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APERIODICITY AND ORDER

Volume 2

Introduction to the Mathematics of Quasicrystals

Edited by

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ACADEMIC PRESS, INC.
Harcourt Brace Jovanovich, Publishers
Boston San Diego New York
Berkeley London Sydney
Tokyo Toronto

ACADEMIC PRESS, INC.
1250 Sixth Avenue, San Diego, CA 92101

United Kingdom Edition published by
ACADEMIC PRESS INC. (LONDON) LTD.
24-28 Oval Road, London NW1 7DX

Library of Congress Cataloging-in-Publication Data

Aperiodicity and order.

Vol. 3 edited by Marko V. Jarić and Denis Gratias.
Includes bibliographies and indexes.
Contents: v. 1. Introduction to quasicrystals—
v. 2. Introduction to the mathematics of quasi-
crystals— v. 3. Icosahedral structures.
1. Metal crystals. I. Jarić, Marko V.,
Date- . II. Gratias, Denis.
QD921.A67 1987 530.4'1 87-11527
ISBN 0-12-040602-0 (v. 2)

Printed in the United States of America
89 90 91 92 9 8 7 6 5 4 3 2 1

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Preface

Quasicrystals, whose diffraction patterns have non-crystallographic symmetries but, nonetheless, consist of sharp peaks, brought a dramatic change to our way of thinking about the structure of solids. This revolution required not only the development of new physical concepts, but it also demanded new mathematical tools. While the first volume of this series offered an introduction to the physics of quasicrystals, this companion volume is intended to provide mathematical concepts and results necessary for a quantitative description or analysis of quasicrystals. An effort has been made not to remain purely mathematical, but rather to emphasize the subjects of interest to physicists, crystallographers and metallurgists, focusing on the results and their possible ramifications, thus occasionally sacrificing the rigor of the proof.

The three mathematical areas most relevant to quasicrystals are the theory of almost periodic functions, the theory of aperiodic tilings, and group theory.

The theory of almost periodic functions, developed in the twenties and described in the classic book by Besicowich, is as important in the field of quasicrystals as the theory of periodic functions is in the field of crystals: It is its cornerstone. A practical dimension of this theory was recognised in the 1970s by theorists like DeWolf, Janner and Janssen who realized that an incommensurate crystal can be always identified with a cut through a higher dimensional crystal. For example, it is very easy to understand diffraction from incommensurate crystals in terms of the associated hypercrystals. This extremely useful point of view, equally applicable to quasicrystals (which can be considered as a special case of incommensurate crystals), was described in a chapter by Per Bak and Alan Goldman in the first volume of this series and is not extensively elaborated here.

Although at the time of the discovery of quasicrystals the theory of quasiperiodic functions had been known for nearly sixty years, it was the mathematics of aperiodic Penrose tilings, mostly developed by Nicolaas de Bruijn, that provided the major influence on the new field. For example, de Bruijn's "strip-projection" and "multigrid" methods, although not as general as the above mentioned "cut method," are widely used for quasicrystal description. Although quasicrystals, even if truly quasiperiodic, are generically not tilings, a reason for the importance of quasiperiodic tilings lies in the simplicity of the geometric "atomic" models they provide. Even before the discovery of quasicrystals, such geometric models were explored by Paul Steinhardt in connection with icosahedral and pentagonal order in metallic glasses and supercooled liquids. The tiling picture of quasicrystals, while approximate, is proving to be valuable in conceptual understanding of quasicrystal properties such as growth, low temperature excitations, and structural relationship with large unit-cell crystals.

Group theory, which traditionally provides an indispensible tool in studies of ordered structures and their defects, should also be of great importance in quasicrystals with high symmetry. Indeed, several years before the discovery of icosahedral quasicrystals, Peter Kramer invented a group theoretical method for constructing quasiperiodic tilings, like the Penrose tiling, and he used this method to construct a three-dimensional icosahedral tiling, which now provides a geometric conceptual model of icosahedral quasicrystals. Undoubtedly, group theory, which has already been used in the theories of elasticity, phase transitions, and defects in quasicrystals, will continue to play an important role in studies of quasicrystals.

In conclusion, let me emphasize that this volume is intended as a pedagogical introduction for the beginners in the field of quasicrystals. However, with its extensive collection of the results and bibliography, it can also serve as a reference for already experienced researchers working in this field. In particular, the material contained in this and the previous volume of the series provides an easy access to the next volume, devoted to various models of structures with icosahedral symmetry.

Chapter 1

A Brief Introduction to Tilings

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ISBN 0-12-040602-0

1 Tilings and Crystals

1.1 Introduction

No one knows when and where clay tiles were first designed for floors, stoves, and walls; probably they were developed in very ancient times, together with bowls, jugs and other earthenware. Equally ancient—perhaps more so—are the first three dimensional tiles, the familiar bricks. Their designers must have been the first to consider the problem of filling the plane, or a region of space, with congruent objects, a problem which continues to challenge designers today.

Also lost in the mists of time are the earliest musings of natural philosophers on the existence of atoms, a question which leads directly to the problem of designing tiles: if the universe is built from indivisible atomic units (and if there is no vacuum), then in order to fit together to fill space without gaps, these units have to have definite shapes.

In his dialogue *Timaeus*, Plato designated four of the five regular solids (Fig. 1), the regular hexahedron (the cube), octahedron, tetrahedron and

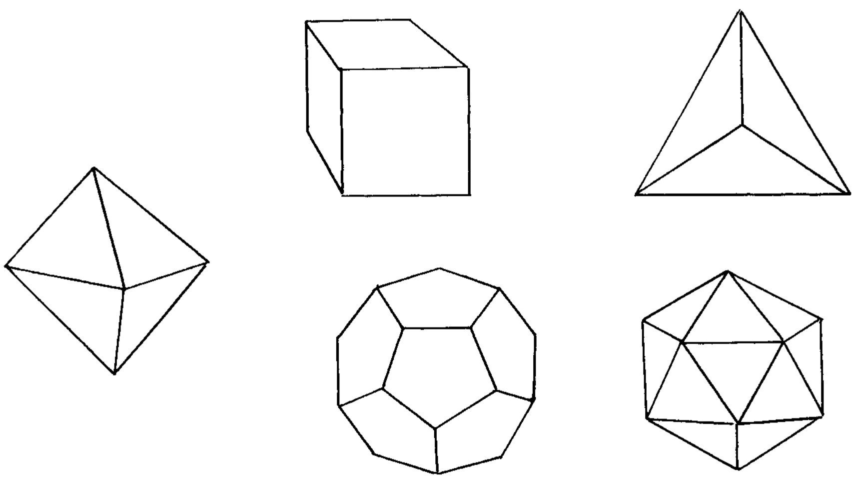

Figure 1. The five regular solids played a key role in Plato's cosmology.

icosahedron, to be the shapes of the ultimate particles of earth, air, fire and water, an association which captured the imaginations of artists and natural philosophers through the Renaissance, despite Aristotle's cogent refutation of this theory. Aristotle argued that "In general it is incorrect to give a form to each of the singular bodies, in the first place, because they will not succeed in filling the whole." It is clear that, although he did no calculations and made no models, Aristotle realized that some of the regular solids do not fill space. In fact, only the cube does; the others do not[1].

Eventually of course the vacuum was shown to exist, and today few people propose rigid tiling models for air, fire, or water. But tilings continues to be a useful way to study the solid state (Senechal, to appear). For over 150 years, since Haüy showed how various crystyal forms can be built from congruent bricks (Fig. 2), tilings have been used to illustrate geometrical properties of crystals such as the relation between internal crystal structure and external polyhedral form. Today the image of a pattern partitioned into congruent blocks is so central to our concept of crystal structure that it is difficult to imagine how it might be otherwise.

Thus tiling theory emerges from the confluence of two rich sources—the practical problem of designing useful and decorative tiles and bricks, and the scientific problem of modeling the internal structure of crystals. A third source—mathematical creativity—also plays a crucial role, that of organizing, simplifying, and generalizing basic concepts, devising classification schemes, and posing (and sometimes solving) new questions. For example, as the tiling model for crystal structures has been refined, challenging geometrical problems have arisen. Which polyhedra fill space? If a polyhedron does fill space, in how many different ways can it do so and what properties will the tilings have? What conditions can one place on a set of tiles to force nonperiodicity? Although we have partial answers to these questions, in many respects they remain unsolved.

The literature on tilings is voluminous, but until recently it has been almost inaccessible, spread among the journals of many fields (design, mathematics, crystallography, biology, etc.) and written in many languages. Moreover, it is often imprecise and frequently contains serious errors. Fortunately, 1987 saw the publication of a definitive treatment of tilings and patterns in the plane, *Tilings and Patterns*, by Branko Grünbaum and Geoffrey Shephard. (Hereafter we will refer to this book simply as G&S.) This 700 page monograph-textbook is a richly illustrated, rigorous yet very readable account of the state of the theory today. There is no comparable treatment of tilings and patterns in three dimensional space; indeed, there is no comparable theory! However a great deal of useful information about

[1] Aristotle thought that the regular tetrahedron was also a space filler.

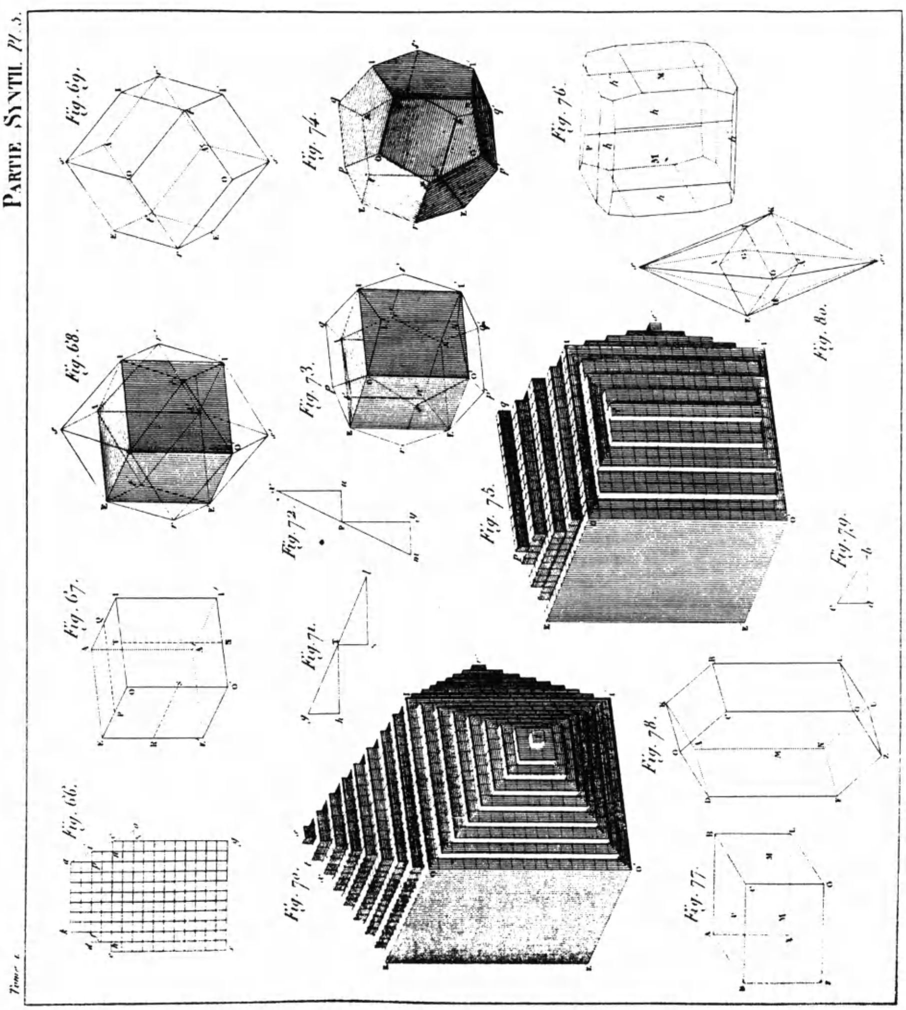

Figure 2. Hauy's concept of the relation between structure and form.

crystallographically significant tilings in n dimensions ($n \geq 2$) can be found in *Geometrical Crystallography* by Engel (1986). H. S. M. Coxeter's *Regular Polytopes* (1973) is also required reading.

The purpose of this chapter is to sketch those aspects of the theory of tilings in two and three dimensional space that are important for understanding some of the ways in which "classical" mathematical crystallography is being generalized to include possible models for aperiodic crystals. Although the aesthetic attractiveness of tilings suggests that the theory is accessible, it is not as easy as it looks. Unfortunately, the recent literature shows that many of its subtleties continue to be ignored. If the theory is to develop in a satisfactory way, investigators must be aware of what is and is not known about tilings, and why.

1.2 Analogies and Models

In Figs. 3, 4 and 5 we see some of the ways in which tilings are used today to illustrate particular aspects of crystal structure: patterns of crystal growth, twinning and structural defects. Underlying these and similar analogies is the concept of the structure of an ideal crystal as a periodic tiling of space by parallelopipedal "unit cells" which contain the translational repeat units of the atomic pattern of the crystal. Deviations from strict periodicity in these tilings are thought to be analogous to deviations from the ideal crystalline state.

In addition to providing visual analogies, tilings are also used as working

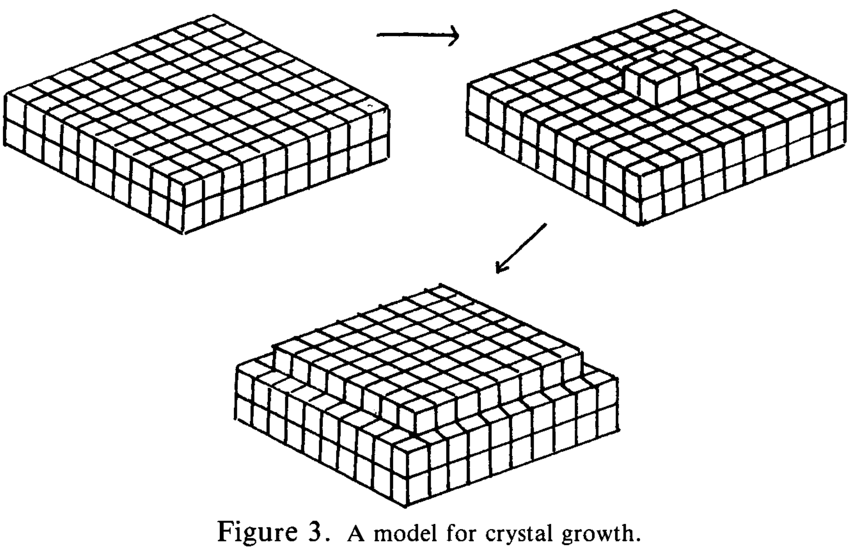

Figure 3. A model for crystal growth.

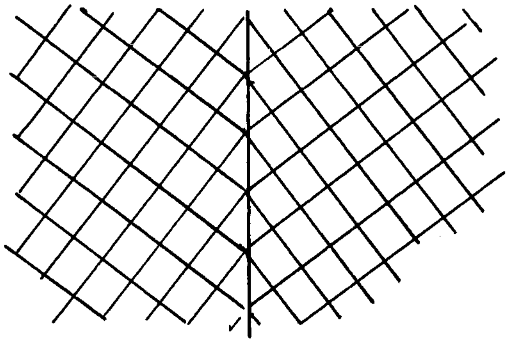

Figure 4. A tiling model for a twin boundary.

models for the exploration of crystallographic problems. In these cases the tiling is not only an illustration of a crystallographic idea; it is an alternative formulation of the idea. It is hoped that the solution of the tiling problem (if it can be solved), properly reinterpreted, will provide the solution to the crystallographic problem. For example, the Russian crystallographer E. S. Fedorov (1885), erroneously believing that crystal structures could be unambiguously partitioned into polyhedral cells, interpeted the problem of classifying crystals as the problem of determining which polyhedra fill space. Defining a *parallelohedron* to be a convex polyhedron which fills space when arranged face to face, in parallel position (all of these terms are defined in Section 2.1), Fedorov proved that there are exactly five parallelohedra (Fig. 23) and used them as the basis for his classification of crystals.

One hundred years later, tilings are again being used to model important crystallographic phenomena, but now we are in a new period in which the

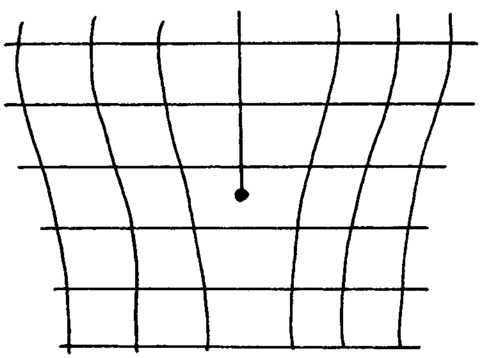

Figure 5. Model of a crystal dislocation.

necessity of the laws of periodicity associated with classical crystallography is being challenged. The announcement in 1984 of the discovery of "quasicrystals" was startling because the diffraction pattern of these crystals, with its five-fold symmetry, breaks the "crystallographic restriction" (Section 2.3) and thus challenges the crystallographic paradigm that the structure of a crystal is a repeating pattern like that envisioned by Haüy and Fedorov. Because certain nonperiodic tilings appear to have diffraction properties analogous to those of the quasicrystals, the discovery of quasicrystals has generated new research in tiling theory. A closely related problem has been discussed by C. Radin (to appear), who argues that "the crystal problem" (why are solids crystalline?) is equivalent to what he calls "the tiling problem": how is the periodicity of a tiling forced by matching rules?

2 Basic Concepts

2.1 Tiles and Tilings

A *tiling* is a partition of the plane or space by copies of objects which we call *tiles*. By "partition" we mean that every point belongs to at least one tile, and if a point belongs to more than one tile, then it must lie on the tiles' shared boundary.

This definition is widely accepted but subtleties arise when we try to define the word "tile." In fact, the word "tile" cannot be defined independently of "tiling" since there is, in general, no criterion for determining whether or not copies of a given set of objects fill the plane or space. On the other hand, although most shapes do not tile (here we are using the word as a verb) there are surprisingly many space-filling shapes besides the standard brick. Some are infinite, some are even toroidal (Fig. 6)! Thus when we are dealing with tiles in a specific context it is necessary to place restrictions on their sizes and shapes in order to make the problem of analyzing them more tractable. In most cases—and in this chapter unless stated otherwise—a *tile* is defined to be a finite region of the plane or space with a polygonal or polyhedral boundary that can be (continuously) deformed into a disk or a ball (accordingly as the tile is two or three dimensional)[2].

Let us recall some of the basic properties of finite polygons and polyhedra. A *polygon* is a finite sequence of line segments or *edges*, joined end to end to form a closed circuit which does not intersect itself. Each edge

[2] A disk (ball) is a circle (sphere) together with its interior.

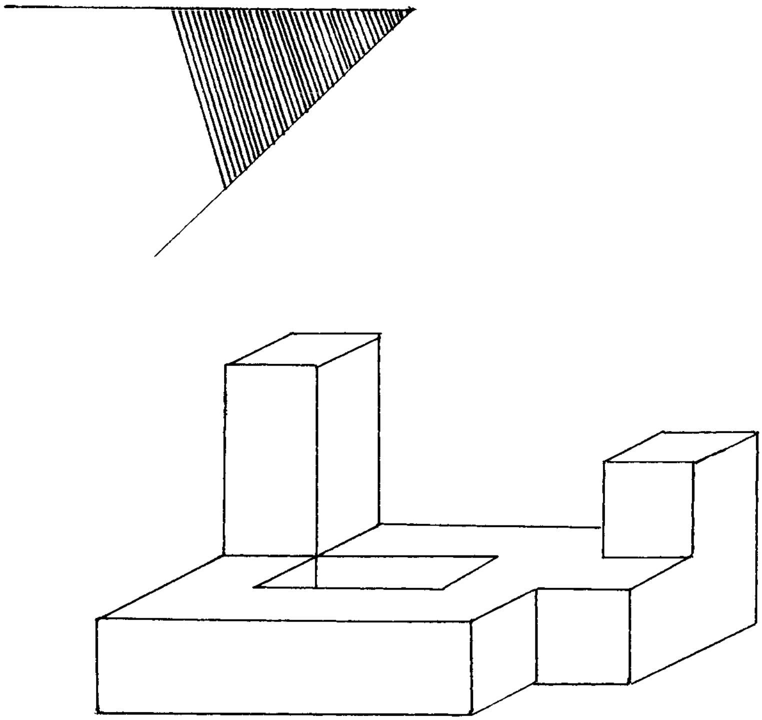

Figure 6. An object which tiles space need not be bounded or convex.

has two endpoints, which in turn belong to exactly two edges. The endpoints are the *vertices* of the polygon. The interior angle formed by two adjacent edges is called a polygonal angle. A *polyhedron* is the union of a finite number of plane polygonal regions, which are called the *faces* of the polyhedron. We require that the faces share whole edges (exactly two faces share each edge) and that it is possible to trace a path from any face to any other which does not pass through any vertex. We will assume that adjacent faces are not coplanar. Obviously, the edges and vertices of a polyhedron coincide with the edges and vertices of its faces. The number of edges meeting at a vertex v is called the *valence* of v. The convex hull of the midpoints of these edges is called the *vertex figure* of v. The interior angle formed by two faces is called a dihedral angle of the polyhedron. For example, a cube has six faces, all of which are squares. Adjacent faces always meet at right angles; thus all the dihedral angles of the cube are 90° Three faces meet at each of its eight vertices, which are thus 3-valent.

A region is *convex* if any two of its points can be joined by a line segment which lies entirely within it (the wedge in Fig. 6 is convex, the toroid is not).

Most of the tilings one encounters in crystallographic problems have convex tiles, but it should not be assumed that all important tilings have this property. Indeed, it is sometimes useful to deform convex tiles into non-convex ones in order to restrict the ways in which they can fit together. We will sometimes refer to the *convex hull* of a set of points; this is the smallest convex region containing them. For example, a convex polygon or polyhedron is the convex hull of its vertices.

The terms vertex, edge and face are also used in the description of tilings, but in a somewhat more subtle way. We define a vertex of a plane tiling to be a point where three or more tiles meet. The vertices and edges of the tiling may or may not be identical with those of the tiles: for example, in Fig. 7 the vertices of the tiling are not vertices of all of the tiles to which they belong. Similarly, a vertex of a three dimensional tiling is a point at which at least four tiles, not all sharing an edge, meet; such a point need not be a vertex of all of the tiles. Nor need the edges and faces of a three dimensional tiling always coincide with those of the constituent tiles. In the special case in which the tiles in a plane tiling share whole edges, the tiling is said to be *edge-to-edge*; similarly, if the tiles in a three dimensional tiling meet along whole faces, then the tiling is said to be *face-to-face*.

In most tilings of crystallographic interest, the tiles are all congruent copies of, or combinatorially equivalent to, a finite set of tiles, called *prototiles*. A tiling is said to be *k-hedral* if the number of prototiles is k. When $k = 1$, the tiling is *monohedral*.

A tiling is *normal* if the sizes of the tiles are uniformly bounded from above and below; by this we mean that there are circles or spheres of fixed radii d and D which can be, respectively, inscribed in and circumscribed

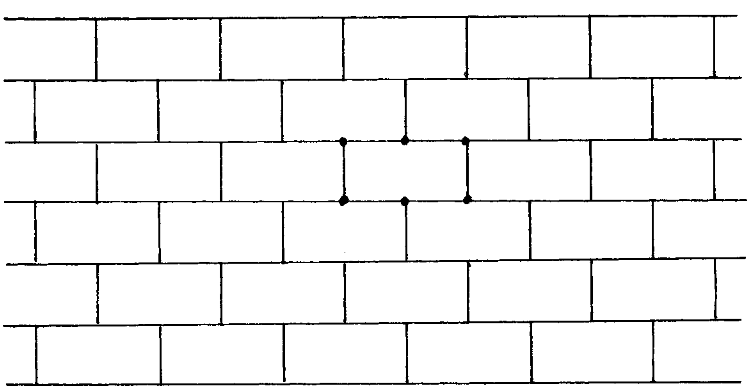

Figure 7. In this common tiling by rectangular bricks, six vertices *of the tiling* belong to each tile, although each tile, taken alone, has only four vertices.

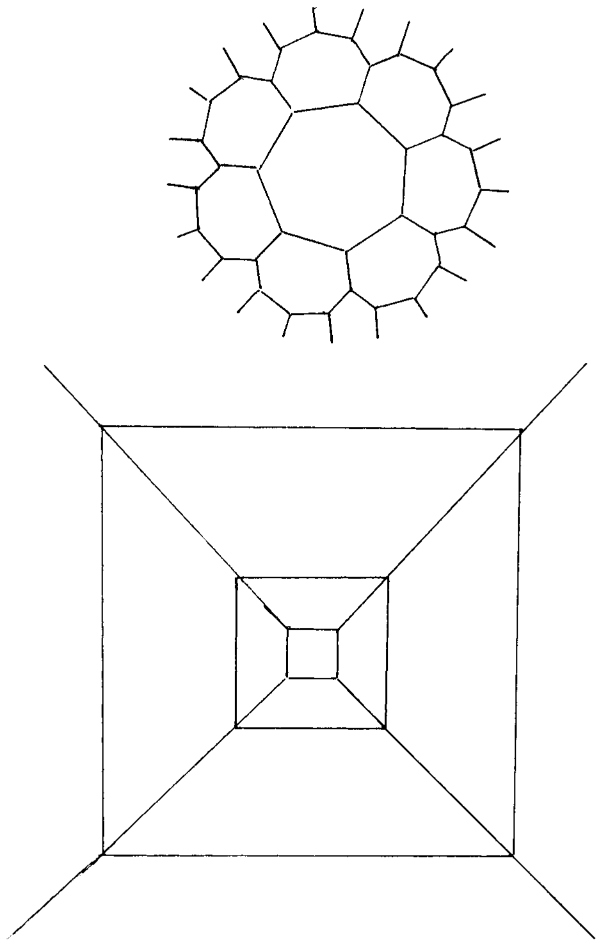

Figure 8. Two tilings which are not normal. In one, the heptagonal tiles become arbitrarily small; in the other, the quadrilateral tiles become arbitrarily large.

about each tile[3]. k-hedral tilings are necessarily normal if congruent copies are used. Two nonnormal tilings ae shown in Fig. 8.

Finally, following G&S we will say that two tilings are *congruent* if one can be brought into coincidence with the other (matching edges to edges and vertices to vertices) by a rigid motion (Section 2.2), while if coincidence

[3] In the literature, an edge-to-edge or face-to-face tiling is sometimes called a normal tiling, but we will always use "normal" as we have defined it here.

requires a change of scale, the tilings will be said to be *equal*. If coincidence requires any other kind of (continuous) deformation, then the tilings are *combinatorially equivalent*[4]. Congruent tilings are of course equal, and equal tilings are also combinatorially equivalent.

2.2 Symmetry

While we all have intuitive ideas of the meaning of terms like "highly symmetrical," "asymmetrical," and so forth, the mathematical study of symmetry is based on careful definitions.

The symmetry of an object is characterized by the symmetry operations that can be performed on it. A symmetry operation or, briefly, a *symmetry*, is a rigid motion, one which does not alter the distances between any pair of points in the plane or in space. Rotations and reflections are examples of symmetries.

In any dimension, the number of different kinds of rigid motions is finite. A simple fixed-point argument shows that in the plane, if three noncolinear points are fixed then all points are fixed and hence the motion is the identity (no motion at all); if two points are fixed so are all points of the line on which they lie and the motion is reflection cross that line; if exactly one point is fixed then the motion is a rotation; and translation and glide reflection are the isometries which have no fixed points. In space, three coplanar, noncolinear fixed points imply that the motion is a reflection in a plane; two, that it is a rotation about an axis; and one, inversion or rotary inversion. In addition to translation and glide reflection, screw rotation (a motion combining rotation and translation parallel to the rotation axis) also has no fixed points. Only the identity has four fixed points which are not coplanar.

An object is said to have a given symmetry if the corresponding operation brings the object into self-confidence. Notice that if we can perform a symmetry operation on an object we can also reverse it; thus for each symmetry we also have an inverse symmetry. The set of all symmetries of a given object form a group in the technical mathematical sense if we include the identity, because then the composition (successive application) of two symmetries is always again a symmetry. Not surprisingly, this group is called the symmetry group of the object. It is a very instructive exercise to find the symmetries of the regular solids (Fig. 1) and of the regular tilings of the plane (Fig. 31), and to determine the results of various compositions.

[4] Combinatorially equivalent tilings are sometimes said to be isomorphic.

The subunits of a pattern (faces of a polyhedron, tiles of a tiling, points of a point set) which can be brought into coincidence with one another by the operations of a symmetry group are said to be *symmetrically equivalent*. Equivalence in this sense is stronger than congruence: the faces of a cube are equivalent not only because they are congruent squares, but also because any one of them can be moved, by a symmetry operation of the cube, to the position of any other in such a way that the cube appears not to have been moved at all. The symmetry group is said to *act transitively* on the equivalent subunits; we also say that the subunits are a transitive set.

A polyhedron or tiling on whose faces or tiles a symmetry group acts transitively is said to be *isohedral*. If group acts transitively on the vertices of a polyhedron or tiling, then it is *isogonal*, and if its edges are equivalent it is *isotoxal*. (Apparently no word has been coined yet for equivalent two dimensional faces in a three dimensional tiling!)

The regular polyhedra are isohedral, isotoxal and isogonal. Their face centers, edge midpoints and vertices lie on three concentric spheres. These polyhedra are the convex hulls of their vertices, which lie on the outermost sphere. The convex hulls of their face centers, which lie on the innermost sphere, are also regular polyhedra. The convex hull of the edge midpoints of a regular polyhedron is not necessarily regular, but it is interesting nevertheless, since it is always isogonal and has the same symmetry group as regular polyhedron from which it is derived. For example, the convex hull of the edge midpoints of the cube (and of the octahedron) is a cuboctahedron, an isogonal polyhedron with six square and eight triangular faces, while the convex hull of the edge midpoints of the icosahedron (and dodecahedron) is an isogonal icosidodecahedron, which has twelve pentagonal and twenty triangular faces (Fig. 9).

The symmetry group of any polyhedron is finite. Finite symmetry groups are sometimes called *point groups*; they are subgroups of the symmetry group of the sphere. These finite groups fall into four classes: the symmetry

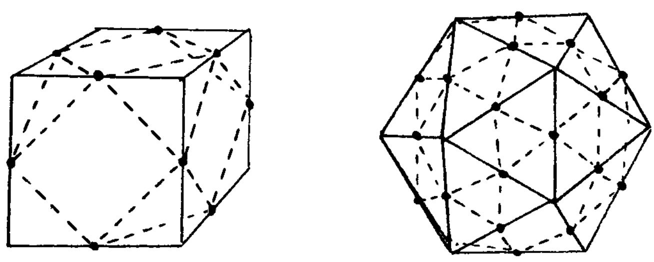

Figure 9. The isogonal cuboctahedron and icosidodecahedron (broken lines) are the convex hulls of the edge midpoints of the cube and the icosahedron.

group of the cube (and regular octahedron) and its subgroups, the symmetry group of the regular icosahedron (and dodecahedron) and its subgroups, and the symmetry groups of right k-gonal prisms and of pyramids with k-gonal bases. There is considerable overlapping of subgroups. The symmetry groups of tilings are more complicated, since they are infinite. A good introductory discussion can be found in G&S.

2.3 Lattices and the Crystallographic Restriction

An infinite set of points equally spaced along a line is a *one dimensional lattice*. If all of these points are translated in the plane by a vector which is not parallel to the line, we obtain a copy of the lattice which is parallel to it; if this translation is repeated ad infinitum (in both positive and negative directions) we obtain a *two dimensional lattice*. Similarly, a *three dimensional lattice* is obtained by successive translations of a two dimensional one. Since the time of Haüy, the lattice has been the fundamental concept underlying theories of crystal structure.

It is sometimes useful to think of an n-dimensional lattice (n may be any positive integer, including two or three) as the set of endpoints of sums and differences of n independent translation vectors $x_1, \ldots, x_n$. Then every lattice point is associated with a unique vector $a_1x_1 + \cdots + a_nx_n$, where the coefficients a_i are integers. The vectors x_i are called a basis for the lattice. The basis vectors are not unique; there are infinitely many sets of n vectors which generate the same lattice.

A set is *discrete* if there is a minimum distance between its points. Since the shortest vector in a lattice defines a minimum distance, a lattice is thus a discrete, translation-equivalent set of points. Because the points of a lattice are equally spaced along any lattice row, every lattice point is a center of inversion for the lattice as a whole, and so is the midpoint between any pair of lattice points. The least symmetric lattices have no other symmetries.

Crystallographers classify lattices into families according to the symmetries which fix (any of) their lattice points and according to the way in which this symmetry group acts on the lattice. The symmetry of lattices is governed by geometrical laws. In particular, a lattice can have m-fold rotational symmetry only if $m = 1, 2, 3, 4$ or 6. This result is known as the *crystallographic restriction*.

A simple geometric argument shows why five-fold symmetry is incompatible with a planar or three dimensional lattice. If a lattice had five-fold symmetry, then the set of points about which this rotation takes place must, by translational symmetry, include a lattice. Assume, then, that the points of a plane lattice have five-fold symmetry, and that P is such a point. In any lattice, there is a minimum distance between points; let Q be a lattice point at minimum distance from P. By hypothesis, P is surrounded by five such Q's and Q is surrounded by five such P's. We see (Fig. 9) that this contradicts the assumption that the distance between P and Q is minimal. The conclusion holds for three dimensional lattices too, since a rotation about an axis effects rotations in the lattice planes normal to the axis.

It follows that if a crystal has a periodic structure, it cannot have five-fold symmetry (Fig. 10). Even its external form cannot be a regular icosahedron (or dodecahedron), since a crystal's form cannot have a symmetry forbidden by its internal structure (notice that the dodecahedron in Fig. 2 is not regular). Equivalently, if a crystal exhibits five-fold symmetry then its structure is not periodic.

It is even easier to show that a two or three dimensional lattice cannot have rotational symmetry of order greater than six. Again we consider a plane lattice whose points are centers of rotation, say of order m. Let P be a lattice point and Q a lattice point at minimum distance d from P. Rotating about P, we come to a lattice point Q', also at distance d from P, such that measure of angle QPQ' is $360/m$. But if $m > 6$, the distance from Q to Q' is less than d. This concludes the proof of the crystallographic restriction. Although the argument does not prove that lattices with two-, three-, four- or six-fold symmetry exist, another simple argument shows that successive m-fold rotations ($m = 3, 4$ or 6) about two points generates a lattice with four- or six-fold (and thus two- or three-fold) symmetry.

The crystallographic restriction is dimension dependent (Pleasants, 1985) because the list of possible symmetry operations is. In four- and all higher

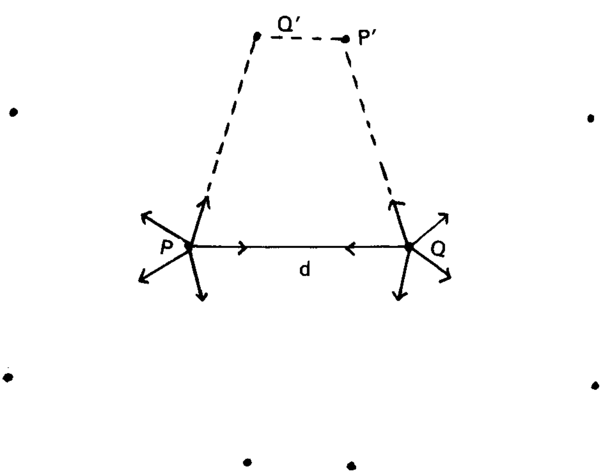

Figure 10. A lattice cannot have points of 5-fold symmetry.