{(t)}) - 2U_{24,24}(\mathbf{R}) + U_{24,24}(\mathbf{R}-\mathbf{e}_3^{(t)}) \\
& + U_{34,34}(\mathbf{R}+\mathbf{e}_3^{(t)}) - 2U_{34,34}(\mathbf{R}) + U_{34,34}(\mathbf{R}-\mathbf{e}_2^{(t)}) \\
& + 2[U_{23,24}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_3^{(t)}+\mathbf{e}_4^{(t)})) + U_{23,24}(\mathbf{R}-\tfrac{1}{2}(\mathbf{e}_3^{(t)}+\mathbf{e}_4^{(t)})) \\
& - U_{23,24}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_4^{(t)}-\mathbf{e}_3^{(t)})) - U_{23,24}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_3^{(t)}-\mathbf{e}_4^{(t)}))] \\
& + 2[U_{23,34}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_2^{(t)}+\mathbf{e}_4^{(t)})) + U_{23,34}(\mathbf{R}-\tfrac{1}{2}(\mathbf{e}_2^{(t)}+\mathbf{e}_4^{(t)})) \\
& - U_{23,34}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_4^{(t)}-\mathbf{e}_2^{(t)})) - U_{23,34}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_2^{(t)}-\mathbf{e}_4^{(t)}))] \\
& + 2[U_{24,34}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_2^{(t)}+\mathbf{e}_3^{(t)})) + U_{24,34}(\mathbf{R}-\tfrac{1}{2}(\mathbf{e}_2^{(t)}+\mathbf{e}_3^{(t)})) \\
& - U_{24,34}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_3^{(t)}-\mathbf{e}_2^{(t)})) - U_{24,34}(\mathbf{R}+\tfrac{1}{2}(\mathbf{e}_2^{(t)}-\mathbf{e}_3^{(t)}))]
\end{aligned}$$

$$\mathbf{X} = (X_1, X_2, X_3, X_4) \leftrightarrow \mathbf{R} = X_1\mathbf{e}_1^{(t)} + X_2\mathbf{e}_2^{(t)} + X_3\mathbf{e}_3^{(t)} + X_4\mathbf{e}_4^{(t)} \quad (3)$$

Some potentials in Eq. (3) are equal to each other (details depend on whether $a^{(t)} = b^{(t)}$ or not). There are a number of functions ε_j analogous to (3). The convexity conditions mentioned above are: all such functions $\varepsilon_j(\mathbf{X})$ must be strictly positive for all $\mathbf{X} \in \mathbb{Z}^4$ such that X_1, X_2, X_3, X_4 are positive.

It is interesting to investigate perturbed states. The deviation of the energy from its minimal value can be expanded in series in powers of $\nabla\varphi$, where $\boldsymbol{\varphi} \in \mathbb{R}^2$ is the window shift and the gradient is taken over the coordinates in the tiling space $\mathbb{R}_t^2$. Suppose that $\nabla\varphi$ is small, i.e., in every large region of $\mathbb{R}_t^2$ the tiling is quasicrystalline obtained by the projection from strips whose slopes differ slightly from those of $\mathbb{R}^2_g$. Along with regular terms $(\nabla\varphi)^2$, $(\nabla\varphi)^3$, etc., the expansion contains the $|\nabla\varphi|$ term:

$$
\begin{aligned}
E - E_{\min} &= \frac{S}{2} \sum_{j=1}^{4} \frac{\theta_j \langle \varepsilon_j \rangle}{n_j} \int \left| \sum_{\alpha, a=1}^{2} A_j^\alpha n_j^a \frac{\partial \varphi_a}{\partial x_\alpha} \right| d^2x \\
\mathbf{A}_j &= 2^{1/2} \mathbf{e}_j^{(g)} \\
\langle \varepsilon_j \rangle &= \sum_{n=1}^{\infty} n(\{n\sqrt{2}\}\, \varepsilon_j([n\mathbf{A}_j] + \delta_{jk} \mathbf{e}_k^{(t)}) \\
&\quad - (1 - \{n\sqrt{2}\})\, \varepsilon_j([n\mathbf{A}_j])) \qquad (4)
\end{aligned}
$$

where $[x]$ is the integer part; $\{x\} = x - [x]$; θ_j are constants describing the change of the area S of the tiling plane when projected onto the surface in the four-dimensional space; $\mathbf{n} \in \mathbb{R}^2$ are four perpendiculars to the sides of the window ($\mathbf{n}_1 = \mathbf{e}_4^*$, $\mathbf{n}_2 = \mathbf{e}_3^*$, $\mathbf{n}_3 = \mathbf{e}_2^*$, $\mathbf{n}_4 = \mathbf{e}_1^*$); the function $\varepsilon_1(\mathbf{X})$ is given in (4); ε_2, ε_3, and ε_4 are written analogously. The subscript $j = 1,..., 4$ is associated with $\mathbb{Z}^4$; $\alpha = 1, 2$ with $\mathbb{R}^2_t$; and $a = 1, 2$ with $\mathbb{R}^2_g$. Looking at Eq. (4), would be easy to believe that any strip bending described by nonzero $\nabla\varphi$ yields nonzero energy. But this is not the case: there exists a family of fields $\boldsymbol{\varphi}(\mathbf{r})$ such that the integrand in Eq. (4) vanishes identically. This means that the strip can be bent without any energy change. So, situation b holds. This fact is connected with the absence of weak local rules for the particular tiling with the octagonal symmetry.[8,9]

I have shown that the high symmetry of the quasicrystals is vital for their formation. I do not believe that an interaction of infinite range (RKKI, van der Waals, etc.) plays an important role in real alloys. But it is easy to believe that there is actually an interaction between the third or the fifth neighbors. If this is the case, the experimentally observed quasicrystals with the 8-, 10-, and 12-fold symmetry are not ideal quasicrystals (in the sense mentioned above), but are associated with slowly bent strips. Sufficiently sharp peaks are observed only due to small $\nabla\varphi$ and finite sizes of the samples. Nevertheless, we suppose that necessary conditions 1–3 must be fulfilled, so nonsymmetric 2D quasicrystals cannot exist.

It should be mentioned that all the above results have been derived for a tiling model. The main assumptions are: there are no fluctuations, no elasticity; the decoration and tiling problems are independent of each other; the interaction has no accidental symmetry (i.e., tiles of different types interact with different energies); and the interaction decreases rapidly when the distance between tiles increases.

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