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Quasicrystals with arbitrary orientational symmetry

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We introduce a "generalized dual method" for generating quasicrystal structures with *arbitrary* orientational symmetry in two and three dimensions.

Quasicrystals[1] are ordered structures with long-range quasiperiodic translational order and long-range orientational order. Incommensurate crystals are a special case for which the orientational order corresponds to an allowed crystal symmetry. For this case, the incommensurate length scales are unconstrained by the orientational symmetry. In general, though, quasicrystals can have disallowed crystallographic orientational symmetries in which case the incommensurate length scales are constrained to special ratios. These noncrystallographic cases have fundamentally different mathematical and physical properties from crystals or incommensurate crystals.[1,2-4] Quasicrystals have a diffraction pattern composed of pure Bragg peaks that densely fill reciprocal space forming a pattern with a symmetry corresponding to the orientational order of the quasicrystal.[1] Recently, Shechtman, Blech, Gratias, and Cahn[5] have reported a rapidly quenched alloy of Al-Mn that exhibits a diffraction pattern of sharp peaks in an icosahedrally symmetric pattern that corresponds to the computed diffraction pattern for an icosahedral quasicrystal.[1,6-8] Icosahedral quasicrystals have therefore been studied in some detail already.

The purpose of this paper is to introduce a "generalized dual method" (GDM) that can generate quasicrystals in two (2D) or three (3D) dimensions with *arbitrary* orientational symmetry. Many orientational symmetries for 3D quasicrystals have already been studied: (i) Quasicrystals with orientational symmetry corresponding to any regular polygon are possible in 2D.[9] The 2D patterns may be trivially extended to form 3D uniaxial structures with periodic or quasiperiodic ordering in the third dimension. (ii) The only intrinsically 3D quasicrystal (other than incommensurate crystals) that has been discussed is the icosahedral quasicrystal (and various duals, e.g., dodecahedral). All these cases are self-similar.[1,10] Examples of icosahedral quasicrystals[11] have been generated with use of the matching and "inflation" methods discussed in Ref. 1, special projections from higher-dimensional periodic (cubic) lattices,[6-8,12] and the GDM discussed in this paper.[10]

The GDM can be used to generate any of the structures obtained by the other methods. One advantage of the GDM is that, for any fixed orientational symmetry, it generates a much wider class of space-filling patterns than any of the other methods. The key advantage of the GDM so far as this paper is concerned, though, is that it is a simple method for generating quasicrystals with *any* orientational symmetry. Although quasiperiodic and orientationally ordered, the new possibilities generally have lower symmetry than the cases described above and, as a result, are not necessarily self-similar. (In this sense, we are broadening the definition of quasicrystal introduced in Ref. 1.)

The GDM is a generalization of a method developed by deBruijn[13] to obtain 2D Penrose tilings.[14] The method of generating a 3D quasicrystal, say, consists of the following five steps: (1) A "star" of N 3D vectors is chosen which determines the orientational symmetry of the quasicrystal. For the regular icosahedron, a star of $N=6$ unit vectors can be chosen, each of which is parallel to one of the six fivefold symmetry axes of an icosahedron. (2) An (infinite) set of periodically or quasiperiodically spaced parallel planes is introduced normal to each star vector, $\mathbf{e}_i$. Randomly spaced planes can produce a structure with long-range orientational order but not translational order. The unit of spacing between planes normal to any given $\mathbf{e}_i$ is proportional to the length of $\mathbf{e}_i$. Each set of parallel planes forms a "grid." Taken together, the grids form an "N-grid" which lies in a 3D space that we will term a "grid space." We note that there are many degrees of freedom in this prescription for the N-grid: The translation of each grid normal to its planes and, for quasiperiodically spaced planes, the incommensurate length scales and the quasiperiodic sequence of spacings between grid planes. For simplicity, we will assume that the degrees of freedom have been fixed so that at most three planes intersect at any given point.[10] (3) Each plane normal to star vector $\mathbf{e}_i$ is labeled by an integer n_i which represents its ordinal position along the $\mathbf{e}_i$ direction. (4) The planes divide grid space into nonintersecting open regions through which no planes pass (the regions can be arbitrarily small). Each such region is specified (uniquely) by N integers $(k_1,k_2,\ldots,k_N)$: if $\mathbf{x}_0$ is any point in the region then k_i is the label of the plane normal to $\mathbf{e}_i$ such that $\mathbf{x}_0$ lies between the planes labeled by k_i and k_i+1. (5) The "dual" is constructed by mapping each open region in grid space into a point $t=\sum_{i=1}^{N} k_i\mathbf{e}_i$ which lies in a 3D space that we shall term "cell space." The points t are the vertices of a packing of quasicrystal unit cells (see Fig. 1). We will refer to a particular packing of unit cells obtained by a dual transformation to a given N-grid as a "packing" (a "tiling" will refer only to 2D).

A further generalization can be obtained by replacing the grid planes with more general grid surfaces.[10] In this case, there are some subtle topological constraints on the shapes of the surfaces in order to ensure that the unit cells do not overlap in the dual packing.[10,15]

A little practice with the GDM reveals the following properties: (i) Each intersection of three planes in grid space divides 3D space locally into eight regions whose duals correspond to the vertices of a single unit cell in cell space. Thus, the dual to an intersection in grid space is a unit cell in cell space. The unit cell is a parallelepiped with its side lengths and angles determined by the lengths and angles of intersection of the three $\mathbf{e}_i$ normal to the intersecting grid-space planes. The number of different triplets of star vec-

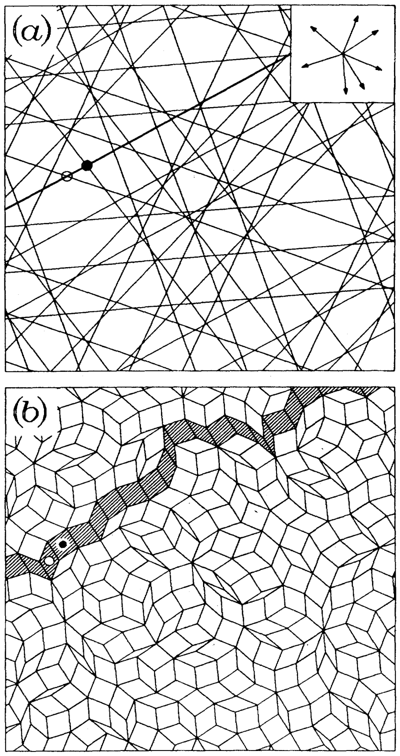

FIG. 1. Illustration of the GDM in 2D for an arbitrary orientational symmetry. (a) A 7-grid based on the star of vectors shown. (b) The dual tiling. The shaded cells in (b) are dual to the intersections along the thick line in (a). (Note that the cells join along edges perpendicular to that line.) The open and solid circles mark the cells associated with the similarly marked intersections of the 7-grid. The cell shape is determined by the angle between grid lines at the intersection. There are 21 different cell shapes in (b).

tors (that is, with different angles of intersection) determines the number of different shapes of unit cells in the packing, which is at most $N(N-1)(N-2)/3!$. (ii) N-grids with periodic spacings can be obtained via projections from higher-dimensional periodic lattices and dual packings can be generated.[12,13,16] The same packings have been obtained by a "direct" projection[6-8,15] (no dual) from five and six dimensions. These methods are all equivalent to the GDM with *periodic* N-grids only and the packings they generate represent a restricted subset of GDM packings for any given symmetry. (iii) When quasiperiodic rather than periodic spacing of planes is used for an N grid, the unit-cell shapes in the packing are unchanged. These depend only on the star vectors that generate the N grid. However, the configurations of cells and, in some cases, the relative numbers of different unit cells in the packing can differ. (iv) The packings generated by a given set of star vectors can be divided into different "local isomorphism" (LI) classes depending on the choice of N-grid parameters. Two packings are in the same LI class if any finite configuration of cells that appears in one packing appears in the other. Two quasicrystals corresponding to packings which are locally isomorphic have the same diffraction peak intensities and elastic energy.[10] For example, the icosahedral packings obtained by the matching rules described in Ref. 1 correspond to one LI class and the packings obtained by projections from 6D correspond to another; a mathematical formalism for determining the N-grid parameters appropriate to these packings is given in Ref. 10. Quasicrystal packings of this type may be of special interest if the matching rules can be realized physically (by bonding of atoms in special clusters, say). In general, though, the physical properties associated with all possible quasicrystal structures should be considered.

The GDM is a simple and powerful method of generating quasicrystals from any fundamental star of vectors. Any quasicrystal generated by the GDM has a long-range orientational symmetry. In particular, each edge of each unit cell is oriented parallel to a star vector. The packing of unit cells is analogous to a Bravais lattice and any decoration of the unit cells with "atoms" will have a diffraction pattern which reflects the orientational symmetry. The construction guarantees that the packing has a well-defined quasiperiodic translational order. As a result, the packing has a diffraction pattern that consists of a dense set of Bragg peaks. The quasicrystal formed from the unit cells also has a nonzero lower bound to the separation between neighboring vertices. Only special cases of high symmetry (e.g., icosahedral) will have the self-similarity properties and close association with algebraic number fields discussed in Ref. 1.

Just as there is a Landau theory for a crystal phase, so there is a Landau description possible for every quasicrystal generated by the GDM method. A Landau theory for the regular icosahedral phase has already been derived by several groups.[2-4,6,17,18] A quasicrystal phase and transitions to and from it are described in terms of a phenomenological Landau free energy that can be expanded in a power series in the density, $\rho(\mathbf{r})$. The expansion is expressed in terms of the Fourier components of the density $\rho_{\mathbf{G}}$, where $\mathbf{G}$ is a vector in reciprocal space. For simplicity, attention is restricted to a subset of $\rho_{\mathbf{G}}$'s which includes the basis vectors, the maximal subset of star vectors such that no vector in the subset can be expressed as an integral linear combination of the others. There is an independent hydrodynamic mode associated with each basis vector. By cutting off the Fourier expansion of $\rho(\mathbf{r})$ to include only a few terms, a density-wave description for the phase can be obtained. As a microscopic structural description for solids at temperatures far from the melting point, though, the density wave description is of limited use.

In Fig. 1 we show a 2D quasicrystal constructed via the GDM by use of seven arbitrary star vectors. As a 3D example, we show in Fig. 2 a slice through a 3D quasicrystal constucted from $N=6$ star vectors which point to six of the twenty face centers of an icosahedron (see inset of Fig. 2). This particular example has some residual icosahedral symmetry that makes it easy to visualize and compare with the more familiar icosahedral (vertex) model. Because of the symmetry, there are only four different rhombohedral unit cells (whereas for $N=6$ there are twenty for arbitrary symmetry or two for the vertex model).

In general, the diffraction pattern consists of a dense set of Bragg peaks that can be indexed by N integers. We do not know an exact method for computing the peak intensities for a general GDM packing with arbitrary symmetry, although there are numerical methods. For the particular case of Fig. 2, each tile vertex is of the form $\sum_{q=i,j,k}(m_q+2n_q/\tau)\mathbf{e}_q$, where i,j,k label different N-grids and m_q,n_q are integers. As a result, the diffraction pattern has the same peak pattern and similar intensity hierarchy as the pattern of an N-grid formed by quasiperiodically spaced planes where the distance

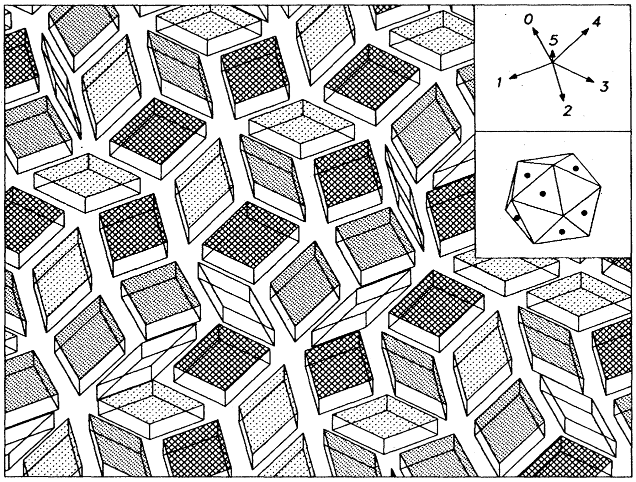

FIG. 2. A slice of a 3D tiling generated from the distorted icosahedral star of vectors described in the text. The cells shown are dual to intersections of grid-space plane normal to $\mathbf{e}_5$. The six star vectors are a subset of the ten independent vectors that point to the face centers of an icosahedron as shown in the insert. The top surfaces of the four different types of cells are shaded differently.

from the origin of the kth plane normal to the $\mathbf{e}_i$ direction is given by $x_k = k + 2/\tau[k/2\tau]$, where the square brackets represent the greatest integer function.[10] The method of computing the diffraction pattern of such an N-grid is described in Refs. 1 and 10. The diffraction patterns in a plane normal to a twofold and broken fivefold axis are shown in Fig. 3. These resemble the patterns obtained for the icosahedral (vertex) model with some noticeable systematic differences due to the broken symmetries.

The fact that the structures are determined by N star vectors and the diffraction patterns are indexed by N integers suggests that the quasicrystals can be understood in terms of projections from higher-dimensional spaces. In fact, the special case of an N-grid with *periodic* spacings between planes and arbitrary symmetry can be obtained by such a projection.[15,16] However, we do not know how to obtain a general GDM packing generated from an N-grid with quasiperiodic spacings as a projection from a higher-dimensional space.

So far it appears that only a few of the many possible quasicrystal symmetries are realized in nature. This might be due to any of several reasons. Perhaps only the symmetrical cases can be ground states or locally stable states for atomic structures. Perhaps only those cases where the structure can be forced by simple local interactions, such as the icosahedral case, are realizable. A fascinating possibility is that perhaps many quasicrystal symmetries are realized in nature, but they have not been detected. In Fig. 1 we show a 2D example of a quasicrystal generated via the GDM using seven arbitrary star vectors. Although the structure clearly has an orientational symmetry, its translational order is not at all apparent. A solid composed of many micrograins of such a quasicrystal phase might be very hard to distnguish from a glass. With many star vectors, the bright diffraction peaks will be very closely spaced (unlike the icosahedral case, say). This, combined with broadening by defects and thermal effects, could make it difficult to detect the translational order without close scrutiny.

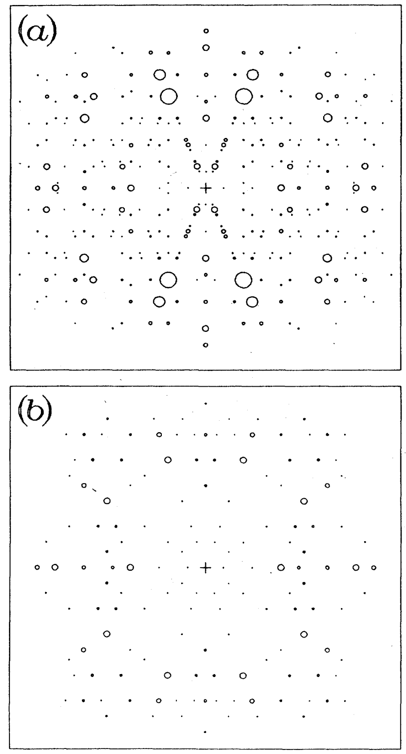

FIG. 3. Diffraction patterns for the distorted icosahedral structure of Figure 2. The radii of the circles are proportional to the Bragg peak intensities. (a) A twofold axis. (b) A broken fivefold axis.

Note added. After submission of our manuscript, we received a manuscript from F. Gahler and J. Rhyner[15] which discusses a "generalized grid method" and its equivalence to direct projection methods. The generalized grid method in their paper corresponds to the GDM for the special case of *periodic* N-grids.

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