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Symmetries of Quasicrystals

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We consider tiling models of "round quasicrystals" which would have diffraction patterns which are fully rotation invariant—rings instead of Bragg peaks. They can be distinguished from glasses by self-similarity of the pattern of radii of the rings.

KEY WORDS: Quasicrystals; diffraction; symmetries; aperiodic tiling; hierarchical quaquaversal.

Quasicrystals were discovered by Schectman *et al.*[1] due to the unusual 10-fold rotational symmetry exhibited in certain directions by their (electron) diffraction patterns, a symmetry which is well known to be impossible for ordinary crystals. An early model to explain this symmetry was put forward by Levine and Steinhardt,[2] based on an observation of Mackay[3] that a particle configuration associated with a (3-dimensional version of) a planar Penrose tiling (Fig. 1) would have diffraction patterns with just such a forbidden symmetry. (The *qualitative* features of the diffraction patterns are independent of precisely how scatterers would be associated with each type of tile, as long as no accidental symmetries are introduced by their placement.)

There was some confusion at first due to the "local 5-fold symmetry" of the Penrose tilings; the Penrose tilings have arbitrarily large regions with centers of 5-fold rotational symmetry. Any such symmetry in the real-space pattern of the scatterers will produce the same rotational symmetry in the diffraction pattern in certain directions. But it was eventually realized that the key feature of the Penrose tilings is their "statistical 10-fold symmetry" —the fact that every finite pattern of tiles in the tiling appears in 10 different rotational orientations with the same frequencies, the *equality* of the

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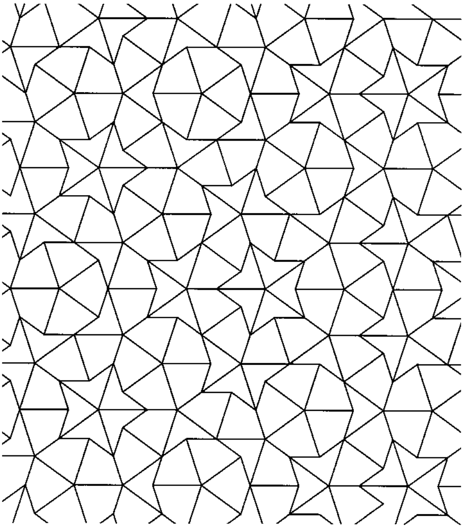

Fig. 1. A Penrose tiling.

frequencies giving rise to the 10-fold rotational *symmetry* of the diffraction patterns;[4, 5] diffraction is only sensitive to such frequencies, and the well known transference, of a real-space symmetry of the pattern of scatterers to a symmetry in the diffraction intensity, is only a special case of this.

The other crucial feature in this modelling was that the Penrose tilings, and its various "aperiodic" analogues in 2 and 3 dimensions, have "matching rules," which means that when the matching rules are used to make a tiling from the tiles, the only tilings *possible* are the desired ones. Matching rules govern how the boundaries of tiles may abut; Fig. 2 shows the two basic quadrilateral shapes (the "kite" and "dart") for Penrose tiles, and Fig. 3 shows how they can be modified by matching rules so as to restrict

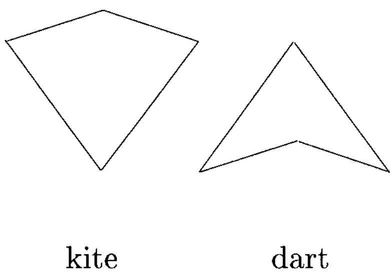

Fig. 2. The kite and dart.

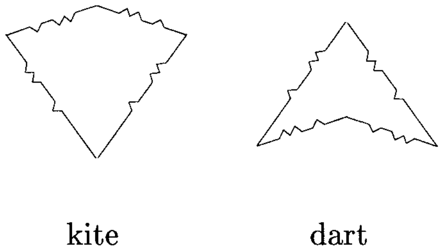

Fig. 3. The modified kite and dart.

how edges of tiles may abut in a tiling. (If the Penrose matching rules are not enforced, the quadrilateral Penrose tiles could be used to make uninteresting tilings.) The restricted tilings made using matching rules can be thought of, at least heuristically, as energy ground states—abutting tiles having "low energy" when their boundaries match and having "high energy" otherwise; an allowed tiling (as opposed to a tiling made without respect to the matching rules) is clearly a lowest energy configuration. Many generalizations of the Penrose tilings were produced over the years using a projection technique due to N. G. de Bruijn.[6] This technique proved able to produce tilings, some with matching rules, with other forbidden rotational symmetries.[7]

A new phenomenon, not possible from tilings made by a projection technique, was exhibited by the planar "pinwheel" tilings,[8] and the recently published[9] 3-dimensional "quaquaversal" tilings (Figs. 4 and 5), based on generalizing a different feature of the Penrose tilings—the fact that they have a *hierarchical structure*. Such tilings can be made using "inflation," that is, using rules for dividing tiles into smaller versions of themselves. Figure 6 shows the inflation rule for pinwheel triangles. A pinwheel tiling can be made by starting with a 1, 2, $\sqrt{5}$ right triangle, dividing it, then expanding about some point by a linear factor of $\sqrt{5}$, as shown. Then apply the division rule to each of the 5 triangles thus produced and expand the result about some point, getting 25 triangles, etc. The Penrose tilings can also be obtained by an inflation rule instead of matching rules, but we will not show this because it is a bit more complicated.[10]

So the Penrose tilings can be thought of as produced in any of three essentially different ways—by enforcing matching rules, or by a projection technique, or by an inflation process. The pinwheel and quaquaversal tilings can be produced either by matching rules or an inflation process,[8, 11] but not by projection. (It is necessary that the tilings used to model materials be enforced by matching rules if one wants the structure to be thought of as an energy ground state.)

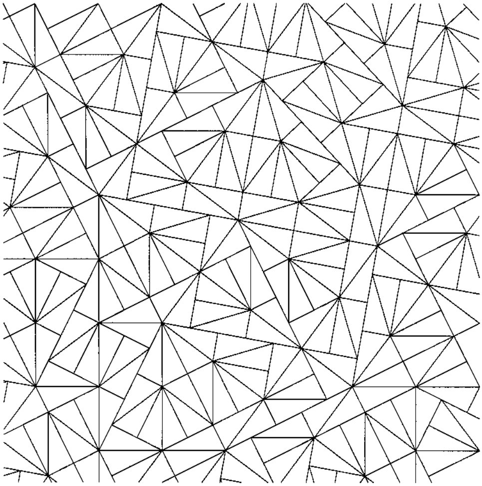

Fig. 4. A pinwheel tiling.

The new feature that the pinwheel and quaquaversal tilings have in common is that they give rise to diffraction patterns which are *fully rotation invariant*;[12, 9, 5] the diffraction consists of uniform rings rather than isolated Bragg peaks. (It is not known if the rings are sharp or diffuse.) In accordance with the above this just corresponds to the fact that any finite structure in such a tiling appears in that tiling with the same frequency (density) in all rotational orientations.

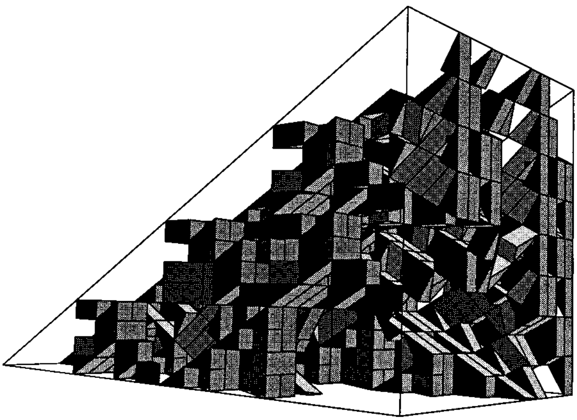

Fig. 5. A quaquaversal tiling.

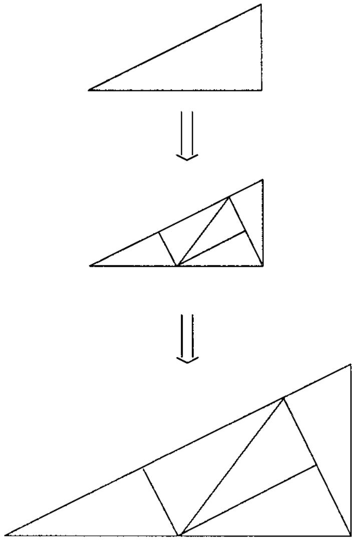

Fig. 6. The pinwheel substitution.

The value of this rotational symmetry of these tilings is that it points to a possibility not previously known. Given diffraction data from bulk material which is rotation invariant (made of rings), it has been natural to suppose the material *must be* either a conglomerate of randomly oriented microcrystals (like a powder) or a glass, i.e., a frozen liquid. These new models point to the *possibility* that the material could be a single quasi-crystal—a fully deterministic structure.

We emphasize that although the above tiling models (pinwheel and quaquaversal) of what might be called "round quasicrystals" are to some extent closely related to the older models built on Penrose-like tilings, there are two points worth noting. First, it is not possible to make a model of a round quasicrystal using projection techniques, which have been much more widely used than inflation techniques. And second, the very possibility of round quasicrystals is not well known, so that diffraction data showing rings could easily be misinterpreted as indicating something other than a fully deterministic (quasicrystalline) structure.

A possible way to distinguish such a quasicrystal from a glass would be to examine the radial part of the diffraction data: if the material structure is hierarchical as in a quaquaversal tiling, then whatever pattern of rings there is with radii between r_1 and r_2 will appear also with radii between κr_1 and κr_2, where κ is the inflation factor of the hierarchy; $\kappa = 2$

for the quaquaversal structure. We show next how this diffraction symmetry comes about for hierarchical structures.

We use the ergodic theory framework for diffraction[13, 5] as it is particularly convenient for symmetries. In this framework one embeds the structure of interest (for us a set of scatterers) in a space X of similar ones, by making all translates of the original one and completing that set in some natural metric (thus creating X). This is completely analogous to the way one analyzes a time series, by embedding it in a family of sequences. In the usual way, the ergodic theorem associates frequencies in the structure of scatterers, as discussed above, with a probability distribution on the space X of such structures. Then this distribution allows one, just as in quantum mechanics, to represent translations on X by unitary representations on the Hilbert space $\mathscr{H}$ of square integrable functions on X, the unitarity resulting from the fact that the frequencies are invariant under translation of the structures.

Now let us consider symmetries other than translations. As noted above the frequencies of Penrose tilings are invariant under rotation by $2\pi/10$, and the frequencies of pinwheel and quaquaversal tilings are invariant under all rotations. We can therefore repeat the argument at the end of the last paragraph to show that rotation by $2\pi/10$ is unitarily implemented on the Hilbert space for Penrose tilings, and all rotations are unitarily implemented on the Hilbert spaces for pinwheel and quaquaversal tilings. In a more interesting way we can also repeat the argument for each hierarchical system for its similarity κ, which shows that stretching by $\kappa = (1+\sqrt{5})/2$ for Penrose tilings, $\kappa = \sqrt{5}$ for pinwheel tilings and $\kappa = 2$ for quaquaversal tilings are each unitarily implemented.

What is less obvious is that such rotational and similarity symmetries lead to symmetries of the spectral projections of translations. For a system such as the quaquaversal tilings it follows from the above and the uniqueness of the spectral projections E_s of translations that the projections are symmetric in the sense that for any rotation R^{Θ} of Euclidean space we have $E_{R^{\Theta}s} = U^{\Theta} E_s U^{-\Theta}$, where U^{Θ} is the unitary representation on $\mathscr{H}$ of the rotation R^{Θ}, and $E_{\kappa s} = U^{\kappa} E_s U^{-\kappa}$, where U^{κ} is the unitary representation of the inflation κ. Diffraction intensities are given by $I(s)\, ds = d\langle f, E_s f\rangle$ where $f \in \mathscr{H}$ incorporates the structure of the charge density of a single scatterer.[13, 5] Therefore $I(R^{\Theta}s)\, ds = d\langle U^{\Theta}f, E_s U^{\Theta}f\rangle$ and $I(\kappa s)\, ds = d\langle U^{\kappa}f, E_s U^{\kappa}f\rangle$. If the charge density of the scatterers is rotation invariant then $U^{\Theta}f = f$ and the diffraction intensity is rotation invariant: $I(R^{\Theta}s)\, ds = I(s)\, ds$. This is why diffraction off such a structure gives rings rather than Bragg peaks.

We now come to the issue of distinguishing quasicrystals from glasses. Of course the charge density cannot be invariant under the similarity κ, so we cannot conclude as much as we could for rotations. However we still have $I(\kappa s)\, ds = d\langle U^{\kappa} f, E_s U^{\kappa} f\rangle$. We just showed that this function is independent of angle, and therefore only depends on the radial component $|s|$ of s. The diffraction rings are the local maxima of this function of $|s|$. And although we cannot say that the function is invariant under κ, we can still see as evidence of the inflation symmetry a self-similarity in the structure of these local maxima: whatever pattern of rings there is with radii between r_1 and r_2 will appear also with radii between κr_1 and κr_2. In other words the pattern of radii repeats at different scales, although the intensity of the diffraction at corresponding radii need not be simply related.

In summary, quasicrystals with a hierarchical structure exhibit a self-similarity in the radial part of their diffraction, and this could be used to distinguish "round quasicrystals" from glasses. Distinguishing them from conglomerates of randomly oriented microcrystals is presumably easier.

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