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Structure Model for Icosahedral Quasicrystal Based on Ammann Tiling

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We derive a structure model for icosahedral quasicrystals. The model is based on a statistical approach involving the concept of average unit cell. This approach enables limiting calculations to real space as opposed to higher-dimensional analysis involving to unphysical atomic surface modeling. We start with the three-dimensional Ammann tiling with its two rhombohedral prototiles. For monoatomic decoration of the lattice nodes the perfect agreement with the higher-dimensional description was recently shown. In this paper we discuss the shape of the average unit cell and the first attempts for decoration scheme.

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1. Introduction

Icosahedral quasicrystals (i-QCs) are three--dimensional aperiodic structures with fivefold symmetry in their diffraction pattern. They are the most frequent quasiperiodic structures occurring in ternary and binary systems [1]. However, there are only few refined i-QCs structures [2–4]. Current models for structure solutions of i-QCs are based on cluster analysis (either three-dimensional physical or in higher-dimensional space).

The method used in this work is based on a statistical approach applying the concept of average unit cell (AUC) [5]. The AUC is a statistical distribution of projections of considered quasicrystalline lattice nodes onto a periodic reference grid. The distribution is uniform and dense and follows a scaling rule. Since quasicrystals are two-scale structures, two reference grids with the ratio of lattice constants given by τ need to be introduced. Thus two coordinates u and v are obtained by a projection and the distribution $P(u,v)$ fully defines the AUC. It is known [5–8] that for a quasicrystal the AUC is non--zero only along a line segment with a τ^2 coefficient in (u,v)-plot. This characteristic is called TAU2-scaling [6].

The main advantage of the statistical method is that it suffices to consider 3D physical space. It was also shown that the shape of the AUC is directly related to the shape of the atomic surface in the higher-dimensional description [9]. The statistical approach has already been successfully applied to decagonal Al–Ni–Co and Al–Cu–TM (TM = Co, Ir, Rh) phases [10, 11]. The starting structural model for i-QCs within the statistical approach is the 3D Ammann tiling (AT), also called Ammann–Kramer–Neri tiling, which is just the generalization of the rhombic Penrose tiling to 3D [12]. The structural units of AT are the golden prolate and oblate rhombohedra; their volumes are in the ratio $\tau \approx 1.618$, the golden mean. The first application of the statistical approach to i-QCs was shown in [13, 14].

2. Average unit cell of the 3D Ammann tiling

The shape of the AUC for AT is the Keplerian triacontahedron. Its faces are golden rhombuses (Fig. 1, left). This solid exhibits icosahedral symmetry; in particular, it has six fivefold axes. The analytical result shows that the AUC is linearly related to the atomic surface by $u_i = -r_i^{\perp}/\tau$ ($r_i^{\perp} = x^{\perp}, y^{\perp}, z^{\perp}$; $i = x, y, z$), where $r_i^{\perp}$ is the perpendicular space coordinate while u_i is the corresponding parallel (physical) space coordinate within the AUC. This is only true for a proper choice of the reciprocal space basis used for construction of the AUC, provided that the shape of the AUC is the Keplerian triacontahedron (see in the sequel). The relation between the AUC and the atomic surface is shown in Fig. 1 (right).

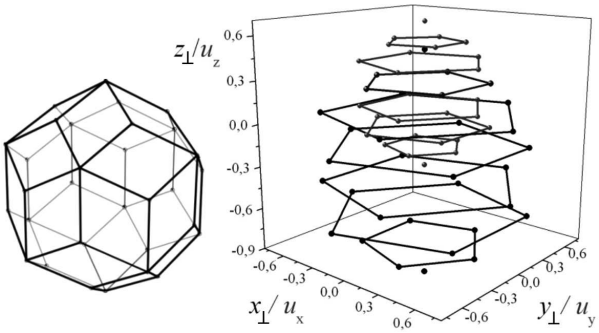

Fig. 1. (left) Keplerian triacontahedron. (right) Atomic surface (black, bigger) and AUC shape (gray, smaller). The coordinates of the AUC nodes are τ times smaller than those of the atomic surface.

The AUC is numerically obtained as a projection of the real structure positions (for a model structure like AT the positions of nodes in AT) on the reference lattice. The statistical distribution of these projections $P(u_i)$ is called AUC. The reference lattice is a square grid with lattice constants related to the reciprocal space vectors $\boldsymbol{k}$ used for indexing the diffraction peaks. In order to

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completely describe a quasicrystal one must introduce modulation vectors ($\boldsymbol{q} = \boldsymbol{k}/\tau$) as well as a second reference grid with a τ times larger lattice parameter. Projections onto both grids simultaneously give a 6D distribution $P(u_i, v_i)$, where v_i are the AUC coordinates with respect to the reference lattice related to the modulation vector $\boldsymbol{q}$. The complete distribution $P(u_i, v_i)$ is again non-zero only along lines $v_i = -\tau^2 u_i$, which simplifies the description.

Here we show some shapes of the $P(u_i)$ distribution for two sets of wave vectors used to construct a reference lattice. We prove that they depend on the choice of these vectors. For the shapes in Fig. 2 the reciprocal space vector basis $\boldsymbol{d}_i^{*\parallel}$ was chosen (see Fig. 3, left, for the definition of the basis vectors).

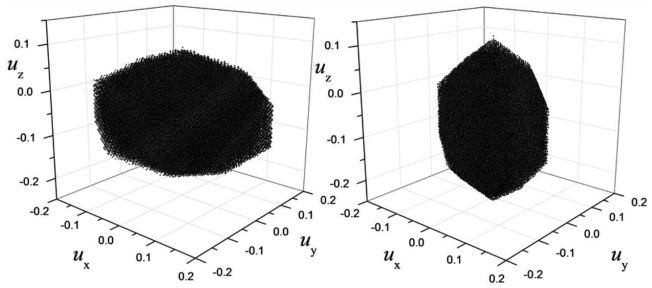

Fig. 2. Dependence of the AUC shape on the choice of basis vectors. The following basis vectors were chosen (Miller indices in brackets, setting $\boldsymbol{d}_i^{*\parallel}$ as a reciprocal space basis was chosen): (left figure) $|\boldsymbol{k_x}| = 13.7\ (0,-5,-5,2,6,2)$,$|\boldsymbol{k_y}| = 7.2\ (0,2,-2,-3,0,3)$,$|\boldsymbol{k_z}| = 17.9\ (9,4,4,4,4,4)$, (right figure) $|\boldsymbol{k_x}| = 22.2\ (0,-8,-8,3,10,3)$, $|\boldsymbol{k_y}| = 11.7\ (0,-3,3,5,0,-5)$, $|\boldsymbol{k_z}| = 4.2\ (2,1,1,1,1,1)$.

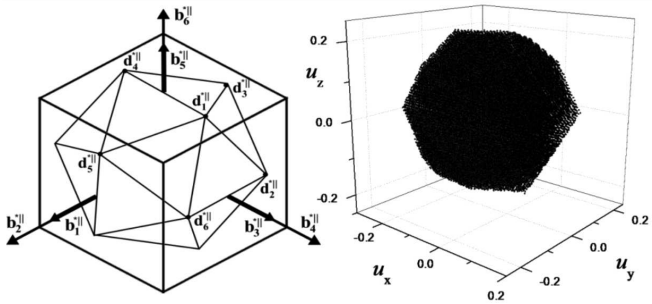

Fig. 3. (left) The two reciprocal space bases. (right) AUC shape with respect to Cartesian basis vectors.

After changing the reciprocal space basis to Cartesian (basis vectors $\boldsymbol{b}_i^{*\parallel}$ in Fig. 3, left) the shape of the AUC becomes uniformly triacontahedral (Fig. 3, right). In this case the peak position can be described as follows:

$$\boldsymbol{k} = h\boldsymbol{q}_{0\boldsymbol{x}} + h'\boldsymbol{k}_{0\boldsymbol{x}} + k\boldsymbol{q}_{0\boldsymbol{y}} + k'\boldsymbol{k}_{0\boldsymbol{y}} + l\boldsymbol{q}_{0\boldsymbol{z}} + l'\boldsymbol{k}_{0\boldsymbol{z}},$$

where

$$\boldsymbol{q}_{0\boldsymbol{x}} = c[1,0,0], \quad \boldsymbol{q}_{0\boldsymbol{y}} = c[0,1,0], \quad \boldsymbol{q}_{0\boldsymbol{z}} = c[0,0,1],$$
$$\boldsymbol{k}_{0\boldsymbol{x}} = c[\tau,0,0], \quad \boldsymbol{k}_{0\boldsymbol{y}} = c[0,\tau,0], \quad \boldsymbol{k}_{0\boldsymbol{z}} = c[0,0,\tau]$$

are the vectors taken along the Cartesian basis vectors $\boldsymbol{b}_i^{*\parallel}$ (Fig. 3, left) and h, h', k, k', l, l' are integer indices. The numerical values: $c = \frac{1}{\sqrt{\tau+2}} \approx 0.5257$, $\tau c = \frac{\tau}{\sqrt{\tau+2}} \approx 0.8507$.

3. Structure factor

The structure factor for AT can be obtained as the Fourier transform of the distribution $P(u_i)$ integrated over AUC. Analytically, it is just the volume integral over the Keplerian triacontahedron. Numerically it is the integral over the distribution of points (i.e. the shapes in Fig. 2 and 3, right). The TAU2-scaling reduces the Fourier transform to a 3D integral, since the full distribution $P(u_i, v_i)$ is non-zero only for TAU2-scaling property.

The structure factor for AT is given by the following formula:

$$F(\boldsymbol{k}) = f_{at} \iiint_{\mathrm{AUC}} P(u_x, u_y, u_z) \exp(\mathrm{i}\boldsymbol{\chi}\cdot\boldsymbol{u})\, \mathrm{d}^3\boldsymbol{u}$$

where

$$\boldsymbol{\chi} = [\chi_x, \chi_y, \chi_z] = [k_x(h'-\tau h), k_y(k'-\tau k), k_z(l'-\tau l)],$$
$$\boldsymbol{u} = [u_x, u_y, u_z] = \left[-x^{\perp}/\tau, -y^{\perp}/\tau, -z^{\perp}/\tau\right]$$

and f_{at} is the atomic form factor, considered equal to 1 for non-decorated AT.

4. Diffraction pattern

The peak intensities for the undecorated Ammann model structure are calculated as the square of the structure factor modulus. For "empty" AT the structure factor was already derived [11, 12] and the agreement of AUC and higher-dimensional description was proven (Fig. 4).

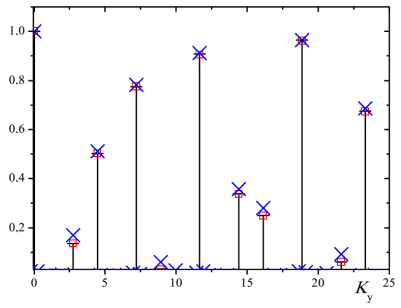

Fig. 4. Diffraction pattern for undecorated AT calculated along K_y-component of the wave vector: dashes — numerical calculations for model structure (Fourier transform of AT points in real space), squares — analytical volume integration over Keplerian triacontahedron, crosses — numerical integration over distribution within AUC.

5. Structural units and decorations

The model based on AT uses only two structural units in real space — prolate and oblate rhombohedra (Fig. 5, left). Our goal is to find the distribution of units with a certain orientation in the AUC and to decorate them with atoms of different kinds. The regions cut out of the AUC

correspond to a given orientation and decoration of the rhombohedra in real space. The distribution of prolate rhombohedra in one basic orientation is shown in Fig. 5 middle and right. This is a picture in perp-space (on the atomic surface). Due to linear relation between atomic surface and AUC, the distribution within AUC is similar. The shape of such distribution is also rhombohedral.

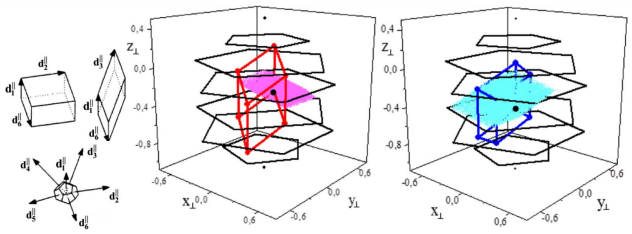

Fig. 5. The distribution in perp-space of oblate (middle) and prolate (right) rhombohedron in basic orientation given by real-space basis vectors (left). The vectors $\boldsymbol{d}_i^{\parallel}$ are related to the reciprocal space vectors $\boldsymbol{d}_i^{*\parallel}$ by an orthogonality condition. The gray area is the part of the distribution corresponding to the marked vertex of a rhombohedron.

The AUC (as well as the atomic surface) can be divided into separate regions related to only two orientations of rhombohedra pairs (prolate and oblate): one pair in the z direction (along some fivefold axis) and one in any other of the fivefold axes. The vertices of rhombohedra of a certain orientation build the distribution. It means that they cut out some part of the AUC. There are altogether $4 \times 8 \times 5 \times 6 = 960$ allowed orientations of such distributions. After applying all icosahedral symmetry operations some overlaps of the distributions are observed. The decoration scheme is to be performed in physical space where the rhombohedra are spanned on neighboring nodes of the Ammann lattice. Yet such rhombohedra are too small to contain any decoration and to fulfill the proper conditions of the refinement procedure.

The decoration scheme can be obtained for instance from the inflation rule. The first three inflation steps for the prolate rhombohedron oriented in the z direction of the physical space are shown in Fig. 6.

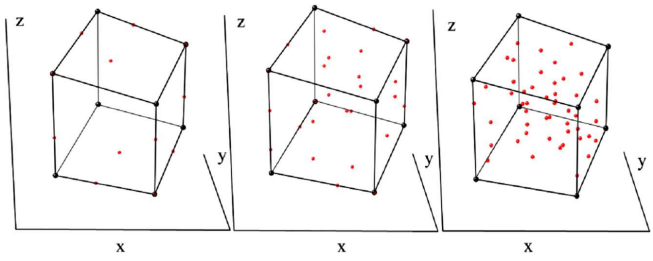

Fig. 6. Prolate rhombohedron in real-space inflated: τ times (left), τ^2 times (middle) and τ^3 times (right). The nodes of AT contained within the inflated rhombohedron are marked.

For the structure refinement it is important to keep the proper ratio between the number of fitting parameters to the number of diffraction peaks. Therefore inflation carried too far maybe of no avail. Inside the τ^3-times inflated rhombohedron there are 27 possible atomic positions which is not sufficient yet for a successful refinement.

6. Summary and outlook

We chose the 3D Ammann tiling with two rhombohedra as structural units as a model of an icosahedral quasicrystal. We derived the structure factor for an empty Ammann lattice using the average unit cell concept that is by the statistical approach. We proved perfect agreement of the AUC results with the commonly used higher-dimensional analysis. We also dealt with atomic decoration and found the possible orientations of rhombohedra and their distributions in the AUC. The inflated rhombohedra indicated the starting atomic positions for the refinement procedure. Currently we study decoration schemes with atoms of different kind for binary and/or ternary systems.

Acknowledgments

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