ORDERLY DISPOSITIONS IN SPACE

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A report on the workshop

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I. <u>Introduction</u>. This workshop brought together a group of mathematicians, physicists, biologists, chemists, and crystallographers whose work involves the study of orderly arrangements of identical objects in space. In many cases, these order properties do not include three dimensional periodicity and cannot be adequately characterized by automorphism groups. The purpose of the workshop was to explore new methods — not limited to group theory — for generating and classifying such patterns.

In the last few years the need for methods to deal with more general arrangements has been increasingly felt. In particular, the discovery of quasicrystals has given special impetus to the study of aperiodic tilings and patterns. When this is taken together with recent investigations into the structure of glasses, the structure of incommensurate crystals, the patterns of molecules in liquid crystals, the geometry of viruses and other biological structures, and other areas of research in geometrical pattern formation, it is clear that a theory of nonperiodic structures which utilizes both local and global information is badly needed.

II. <u>Summary of the lectures</u>. The first two lectures were introductory, intended for workshop participants who were not specialists in the field. In the first lecture, Branko Grünbaum discussed recent developments in tiling theory. In the first place, the theory is now mathematically sound. Proper definitions have been supplied for key concepts, "facts" that had long been taken for granted have been rigorously proved — or disproved! — and intuitive classification schemes have been replaced by well-defined ones. In the process, many new questions have arisen. For example, in most tilings of interest, the tiles are copies of a finite number of tiles, called prototiles. Often, more than one tiling can be constructed with copies of the same set, and there are many open problems concerning the number and nature of the different tilings. Of particular interest are aperiodic tilings, tilings in which the tiles are copies of a set of aperiodic prototiles. (A set of prototiles is aperiodic if every tiling constructed with their copies is necessarily nonperiodic.) The Penrose thick and thin rhombs with oriented edges are the simplest examples of aperiodic prototiles, but others have been constructed by Ammann, and also, earlier, by Robinson and others investigating Wang tiles, squares with colored edges which are juxtaposed so that the colors match. Aperiodic tilings form only a tiny subclass of nonperiodic tilings but they may be especially helpful in understanding nonperiodic physical systems, since in all known examples, their nonperiodicity is forced by matching rules which are analogous to bonding properties. Further, in all known examples the matching rules for aperiodic prototiles ensure that they can be grouped together, in only one way, to form larger copies of themselves. Grünbaum asked whether this inflation, or self-similarity, property is intrinsic to aperiodicity. He concluded the lecture with illustrations of some of the subtleties of tilings of the plane, and posed some problems concerning them.

In the second lecture, Marjorie Senechal reviewed the historical development of the crystallographic paradigm, which holds that by definition a crystal is a solid whose atomic pattern is a three dimensional periodic structure. This paradigm was triply reinforced by the success of group theory in enumerating the crystallographic groups in 1891, the discovery of x-ray diffraction by crystals in 1912, and the universal and timeless aesthetic appeal of symmetrical designs. Two important mathematical consequences of periodicity are that the rotational symmetry of a crystal can only be 2-,3-,4- or 6-fold, and that a point pattern representing the structure can be described as a union of a finite number of congruent point lattices. However in recent years the paradigm has eroded considerably, largely due to the discovery of crystals whose structures are unions of incommensurate lattices, and of crystals whose diffraction patterns indicate the presence of 5-fold symmetry. She reviewed some of the geometrical models that are being used to generate patterns with "forbidden" symmetries, and concluded with a list of open questions.

The next lecture was given by Charles Radin, whose recent research has focussed on the problem of why crystals are symmetrical — if they are. Radin discussed the "Weyl conjecture", which asserts that arrangements of minimum energy should be periodic. For example, in the densest packing of equal spheres in three dimensional space the sphere centers should form a lattice (this is part of Hilbert's 18th problem, and remains unsolved). More generally, consider a set of interacting particles arranged in space. We want to minimize the energy of any arrangement in any box. Radin and his colleagues have shown, however, that minimizers are (generically) *not* periodic. The result can be illustrated by sphere packings, Wang tiles, and other models.

(Later, in a problem session, Enrico Bombieri presented arguments which suggest that densest packings and thinnest coverings by sets in high dimensions will also not be periodic).

Several lectures dealt with attempts to model specific physical systems.

John Cahn discussed his continuing efforts, together with several colleagues, to determine the structure of the icosahedral ("quasicrystalline") phase of $Al_{73}Mn_{21}Si_6$. The observed rotational symmetries of their diffraction patterns are 5-fold, 3-fold, and 2-fold, suggesting that the three dimensional symmetry is icosahedral. One model that has been used to describe the structure is a decoration of the rhombohedral cells of a three dimensional Penrose pattern, but this does not fit the data sufficiently well. Another approach has been to treat these crystals as if they were glasses, but this does not explain enough either. (For a detailed discussion of these models, see the recent article by LaBrecque.) Alternatively, Cahn proposed, we can go back to the fundamentals of crystallography and be completely empirical. Computing the Patterson (autocorrelation) function in three dimensions one obtains a quasiperiodic Patterson which, because six numbers are needed to index the patterns, can be unfolded to 6 dimensions. The six-dimensional structure is periodic, an analogue of the three-dimensional cesium chloride crystal structure. The Patterson superimposes, in small regions, on that of the crystalline phase of $Al_{73}Si_{11}Mn_{16}$, a cubic crystal with 138 atoms in the unit cell. This suggests that the packing in the quasicrystalline and cubic phases are closely related. But they are not identical over larger regions: as you move the quasiperiodic Patterson around, you get substitutions: the first shells look like Mackay icosahedra but as you go out, Mn substitutes for Al, and so forth.

Louis Michel discussed the "zoology" of liquid crystals. A crystal, whatever it is, has long range correlations. In liquid crystals, the correlation is only in orientation, not position. The molecules are usually long or disk-like. For example, nematic liquid crystals have elongated molecules (e.g., ellipsoids) which are parallel, in cholesterics the molecules have electric dipoles and the symmetry is helicoidal, while discrete translations occur in smectics. Explaining that the job of the theoretician is classification, to provide a framework for experimentalists, Michel described the group-theoretic classification of liquid crystals (quotient groups of crystallographic groups). He pointed out that among the many problems awaiting the attention of mathematicians is the description of defects in liquid crystal structure.

Donald Caspar discussed the geometry of viruses. Some viruses are columnar. For example, in sickle-cell hemoglobin, the heme molecules aggregate into long helices which cannot move through capillaries. The helices aggregate to form interlocked helices or "twisted crystals". Any helical structure can be described by a surface lattice; it becomes geometrically interesting when the lattice is perturbed. In biology there are all sorts of imperfect or "quasi" helices, in which the units may be closer together or further apart than on the "perfect" unrolled pattern. (For example, there is a sinusoidal perturbation in tobacco mosaic virus). The helices may be made from nonregular tilings, and the molecules at the vertices can switch bonding properties. In addition to the columnar viruses, there are polyhedral ones. For example, the tomato bushy stunt virus has icosahedral symmetry. Twenty years ago Caspar and Klug proposed a geometrical model for icosahedral viruses, based on the geodesic dome, that incorporated the essential property of self-assembly. This model predicted that six-coordinated units would be hexamers, and fivecoordinated units would be pentamers. Recently, however, Caspar and his colleagues have discovered that all the units are pentamers! The reasons for this, and how the bonding works, are not yet understood. It appears to be more a problem of molecular biology than of geometry.

Another set of lectures concerned properties of aperiodic tilings, in particular the Penrose tilings. In 1981, N.G. de Bruijn published a global derivation of the Penrose tilings, which he obtained as special duals of pentagrids (five superimposed grids, or infinite families of equispaced lines). In one class of these duals (defined by the displacements of the grids from the origin), the edges of the tiles can be labeled so that the original Penrose matching rules are recovered. The grid method can be generalized to produce tilings with arbitary symmetry, but unless adequate matching rules can be found for the prototiles produced by these grids, there is no guarantee that these prototiles enforce tilings which are grid-produced, or enfore tilings with any other kind of long range order." A tiling whose vertices lie at points of a de Bruijn dual of an m-grid can also be obtained by projection from a lattice in m-dimensional space. This "cut-and-project" method gives important global information about the tiling; in particular one can compute the diffraction pattern of the tile vertices.

De Bruijn gave two closely related lectures. In the first, intended for a general audience, he showed that the labeling problem is similar to the "riffle-shuffle" card trick, first presented by a magician, Norman Gilbreath, and later discussed by Martin Gardner in Scientific American. A deck of cards is arranged so that red and black cards alternate. The deck is cut, and the riffle shuffle performed. Then the deck is cut again, this time so that the cut is made between two cards of the same color. Now the performer deals out pairs of cards. Lo and behold, each contains a red card and a black one! The "trick" can be analyzed by means of a finite graph with directed edges, or automaton; the riffle shuffle defines a directed path through the graph. De Bruijn then showed that in a pentagrid, a single line in one of the five grids is cut by lines of the other four grids in a sequence that can be described by a riffle shuffle with an infinite deck of cards, and which can be described as an infinite path through a finite automaton. The automaton prescribes arrowing for the edges of the rhombus tiling. In general, this procedure gives two types of thick rhombs and two types of thin, but under the shift conditions mentioned above there is only one of each type. The procedure can be extended to the three dimensional analogue of the Penrose tiles, providing them with markings which ensure nonperiodicity in every one-dimensional stack of tiles. In the second lecture, de Bruijn showed how the automata can be used to generate Penrose tilings of the plane, and also their one dimensional analogues.

Andre Katz announced the discovery of matching rules for the three dimensional Penrose tiles which ensure nonperiodicity. These rules can be presented as decorations on the faces of the tiles, just as in the two-dimensional case they can be presented as decorations of the edges of the rhombs. There are fourteen different decorations for the thick rhomohedra and eight for the thin ones. Katz's approach is based on the cut-and-project method of obtaining the three dimensional Penrose tiles. From this point of view, the vertices of the tilings are the projections into three dimensional space of the points in a strip in the six-dimensional cubic lattice. The strip is defined to be the set of points in the lattice which project, in the complementary three-space, into a triacontahedron (which is the projection of a unit six-cube). The triacontahedron can be tiled by copies of the two rhombohedra in 120 different ways. A careful analysis of the packings which are consistent with icosahedral symmetry shows that the actual number that occur in this case is far less than 120. Katz was able to discover decorations which permit only those configurations of rhombohedra which do occur, and proved that the corresponding tiling of three space has no gaps or overlaps. This is not only the first example of a bona fide set of three dimensional aperiodic prototiles, it is also the only example of matching rules for which there is (evidently) no associated inflation rule.

Peter Pleasants presented a general method for deriving a class of one dimensional nonperiodic tilings, called prism patterns. The vertices of these tilings are obtained as cross sections of an infinite prism in R^n. Prism patterns with inflation can be constructed for any preassigned multiplier which is a Pisot-Vijayaraghavan number; the multiplier is an eigenvalue of the matrix representing the inflation rule. Prism patterns can be used to constuct tilings in any dimension which are invariant under a finite noncrystallographic symmetry group G; it is only necessary that the inflation multiplier generates the field containing the cosines of the angles between the symmetry directions. In general it is not possible to find matching rules for the tilings because the inflation of copies of a single shape may be realized in different ways in different parts of the tiling. However, if G has no large crystallographic subgroup, then matching rules preventing periodicity can be assigned to the prototiles.

Peter Engel spoke on conditions which force periodicity in tilings. A necessary condition for a tiling to be transitive is that the tiling is monohedral, that is, all the tiles are congruent. This is, of course, not sufficient. On the other hand, if the tiles in a monohedral tilings are surrounded by all the other tiles in exactly the same way, then the tiling is transitive: any tile can be mapped to any other by an isometry which maps the entire tiling onto itself. About ten years ago Delone and his colleagues showed that for a discrete, homogeneous (relatively dense) point set transitivity is ensured if each point is surrounded identically by all the other points of the set *within a finite radius*, for which they gave an upper bound. But how small is this radius? We can translate this into a tiling problem by considering the Dirichlet domains of the points of the set. It has been shown that in the plane, it is sufficient to require the congruence of the tiles and also of their first coronas (the first corona of a tile is the tile together with the tiles which meet it). However, in three dimensions, the congruence of the first coronas is not enough; Engel has found several examples of distinct tilings in which the first coronas are the same. In these cases, however, the second coronas are different. Engel conjectured that in all dimensions greater than two, the congruence of second coronas is sufficient to ensure transitivity.

The remaining two lecturers presented conceptual frameworks for quasicrystallographic patterns and disordered structures.

Aloysio Janner showed that it is possible to regard quasicrystal structure as a special case of incommensurate crystal structure, if we adopt the working definition of an n-dimensional quasicrystal as a projection of a slice of an (n+d)-dimensional lattice. In embedding the quasicrystal in the higher dimensional lattice, the choice of lattice type is not unique. Only the (1+1)-dimensional case has been worked out in complete detail, but already the results are intriguing. In particular, it turns out that not only can the quasicrystal be embedded in a Euclidean plane, it can also be embedded in a plane with an indefinite metric. The self-similarity property of quasicrystals has a simple explanation when seen in this light.

Nick Rivier discussed his ongoing work with J.F. Sadoc and others on the structure of glasses and close-packed compounds. They derive disordered structures by mapping 3-polytopes from curved into flat 3-space. The process, called decurving, consists of iterated decorations of the simplices of the polytope, and results in the introduction of disclinations. Disorder is introduced by local commutations in the sequence of decurving operations. The decurving of {3,3,5} was discussed in detail. This polytope has 120 vertices, all of them 12-connected. At each stage of the decurving process, some of the 12-coordinated points are transformed to 14- or 16-coordinated ones, which form the vertices of disclination networks. The average coordination number approaches 13.333..., in both ordered and disordered structures, a value approached within 10% by all 24 known crystalline tetrahedrally close-packed structures.

III. <u>Discussions</u>. Three discussion sessions were scheduled during the workshop, and many others materialized during and after lectures. The following notes make no attempt at completeness, but it is hoped that they indicate some of the central themes of these discussions.

1. *What do we mean by order?* Several people suggested that a quasicrystal should be defined to be a structure projected from a slice of a higher dimensional lattice onto an irrational subspace. This ensures a discrete Fourier transform, i.e., a nice diffraction pattern. But such a definition may be too restrictive. Is there any important reason to distinguish between patterns which have nice diffraction patterns and those that don't? There was a general consensus that long-range order is significant, but how is it to be defined? In some way, local configurations should carry information about some larger area. It was pointed out that an algorithmic definition of order (such as inflation?) is not helpful: if you are given a Turing machine that cranks out primes, do you know anything more about primes? It was suggested that long-range correlations should be studied; this has not been done even for the Penrose tiles.

2. *What kind of order are we looking for?* Andreas Dress, who generously gave up the time scheduled for his lecture so that there could be another extended discussion, suggested that we should look for topological, rather than metric order. As an example of the relation between local and global structures, he pointed out that in the theory of coverings the topology of the mapping procedure has a lot to do with the possible metrics that can be imposed. For example, it is topology that controls the possible coverings of the sphere by the torus. We need a discretized theory of coverings, and this exists in the work of Coxeter and Tits. He discussed some of the ways that their work could be used to study tilings, including nonperiodic ones. Even matching rules can be studied this way. Other suggestions were also put forward: one might measure the order of a point set by the properties preserved under various transformations (including duality), or the order in a tiling by the number of noncongruent second coronas. On the other hand, it was pointed out that following any one of these approaches might prejudge the situation. Instead of concentrating on point sets or tilings, maybe the objects we study shouldn't even be assumed to be rigid. The very success of crystallography may be a source of our conceptual difficulties. It was agreed that we are now in a period with a lot of ideas and a lot of disorder; we are still searching for the right questions and the right definitions.

3. *Where do quasicrystallographic patterns come from?* The embedding of a discrete nonperiodic structure in higher dimensional space is, as Janner had pointed out, not unique. Per Bak showed that not only might different lattices accomodate it; so could a sequence of surfaces in higher-dimensional space. It was argued that for physical reasons these surfaces should not have discontinuities. This led to a discussion of the kinds of topologies that would be consistent with icosahedral symmetry. Unfortunately it is possible for a structure to have a Fourier density which converges on the cut, but which is itself nonmeasurable. Bak gave an example in which there is a transition to chaos. Mathematicians were urged to consider these analogies.

4. Although it was not explictly discussed during the workshop as a question to be investigated for its own sake, in reviewing notes, xeroxes of transparencies, and preprints before writing this report it was striking how often the concept of self-similarity, or inflation, was invoked in one form or another in lectures and discussions during the week. While it now appears that inflation is not necessarily a feature of either aperiodic prototiles or cut-and-project quasiperiodic point sets, it continues to play an important role in research in these areas, as well as in fractal geometry. Yet there does not appear to be any general agreement about what is meant by inflation, what its properties are, and what it implies for patterns which possess it.

The spirit of the workshop can be summarized by saying that it was a lively one, in which the participants taught, learned from, and argued with one another. We hope that the lectures and the discussions will provide a useful basis for further research.

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