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PERIODIC SURFACES IN THE DESCRIPTION OF QUASICRYSTALS

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To give a full account of the atomic ordering in modulated crystals or in quasicrystals amounts to find suitable periodic surfaces living in a euclidean space of dimension higher than that of the physical 3D space. In a first part, I summarize the main features of quasiperiodic order and show that it is naturally described by the so-called section or cut method. Next, the problem of short range order will be raised: for quasicrystals, the solution consists in building surfaces in higher D whose intersections with the physical space (imbedded in R^D as a vector subspace) provide the atomic sites. As an example, the "atomic surface" of the octagonal quasiperiodic tiling is analysed in more details in the last section.

Quasicrystals were discovered by Shechtmann (1984) in rapidly quenched samples of metallic alloys [1]. Nowadays, in systems like AlFeCu, diffraction spectra are available presenting sharp Bragg peaks; the (icosahedral or pentagonal) symmetry is fulfilled with a high degree of accuracy and the correlation length is of the same order of magnitude as in conventional crystals (> several \(\mu\)).

The observed icosahedral, 5 or 8-fold symmetry preclude periodic ordering but are compatible with quasiperiodic ordering. General references are proposed in [2]. The homogeneity of the bulk material can also be tested (for ex. by TEM) and rules out twins patterns with overall, say, icosahedral symmetry. Actually polycrystalline phases made of icosahedral twins of cubic or rhombohedral symmetry have been observed in the vicinity of icosahedral quasicrystals [3].

1. Quasiperiodicity: the cut method

A function in the real euclidean space \(E = R^d\) is quasiperiodic if its Fourier transform is a sum of Dirac point measures carried by a finitely generated module. The Dirac peaks correspond to Bragg reflexions in diffraction experiments. A module is the set of all combinations, with integer coefficients, of a finite set of independent vectors called a basis. In the case of the Fourier or Frequency module, the vectors belong to the reciprocal space \(E^*\). For example six independent vertices of a regular icosahedron generate a D=6 dimensional module in \(R^3\).

Such modules can always be considered as linear projections of regular D-dimensional lattices. By inverse Fourier transform the quasiperiodic function is a section by (meaning, here, a restriction to) \(E\) of a function defined in \(R^D\) and D times periodic. Doing so requires to imbed the physical space as an affine subspace \(E+t\) of \(R^D\): the orientation of \(E\) with respect to the lattice of periods \(L\) in \(R^D\) is determined by the various incommensurability ratios of the Fourier basis — in almost all known cases, the representation theory of the point group provides most of the parameters — whereas the vector \(t\) accounts for the relative phase shift between the various incommensurate components.

These two characterizations of quasiperiodicity (in direct and in reciprocal space) are equivalent [4]. Indexing the Bragg reflexions determines, up to equivalences intrinsic to the method, the space group of the D-periodic structure as well as its orientation with respect to the physical space. In this idealized picture, defects, grain patterns etc., are left aside.

Notice that, as described so far, the section method - whence quasiperiodicity - only fixes the long range order of the model. To account for the short range or local ordering, one ought to specify what is inside a unit cell of the D-lattice. For example, a hyper-surface might be imbedded. If it is almost everywhere transversal, a cut by \(E\) would produce a quasiperiodic 2D surface. Since no such symmetry has (yet?) been reported for real surfaces

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or interfaces, I leave this subject; but the tools are here...

For quasicrystals, the "local problem" has not been satisfactorily solved yet. The question is of importance, however, because it is not restricted to geometry in a purely descriptive point of view; answers might have issues as to explain the onset of quasiperiodic order or the stability of quasicrystals. Even phenomenological properties like transport or susceptibilities are believed to depend on local features. After studies on 1-dim systems, locally different models, even with common long range order, may have different behaviors. This contrasts with conventional crystals where, e.g., the band structure follows from just the action of the discrete translation group.

At small scale, a solid phase is made of atoms with well defined equilibrium sites (small rigid clusters may eventually be substituted to atoms in a structural description). Since a site is an isolated point, in order to get it as the result of a generic cut by \( E \), the object to be cut must be a surface (manifold) of codimension \( d \) in \( \mathbb{R}^D \).

An example: modulated crystals

The first application of the cut method to solid state physics is due to Janner and Janssen [5]. In a modulated crystal every atomic site \( r_n \) is referred to a site \( n \) in a regular crystal and the displacement of \( r_n \) with respect to \( n \) is provided by a vector field \( u \) called the modulation: \( r_n = n + u(n) \). The field \( u \) is periodic on \( E \) and its invariance lattice \( U \) is incommensurate with the reference lattice \( L = \{ n \} \). Usually, the field \( r \rightarrow u(r) \) is an analytic and slowly varying function whose Fourier transform, "convoluted" with the reciprocal lattice \( L^* \), appears as satellite spots around each main reflexion. If \( d' \) is the dimension of the Fourier spectrum of \( u \) (which is the lattice dual to \( U \); \( d-d' \) is the dimension of the maximal subspace along which \( u \) is constant), then both a \( D \)-lattice \( \Lambda = \mathbb{R} \times \mathbb{R} \) isomorphic to \( L \times U \) and the physical space \( E \) can be imbedded in \( \mathbb{R}^D \) (\( D=d+d' \)) in such a way that \( p(\Lambda)=L \) and \( p'(\Lambda)=U \). Here, \( p \) and \( p' \) are linear projectors onto the space \( E \), \( p \) has kernel \( E_{\mathbb{R}} \) spanned by \( U \), \( p' \) has kernel \( E'_{\mathbb{R}} \) spanned by \( L \). In \( \mathbb{R}^D \), the graph of \( y \rightarrow u(p(y)) \), \( y \in E_{\mathbb{R}} \), is a \( d' \)-dimensional surface "above" \( E_{\mathbb{R}} \), periodic along the \( L \) directions. Building replicas of this graph along the lattice \( L \) generates a surface \( A \) which is \( \Lambda \)-periodic. Its intersection with \( E \) is precisely the modulated crystal.

**Remarks**

1. If the amplitude \( ||u|| = \sup_r |u(r)| \) is not too large, the various "sheets" never touch each other. So the atomic surface \( A \) is disconnected. There is one connected component for each atom or, equivalently, for each reference crystal site.

2. The "modulo \( \Lambda \)" operation projects the surface \( A \) onto a closed surface in the torus \( T^D \). Then \( (y, \alpha) \rightarrow \alpha u(p'(y)) \mod \Lambda, (y, \alpha) \in E_{\mathbb{R}} \times [0, 1] \), is a continuous interpolation (isotopy) from the atomic surface \( A^c \) to a basis torus \( T^d \) in \( T^D \). It induces a uniformly bounded and continuous displacement between the modulated crystal and the reference lattice. Quasicrystals are not of this type, as we shall see.

3. For small enough \( ||u|| \) again, there is a lower bound to the pair distances. Beyond that minimal value, the
inter-atomic distances vary indefinitely throughout the modulated crystal: in this case, the pair correlation function has a smooth and continuous profile beyond the core.

4. If the pair interactions decay sufficiently fast at large distances, and in absence of external fields, the (free) energy does not change if one shifts the phases between the various incommensurate components. This yields an infinite class of degenerate equilibrium states which are obtained, in the cut method, by parallel shifts \( E \rightarrow E + t \) of the cutting space. The Goldstone modes corresponding to this "broken symmetry" are called phasons.

2. Atomic surfaces in quasicrystals.

At first glance, two major features make the quasicrystals different from modulated crystals: a) the main Bragg reflexions do not stay on, or close to, any lattice. b) the Patterson diagrams \([6,2]\) (representing the two-points correlation function) show highly pronounced peaks which entail that the local atomic patterns are somehow rigid and that the whole structure is built out of a finite number of different types of "pieces", just as a tiling. This explains the analogy there is between quasicrystals and quasiperiodic tilings like Penrose's tilings of the plane or the icosahedral tiling of the 3d space by Hammann's rhombohedra \([2]\). These tilings are known to be quasiperiodic as they can be produced by a cut technique \([7]\).

It must be recalled, however, that the Penrose tilings are very special in that the tiles are rhombi (rhombohedra) which turn out to be projected facets of the simple cubic lattice in higher dimension. If one wants to fit the basic crystallo-chemical data (typical inter-atomic distances, densities, diffraction intensities, etc.), in particular those concerning local order, then the various possible decorations of the Hammann tiles do not yield satisfying solutions. On the other hand experiments do not provide tiles, but only correlations. So there is no need, a priori, to base the analysis of the structure on any specific tiling. The structural problem can be stated directly in terms of atomic positions, that is, taking into account the (global) quasiperiodic order, as finding the correct atomic surface. As mentioned, no definite model has yet been proposed for any of the known quasicrystals. On a more general level, at least for quasicrystals, like the icosahedral ones, involving atomic surfaces of dimension strictly larger than two, a complete classification is still missing as far as I know. Here is, nevertheless, a list of basic properties, either observed or conjectured, a surface should satisfy in order to be a "reasonable" candidate:

1. The surface has dimension \( D - d \), imbedded in the \( D \)-dimensional affine space. For example \( d = 3, D = 6 \) for icosahedral quasicrystals. In particular, the surface is not "separating" as soon as the physical dimension \( d > 1 \).
2. It is periodic, the space group being deduced from indexing of the diffraction spots.
3. It is locally "flat", and the transversal (w.r.to the cut \( E \)) components are all parallel (to the space \( E^\perp \) perpendicular to \( E \), by a suitable choice of scalar product in \( \mathbb{R}^D \)).
4. It is closed (free of boundaries or "holes")
5. Its topology is different from that of modulated crystals.
Comments
*1, 2 and 3 summarize features already discussed. In particular, 3 is the local rigidity property.
*All five properties are compatible with an underlying tiling in the physical space. This will be explicitly demonstrated in the last section on a specific example: the octagonal tiling. A fragment is shown on the left hand side of the above figure.
*Point 5 means that the surface is not equivalent to a periodic family of (D-d)-planes. In the known examples, this property is a consequence of the point symmetry. Topology has been proposed as a mean of classification by Katz [8]. The octagonal tiling will provide an example.
*Point 4 needs explanations that I can only sketch here. One physical reason for wanting closed surfaces is mere conservation of matter in "phason modes" [8,9,10]. These modes can be described by long wavelength fluctuations in the phase shift t, which then becomes a transversal field over E : t(x). If there were boundaries in A, by "ergodicity" of the cut in the torus, "tuning on" an arbitrarily small shift would make the cut E+t(x) leave the atomic surface at some point, thereby canceling an intersection site, thus an atom! The only setting compatible with conservation of atoms is when the local event "E+t(x) leaves A" occurs at the same time as "E+t(x') enters A" somewhere else but not too far from x. In the long variation limit t(x) = t = const. this implies that the two boundary elements crossed at x and x' respectively are related by a vector of E. Then it does not cost anything to consider these two elements as joined together by a non-transversal piece, since parallel pieces are not "seen" in generic cuts. The feasibility of such a closure everywhere on A is of course a severe constraint to the profile of the transversal components (see the example in sec.3).
*Another criterion enforcing the closure (pt. 4) is "homogeneity" in the density if bulk matter. Take the $Z^D$ orbit of any smoothly bounded open region in $E^\perp$ and look at the intersection with $E$: this is a perfectly quasiperiodic discrete set in E. But generically this set contains a finite density of vacancy-like, or interstitial-like, sites which are, however, not defects. Globally, these inhomogeneities (weak enough to remain compatible with quasiperiodicity), preclude transitions to any periodic structure by uniformly bounded displacements [11,12], in contradiction with the apparent "proximity" of the crystalline approximant phases [3].

3. The "atomic surface" of the octagonal tiling.

In this section, I describe the surface, periodic in D=4 dimensions, which gives the set of vertices of the octagonal tiling. This is the simplest example of an authentic quasicrystal; indeed in d=1 dimension, quasicrystals are known to be unstable states for classical systems with short range interactions; on the other hand, all codimension 1 quasiperiodic structures are basically equivalent to modulated crystals. So the lowest significant dimension is $D = 2+2 = 4$. Quasicrystals with 8-fold symmetry have actually been observed [13]. The geometrical analysis has many analogies with that of icosahedral tilings [14], but describing realistic models for icosahedral quasicrystals would be technical.

The 8-fold group $C_8$ can be represented by signed permutations of the standard basis $e_1, \ldots, e_4$ of the 4-dimensional vector space $R^4$: $e_1 \to e_2 \to e_3 \to e_4 \to -e_1 \to -e_2 \to -e_3 \to -e_4 \to e_1$. This representation leaves the lattice $Z^4$ generated by $(e_1, \ldots, e_4)$ invariant, as well as the two complementary planes $E = p(R^4)$ and $E^\perp = p^\perp(R^4)$, where

$$p = 1 - p^\perp = \begin{pmatrix} 1 & 0 & 0 & -s \\ s & 1 & 0 & 0 \\ 0 & s & 1 & 0 \\ -s & 0 & 1 & 1 \end{pmatrix}, \quad s = 1/\sqrt{2}.$$

The projection of the cube $c_4 = \{\sum x_i e_i | 0 \leq x_i \leq 1\}$ into $E^\perp$ is a regular octagon $O = p^\perp(c_4)$. The $Z^4$ orbit of this octagon is the atomic surface of an octagonal quasiperiodic tiling, more precisely the transversal part of this
atomic surface. This means that the intersection $X_0$ of $0 + \mathbb{Z}^4 \equiv \bigcup_{x \in \mathbb{Z}^4} 0+x\mathbb{Z}^4$ with the physical space $E + t$ is the set of vertices of the tiling. The generic vector $t \in \mathbb{R}^4$ indexes the locally isomorphic—and physically equivalent—tilings. Since the $// component of $t$ corresponds to just a global translation in the physical space $E$, we only need to consider shifts $t$ in $E^4 = \mathbb{R}^4 / \mathbb{E}$ in order to get non-congruent tilings.

To build the "non-transversal" part of the atomic surface $A$, we have to look at non-generic cuts (i.e. non-generic $t$) or, a method which is equivalent, to compare two parallel cuts, close to each other. When a vertex $x$ of the tiling belongs to the interior of its octagon (remember that any vertex of $X_0$ is the intersection of some octagon $O+\xi$, $\xi$ in $\mathbb{Z}^4$, with $E+t$), then it stays such for any shift $t'$ in a sufficiently small neighborhood of $t$. Such vertices will be called $t$-stable.

On the other hand, by irrationality of the cut, some intersections are close enough to the boundary to disappear under very small shifts of $t$. But, because of the particular profile of the octagons, a point reappears as an intersection with another octagon, shifted by $b$ with respect to the former. The vector $b$ is the diagonal of a 3-facet of $c_4$ such that $O$ and $O+b$ share a common edge when projected into $E^4$. Thus, as $t$ moves to $t+e_1$, the intersection simply "jumps" from $x$ to $x'=x+p(b)$ in a way analogous to a local atomic diffusion process. A rectangular bridge $e_3 \times b/\parallel$ between the edges of the two octagons in $\mathbb{R}^4$ will account for this flip-flop.

Similar rectangles must be inserted all over through the lattice $\mathbb{Z}^4$ and in accordance with the $C_8$ symmetry. In this manner, all the edges of the octagons $O+\xi$ are transformed into inner "dihedral" edges in $A$.

To handle the remaining boundaries (the $b/\parallel$ sides of the rectangles), let the shift $t$ move along a small circular path around a vertex of an octagon. The corresponding atom, staying in the surface $A$ and in the physical space $E+t$ all along, flips from $x$ to $x'=x+b/\parallel$, etc. and goes over the 8 vertices of a small octagon of side $b/\parallel$. A $t$-path of $6\pi t$ is necessary before the vertex $x$ comes back to its starting point. Since all the $b/\parallel$ edges of the rectangles are sides of similar small octagons translated by $\mathbb{Z}^4$, filling all the small octagons will cancel the remaining boundaries and make $A$ a closed 4-periodic surface.

Notice that the horizontal pieces—rectangles and small octagons parallel to the physical space—do not appear in generic cuts, but they play a crucial role in the theory of elementary excitations (see sec.2).

In the surface $A$, an octagon $O+\xi$ is connected to 8 nearest neighbors at $\xi \pm b_i$ where

$$ (b_1, b_2, b_3, b_4) = \begin{pmatrix} 0 & 1 & 1 & 1 \\ 1 & 0 & -1 & -1 \\ -1 & 0 & 1 & 1 \\ 1 & 1 & 1 & 0 \end{pmatrix} $$

But $b_1, b_4$ is a basis of $\mathbb{Z}^4$ ($\det(b_1, \ldots, b_4) = 1$), so that all the octagons now belong to one single connected component and $A$ is connected.

The projection $A^c$ of $A$ into the torus $T^4 = \mathbb{R}^4 / \mathbb{Z}^4$ is a compact manifold whose genus is 2, as found, e.g., by Euler formula applied to the polygonal decomposition of $A^c$ inherited from that of $A$. There are 1 O, 1 small O and 4 rectangles per unit cell of $\mathbb{Z}^4$.

The sum of the angles around a vertex of the polygonal surface is $5\pi/2$, so the vertices are loci of negative curvature. Since $A^c$ is homeomorphic to a torus of genus 2, a universal covering of $A^c$ can be drawn by decorating the $[8,8]$ tiling of the hyperbolic plane $H^2$ in a way analogous to the $H^2$ representations of triply periodic surfaces with genus 3 by Charvolin and Sadoc. The covering of $A^c$ is obtained by "blowing" the edges.
of \{(8, 8)\} into rectangles; this opens small octagons at every vertex of \{(8, 8)\} which correspond to the small octagons parallel to $E$ in $\mathbb{R}^4$.

To recover the 4-periodic surface $A$, one has to quotient $H^2$ by the fundamental group $G=\pi_1(A)$; $G$ is a normal subgroup of $T$, itself a pure translation subgroup of $\{8,8\}$ with octagonal fundamental domain. A presentation of the translation group $T$ is: $b_1^{\pm 1}, \ldots, b_4^{\pm 1}; b_1 b_2^{-1} b_3^{-1} b_1^{-1} b_2 b_3 b_4 = 1$. The automorphism group of $A$, $Z^4$, is obtained as $T/G$ ($G$ consists of the elements "forced to identity"). Now $Z^4$ is abelian. So $G$ contains the commutator subgroup of $T$; But since $T$ has four generators (plus the inverses), $T/\text{com}(T)$ is already isomorphic to $Z^4$ and we deduce that $G=\text{com}(T)$.

**Conclusion.** To summarize, I have given, first, a short introduction to quasiperiodicity. This type of ordering is now common in solid state physics. Whether it will show up in real interfaces or surfaces is still a question. With minor modifications, the cut method would provide a geometrical description of such surfaces. In the second part, I have focused on the atomic ordering in realistic quasicrystals. A fully periodic surface naturally comes in, imbedded in the higher dimensionnal space. This provides physical issues to theoretical investigations on periodic surfaces not necessarily imbedded in 3D. For example, it would be helpful to have a classification of periodic icosahedrally symmetric 3-surfaces.

Some features of real oriented surfaces are related to their separating space into disconnected regions. E.g. the minimal genus of a triply periodic surface, modulo the lattice in 3D, is $g=3$. These do not necessarily hold in higher $D$. A priori, in $D>3$ dimensions, the minimal genus of a maximally periodic orientable 2-surface taken modulo the lattice is $g=D/2$. The atomic surface of the octagonal quasiperiodic tiling is an example.

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*Left:* "Exploded" view of $A$, made of rectangles, large and small octagons.

*Right:* Universal covering of $A$ in $H^2$, with 4 generators of $T$. The $\{8,8\}$ tiling is dotted.
References


Introduction to Quasicrystals. series ed. by M.Jaric, Academic press.


