A self-similar ordered structure with a non-crystallographic point symmetry

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A new class of self-similar ordered structures with non-crystallographic point symmetries is presented. Each of these structures, named superquasicrystals, is given as a section of a higher-dimensional “crystal” with recursive superlattice structures. Such structures turn out to be limit-quasiperiodic, distinguishing themselves from quasicrystals which are quasiperiodic. There exist a few real materials that seem to be promising candidates for superquasicrystals.

The Penrose patterns reported in 1974, originally introduced as self-similar structures produced by inflation, are aperiodic structures with long-range positional orders as well as non-crystallographic point symmetries. Since then, several classes of self-similar ordered structures, such as quasicrystals (QCs) discovered in 1984, it has been found that they can be described as quasiperiodic point sets, given as sections of higher-dimensional crystals, or hypercrystals. From the viewpoint of aperiodic or-crystals, QCs including limit quasiperiodic structures (LQPSs) are aperiodic structures with long-range positional orders, namely, limit quasiperiodic structures (LQPSs). In this Letter, we will discuss the following three topics: i) A limit periodic structure can be described as a recursive superlattice structure. ii) A limit periodic structure can be described as a section of a supercrystal that is a higher-dimensional limit periodic structure. iii) There can exist superquasicrystals (SQCs), which are LQPSs with noncrystallographic point symmetries. Subsequent arguments will start from a brief review of QCs. Then a concrete example of octagonal SQCs is introduced, and its properties including its structure factor are investigated. Finally, the theory is generalized.

In classical crystallography, crystals are classified into space groups, in which point groups and translational groups are combined together. Translational symmetry restricts severely point groups for crystals, allowing only 2-, 3-, 4-, and 6-fold rotational symmetries. Yet noncrystallographic point groups are allowed for QCs including D_{8h} (octagonal), D_{10h} (decagonal), and D_{12h} (dodecagonal) in 2D and Y_{h} (icosahedral) in 3D (for a review for QCs, see [1]). A QC having one of these point groups is a d-dimensional section of a hypercystal in 2d-dimensions, with d = 2 or 3 being the number of physical space dimensions. The entities of the hypercystal are not atoms but geometric objects called hyperatoms (atomic surfaces, windows, acceptance domains, etc.). We consider a hypercystal constructed by locating one kind of hyperatoms onto the sites of a Bravais lattice Λ in the 2d-dimensional space E_{2d}. The point group G of Λ is isomorphic with a noncrystallographic point group G in d dimensions. More precisely, G is a direct sum of two groups G and G^⊥ that act on the physical space E_{d} and the orthogonal complement E_{d}^⊥, respectively, where all the three groups G, G, and G^⊥ are isomorphic with each other.

We now begin with some preliminary investigation of an octagonal QC in 2D, which will set the basis of introducing SQCs in a later paragraph. For the octagonal QC, the relevant lattice Λ is a hypercubic lattice in 4D. Let Λ be its projection onto the physical space E_{2}. Then, it is an additive group composed of vectors such that the addition of its two members belongs to it and any member, ∈, and its inversion, − ∈, belong together to it: Λ is mathematically a module. Every “lattice vector" ∈ in Λ is uniquely defined as an integral linear combination of the generators, e_{i} (i = 0 - 3), which are the projections of the four standard basis vectors of Λ onto E_{2}. These generators point toward four successive vertices of a regular octagon centered at the origin, and the point symmetry of Λ is equal to D_{8}. Λ is a dense set in E_{2} because the number of its generators is more than two. If one defines Λ^⊥ and e_{⊥} in E_{2} in a similar way, it can be shown that e_{⊥} = (-1)e_{i}, which means that Λ and Λ^⊥ are identical as sets of vectors. Obviously the 4D vectors e_{i} := (e_{i}, e_{⊥}) are the basis vectors of Λ, and the pair of a lattice vector ∈ in Λ and its conjugate ∈' in Λ^⊥ form a 4D lattice vector : = (∈, ∈') in Λ. If the hyperatom is a regular octagon such that its sides are translates of the eight vectors, ±e_{⊥}, the resulting octagonal QC is a discrete subset of Λ, yielding an octagonal tiling with a square and a 45° rhombus; the “bonds” of the tiling are translates of ±e_{i}.

We have already seen several examples of triplets of objects, {X, X^⊥, X}, associated with the three worlds, E_{2}, E_{2}^⊥, and E_{A}, where the latter two of the triplets are uniquely determined by the first, X. There exists an important triplet of linear maps {ϕ, ϕ^⊥, ϕ} with ϕ (resp. ϕ^⊥) being a scaling transformation with the ratio τ := 1+√2 (resp. τ := 1−√2) whereas ϕ is a 4D map defined as ϕ = φ ⊕ φ^⊥ or, equivalently, ϕ = (φ 0 -φ^⊥); we may
The self-similarity is represented as the inflation rule for the octagonal QC above has a self-similarity with ratio \( \tau \). We may call \( \tau \) a bi-scaling. Note that \( \tau \) is the algebraic conjugate of \( \tau \) and written as \( \bar{\tau} = -1/\tau \). A simple geometrical consideration proves that \( \tau \mathbf{e}_1 \) and \( \tau^{-1} \mathbf{e}_1 \) belong both to \( \hat{\Lambda} \), and \( \Lambda \) has the scaling symmetry, \( \tau \Lambda = \Lambda \), where \( \tau \Lambda \) stands for the set of all the vectors of the form \( \tau \ell \) with \( \ell \) in \( \Lambda \). It follows that \( \bar{\varphi} \Lambda = \hat{\Lambda} \). In fact, we may write \( \bar{\varphi}(\mathbf{e}_0 \mathbf{e}_1 \mathbf{e}_2 \mathbf{e}_3) = (\mathbf{e}_0 \mathbf{e}_1 \mathbf{e}_2 \mathbf{e}_3) \mathbf{M} \) with

\[
\mathbf{M} = \begin{pmatrix}
1 & 1 & 0 & -1 \\
1 & 1 & 1 & 0 \\
0 & 1 & 1 & 1 \\
-1 & 0 & 1 & 1 \\
\end{pmatrix}
\]

being a unimodular 4D matrix \( \mathbf{M} \); \( \det \mathbf{M} = 1 \). We shall call \( \mathbf{M} \) the companion matrix of \( \bar{\varphi} \). Using the fact that \( \varphi \) is expansive and \( \varphi^\perp \) contractive, we can show that the octagonal QC above has a self-similarity with ratio \( \tau \). The self-similarity is represented as the inflation rule for the two types of tiles (see Fig.1). Note that a recursive structure is built in the QC by its self-similarity.

Now let us divide \( \hat{\Lambda} \) into two sublattices with respect to the parity of the sum of indices of the lattice vectors. The two sublattices are equivalent, and the volume of their unit cell is twice that of \( \Lambda \); the multiplicity of each sublattice is equal to 2. Every “bond” of the octagonal tiling connects two vertices with opposite parities. The vertices of the inflated tiling come evenly from the two equivalent sublattices because the scaling \( \varphi \) does not change the parity of a lattice vector. For the sake of a later argument, we shall investigate the even sublattice, \( \Lambda_e \), of \( \hat{\Lambda} \). It is a simple algebra to show that \( \mathbf{e}_i' := \mathbf{e}_i + \mathbf{e}_{i+1} \) with \( \mathbf{e}_1 := -\mathbf{e}_0 \) are generators of \( \Lambda_e \), i.e., the projection of \( \hat{\Lambda} \). We may write \( \mathbf{e}_i' := \sigma \mathbf{e}_i \) and, hence, \( \Lambda_e = \sigma \Lambda \), where \( \sigma \) is a similarity transformation that is a combined operation of a scaling through \( 2 \cos(\pi/8) = (2 + \sqrt{2})^{1/2} \) and a rotation through \( 22.5^\circ = (\pi/8) \). The conjugate \( \sigma^\perp \) of \( \sigma \) is a scaling through \( 2 \sin(\pi/8) = (2 - \sqrt{2})^{1/2} \approx 0.765 \) and a rotation through \(-157.5^\circ = -7\pi/8 \). The 4D lattice \( \Lambda_e \) is written with \( \hat{\sigma} = \sigma \oplus \sigma^\perp \) as \( \Lambda_e = \hat{\sigma} \Lambda \); we may call \( \hat{\sigma} \) a bi-similarity transformation. The companion matrix of \( \hat{\sigma} \) is given by

\[
\begin{pmatrix}
1 & 0 & 0 & -1 \\
1 & 1 & 0 & 0 \\
0 & 1 & 1 & 0 \\
0 & 0 & 1 & 1 \\
\end{pmatrix}
\]

which is not unimodular: \( \det \mathbf{M} = 2 \). We may consider \( \Lambda_e \) to be a superlattice of \( \hat{\Lambda} \). An infinite series of modules is generated as \( \Lambda_n := \sigma^n \Lambda \), \( n = 1, 2, \ldots \), each of which is a projection of the relevant superlattice \( \Lambda_n (= \hat{\sigma}^n \Lambda) \); the index of \( \Lambda_n \) in \( \Lambda \) is equal to \( 2^n \). Note that \( \Lambda_2 = (2 + \sqrt{2}) \Lambda = \sqrt{2} \Lambda \) because \( \Lambda \) is invariant against both the \( 45^\circ \) rotation and the \( \tau \)-scaling.

We show in Fig.2 another self-similar octagonal tiling produced by an inflation rule. It is shown later on that the vertices of the tiling form an SQC. Since the “bonds” of the tiling are translates of \( \pm \mathbf{e}_i \), the SQC is a discrete subset of \( \Lambda \), and is given as a section of some 4D structure on \( \hat{\Lambda} \); we shall call the 4D structure a supercrystal. Inspecting Fig.2 we find that the vertices of the inflated SQC come only from the even sublattice, \( \Lambda_e \), and the inflation is equivalent to a similarity transformation through \( \sigma \). Moreover, the double inflation of the SQC in Fig.2 is equivalent to the \( 2 + \sqrt{2} \)-scaling of the SQC. It is now evident that the recursive structure being built in the SQC by its self-similarity requires that the supercrystal has a recursive superlattice structure associated with the
infinite series of superlattices. Therefore, the supercrystal is formed by hyperatoms whose shapes, sizes, and/or orientations depend on the local environments of the relevant sites of the supercrystal; the hyperatoms are not uniform.

While a QC or an SQC is defined as the projection of a subset of \( \Lambda \) onto \( E_2 \), its conjugate set is defined as the projection of the same set onto \( E_2^\perp \). The conjugate set of the octagonal QC is a dense set bounded by the same octagon as the hyperatom. Hence the shape of the hyperatom is retrieved from the conjugate set. The conjugate set of the octagonal SQC is not so simple because hyperatoms are not uniform. If we ignore higher order superlattice structures than a specified order, \( n \), we obtain a hypercrystal whose Bravais hyperlattice is equal to \( \Lambda_n \); its unit cell includes distinct hyperatoms. It yields the \( n \)-th approximant QC to the SQC. Our investigation of hyperatoms of successive approximants strongly indicates that the hyperatoms of the present SQC are topologically disks but their boundaries are fractals. Nevertheless the present SQC satisfies the so-called gluing (or closeness) condition for the gluing condition, see \([13]\).

The two maps, \( \varphi \) and \( \sigma \), are Pisot maps; a general Pisot map \( \psi \) is a member of the triplet \( \{ \psi, \psi^+, \psi^- \} \) such that i) \( \psi \) is a bi-similarity transformation. ii) \( \psi \) is expansive but \( \psi^+ \) contractive. iii) \( \psi \Lambda \) is identical to or a submodule of \( \Lambda \). Conversely, any submodule of \( \Lambda \) can be given as \( \psi \Lambda \) with a Pisot map \( \psi \) provided that the submodule is geometrically similar to \( \Lambda \) \([12]\); the index of the submodule in \( \Lambda \) is given by \( m := |\text{det } M| \) with \( M \) being the companion matrix of \( \psi \). The structure factors of self-similar structures based on Pisot maps are known to be composed only of Bragg spots \([7, 8]\). The companion matrix of the Pisot map associated with a QC is “unimodular” because \( m = 1 \), while the one with an SQC is not because \( m \geq 2 \). A recursive superlattice is considered to be a Bravais hyperlattice, and an infinite number of SQCs are constructed on it. Then, it is important to classify possible recursive superlattices with non-crystallographic point symmetries that are physically important. A recursive superlattice is constructed on the base lattice \( \Lambda \) by the use of a Pisot map \( \psi \).

We begin with the case of octagonal SQCs in 2D. The module \( \Lambda \) and its submodule \( \psi \Lambda \) have the point group \( D_8 \) as their common symmetry group if and only if \( \psi \) is a simple scaling transformation or a similarity transformation including a rotation through \( 22.5^\circ = (\pi/8) \) \([12]\); a Pisot map of the former (resp. latter) type shall be classified into the type I (resp. II). The type I Pisot map is a bi-scaling, \( \hat{\rho} \), specified by so-called a Pisot number, which is a positive number of the form \( \nu := p + q\tau \) with \( p, q \in \mathbb{Z} \) and the magnitude of its algebraic conjugate, \( \bar{\nu} \), is smaller than one. It can be readily shown that \( \rho = p\tau + q\varphi, \hat{\rho} = p\tau + q\bar{\varphi} \) and \( M_p = pI + qM_\varphi \). It follows that \( m = |N(\nu)|^2 \) with \( N(\nu) := \nu\bar{\nu} = p^2 + 2pq - q^2 \). On the other hand, the type II Pisot map is written as \( \psi = \rho\varphi \), so that \( \psi = \hat{\rho}\bar{\varphi} \) and \( M_{\bar{\varphi}} = \hat{\rho}I\bar{\varphi}M_\varphi \). It follows that \( m = 2|N(\nu)|^2 \). Thus an infinite number of recursive superlattices with the octagonal symmetry can be constructed on the octagonal hyperlattice \([12]\), whereas...
there is only one hyperlattice (exactly, Bravais hyperlattice) with the octagonal point symmetry \[10\].

The above consideration for the octagonal SQCs is readily extended to the decagonal case (D_{10}), the dodecagonal case (D_{12}), and the icosahedral case (Y_{b}); the last case is for 3D SQCs. We have succeeded in a complete classification of recursive superlattices with these point symmetries. A simplest decagonal SQC, for example, is specified by a type II Pisot map, and its multiplicity (index) is five, while a simplest icosahedral SQC is specified by a type I Pisot map, and its multiplicity (index) is 64. A complete list will be published elsewhere.

The selfsimilarity of a QC or an SQC is based on a Pisot map associated with the hyperlattice \(L\). The presence of a Pisot map is derived from the \((d + d)\)-reducibility of the point group \(G\) of the hyperlattice \(L\) \[5, 12\]. The \((d + d)\)-reducibility is, in turn, a consequence of the fact that the non-crystallographic point group \(G\) is lifted up to \(\hat{G}\). Thus, the selfsimilarity of a QC or an SQC is closely connected with its non-crystallographic point symmetry. A QC and an SQC are distinguished by whether the relevant Pisot map is volume-conserving or not, respectively. SQCs are an important class of aperiodic or- dered structures with non-crystallographic point symmetries, and the \(QCs\) form a special sub-class which, albeit important, is a definite minority in the class. One may consider an SQC as a bizarre object because of its limit-quasiperiodic nature but it is as a natural object as a QC is as mentioned above.

A 2D QC has a set of parallel quasi-lattice-lines \[2\] so that all its lattice points are located dividedly on them. The same is true for an SQC; the quasi-lattice-lines form a limit quasiperiodic grid, whose spacings take the form \(p + q\tau\) with \(p, q \in \mathbb{Z}\) in an appropriate length unit. This allows us to relate 2D SQCs to the 1D analogues, which have been extensively investigated \[7, 8\]. It is readily shown that SQCs formed by this method satisfy the gluing condition. Moreover, the hyperatoms are shown to be polygons, so that their boundaries are not fractals. The grid method is extended to the 3D case.

An exact mathematical formulation of the limit periodic structure is made by using locally-compact Abelian groups \[8\], which are rather transcendental objects for physicists in condensed matter physics or for crystallographers. We have succeeded in reformulating it on the basis of the recursive superlattice structure, which is basically an “inverse system” \[8\]. We will not, however, present these technical details here; instead, we enumerate several merits of this approach. i) This approach to SQCs is a simple extension of the super-space approach to QCs \[14\]. ii) It explains naturally why the structure factors of SQCs are composed only of Bragg spots. iii) The Fourier modules of SQCs are readily calculated \(cf.\ [7, 8]\). iv) Non-crystallographic point symmetries are easily included.

Quite recently, the MLD (Mutual-Local-Derivability) classification of quasicrystals has been completed \[14\]. It is an urgent task to perform an MLD classification of SQCs. Prior to do it, however, one must specify all the SQCs satisfying the gluing condition.

A superlattice ordering associated with the sublattice with multiplicity (index) five was observed in an Al-Cu-Co decagonal quasicrystal \[15\]. An Al-Pd-Mn icosahedral quasicrystal is known, on the other hand, to exhibit a superlattice ordering such that its hyperlattice constant is doubled \[10\]. A superlattice ordering with a large multiplicity as this case is unusual for conventional alloys but could be interpreted naturally as an icosahedral SQC. Anyway, these two “QCs” are promising candidates for SQCs.

SQCs as well as QCs are quite natural structures from the point of view of the generalized crystallography. It is surprising that such an important class of structures as that of SQCs has been entirely overlooked up to the present. Discovering and/or synthesizing SQCs is a big challenge in material science, and its achievement will be a triumph of the generalized crystallography. Moreover, there exists strong evidence that the nature of the electronic states in an SQC is markedly different from that in a QC \[17\].

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