GENERALISED CRYSTALLOGRAPHY[1]

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In our preoccupation with the detailed problems of our daily work of finding out how atoms are arranged in space, we are in danger of losing sight of the whole picture and of not seeing what the picture shows, nor what it means, nor where our part fits in. In the art world there is a place for the critic. A case has been made that there is also scope for the science critic, and you will indeed see stimulating science criticism, particularly in the pages of Nature, Science and the New Scientist. I think that quite inadequate attention has been paid to the philosophy, ideology and general pattern of our subject, and this is what I want to discuss, bearing in mind that the division of nature and learning into subjects has no natural basis, and is only a concession to human weakness and a convenience for bureaucracy There is, I believe, a genuine crisis of identity in the field of crystallography, which has followed on the success of the modern techniques of structure determination. Like the art critic, I want to attack the traditional beliefs and to support the view that a science which hesitates to forget its founders is lost. I will therefore not mention von Laue, Bragg or Bernal, but immediately commit the blasphemy of suggesting that crystallography is only incidentally concerned with crystals and that its real objectives today must be different. Our objectives must be to understand regular structures.

Our subject is called crystallography purely for historical reasons. But it has taken a great struggle to get it institutionalised and it would be foolish to throw away the name under which we have worked for more than fifty years. Our problem is the eternal one of the relationship between form and content in a particular field.[2]

Crystals form an extremely important reference point or image or concept in our culture and I would like to trace their significance in intellectual history but I must stick to the present argument. However the predominant image, throughout most cultures, is that a crystal is a symbol of purity. W e see this in the Bible, in the Greek philosophers, in the Middle Ages and so on up to the present day where it persists. However the significance of .this mystical view is that it happens to have a firm material basis - crystals are pure. A crystal is the repetition of identical units. The crystalline lens of the eye, our window on to the world, is transparent because its spatial Fourier transform contains no terms with wavelengths in the visible range of 4000 - 8000 A. The working chemist's

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[1] Based upon the general lecture presented at the 10th Conference of the Yugoslav Centre of Crystallography held in Kumrovec, 5th to 7th June, 1975.

[2] The actual word 'crystal' comes, of course, from the Greek word for quartz, which was 'krystallos' and indicated the belief that quartz was just ice which had been petrified by extreme cold. The word 'crystallography' surfaced first in Latin in the »Prodromus Crystallographiae« of M. A. Cappeller (Lucerne, 1723) and in the »Essai de Cristallographie« by Rome de Lisle (1772). Already in 1717, Diderot's Encyclopedia carried substantial articles on crystals as a branch of physical science. If asked to pinpoint the actual moment when the alchemical and mystical view of crystals passed into science, we might mention the book »Elements de la Philosophie et de l' Art du Feu ou Chemie« (Paris; 1641) by William Davidson (1593-1669) who was an Aberdonian and the first British professor of chemistry (albeit in Paris). From the diagrams in his book he clearly thought; regaining the position which the ancient Greeks had reached, that there was some structural connection between the Platonic solids (regular and semi-regular) and the shapes of crystals, but he could not quite cross the bridge. This was left for Hauy (1743-1822).

test for purity is that his product crystallises and only an assembly of identical subunits will do this.[3]

Anyway, although the traditional pre-occupation of crystallography is with crystals, first mineral and then organic and synthetic, it is changing rapidly and there is some organisational flux where crystallographers are tending to leave the central societies dealing with crystals and to adhere more to other societies more concerned with the behaviour of the components which happen to crystallise. I would like to make it a serious aim to restore a central core to crystallography and to show what its new-style central subject matter actually is. I want to claim, thinking of the future, that. crystallography is rapidly becoming the science of structure at a particular level of organisation, being concerned with structures bigger than those represented by simple atoms but smaller than those of, for example, the bacteriophage. It deals with form and function at those levels, particularly with the way in which large-scale form is the expression of local force.[4]

Crystals are just the forms of the simplest ordered structures and we ourselves, perhaps, are the forms of the most complex. Ordered structures go against the general current towards randomness and disorder by using the fluxes of energy through the system. Just as a sailing ship can beat to windward against the prevailing wind by using the wind itself, so an organism can use the flow towards disorder to keep itself ordered. Life depends on the directional stream of energy. It could not operate as a Maxwell demon and obtain its energy from the random thermal motions in an isothermal enclosure. My theme is that a consideration of crystals should lead us to consider other types of ordered structures. Even in an isolated system, where the entropy cannot decrease, the production of crystals with an apparent local reduction of entropy is more than compensated by the more than equal increase in entropy of the rest of the system. You will recall Schroedinger's aphorism that living systems feed on negative entropy, that is on ordered structures, and excrete the residue in greater disorder, metabolism implying catabolism, thus maintaining themselves organised against the prevailing stream flowing towards the isothermal dead sea. When an organism can no longer metabolise and interact with its environment, it dies.

Moving thus from the study of crystals to that of other structures, we may ask what kind of general properties we may look for. The most usual characteristic, which makes both the evolution of the structure and our study of it possible, is that of hierarchy. Simply, in any large structure we can

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[3] Zone refining, used by Bernal in the 1930s for purifying organic materials, but only reported by word of mouth, is only the last of a long line of procedures, followed since the earliest times, partly technology and partly sacrament. which involved passing materials through the fire (cupellation); through the water (crystallisation from solution) or through the air (sublimation) to obtain pure crystalline products. It was the occurrence of the ores of copper, tin, lead and other metals and non-metals (borax, nitre and sal-ammoniac) as crystals which enabled early technologists to recognise and to concentrate them for smelting to give the metals for making the tools which are the basis of our civilisation. All kinds of chemical and biochemical processes followed and were diffused. In the India of the sixth century portrayed by Vatsayana in his Kama Sutra; one of the arts which a courtesan had to learn was mineralogy. This was for the very sound materialist reason that if a girl were to be paid in precious stones it was important that she should be able to distinguish real crystals from paste.

Very much of the apparently mystical literature in many cultures is not, as might appear on first examination, about religion or sex, but about these actual chemical operations. It is not about Gods and Saints, but about, for example the phenomenology of mercury and sulphur. It is just that the present-day scientific concepts and vocabulary had not then separated from those of religion. Curiously perhaps, this separation finally happened at the time of the French Revolution, when the ideas of church and state also separated.

[4] The word 'form' has a very long history from the idealism of Plato to the present, where the French word 'fromage' and the Italian 'formaggio' both meaning cheese, derive from 'form'. Molten cheese may be poured into a mould or form which gives it shape by providing boundary conditions for the transformation to the solid state. Kepler's 'facultas formatrix' (1611) - the power which gives things shape - was an immensely important concept (J. Kepler »The Six-Cornered Snowflake«, Oxford University Press, 1966). A more up-to-date treatment can be seen in the standing waves which are the solution of the wave equations with certain fixed boundary conditions, but I want to examine other kinds of morphogenetic processes also. Etymology is, I would claim, not just a matter of pedantry but, as 1 will try to explain below, a kind of intellectual archaeology where we can see how, like material artifacts, words were refined with the passage of time and successive transmutations.

distinguish smaller sub-structures in which the internal relations are stronger than the external relations binding the sub-structures together. A complex structure can thus (usually) be viewed at different levels in the hierarchy. Level may mean the particular length, or period of time, or mass or energy which characterises the level. A crystal is exceptional in that there is usually an enormous number of sub-units (typically molecules) at the same level of complexity. For example a piece of crystal may contain 10^{24} molecules not combined into macromolecules but simply stacked, following a single rule, as a crystal. The number of sub-units in the same level of an hierarchy is called its span. Crystals have a large span. Other structures in living systems have a much smaller span. This leads to Bernal's aphorism »Crystallisation is death«. Adding units to a crystal simply gives a bigger crystal. Adding units to an organic structure - the simplest is perhaps the assembly of capsomeres into the icosahedral shell of a virus particle cannot be continued indefinitely. After a small number of repetitions the rule has to change, to be replaced by a higher--order rule, thus leading to the more complex hierarchic structures characteristic of life. Life only uses crystals as stores of spare components or as mechanical building bricks such as in shell and bone. I should observe here that the structure analysts, the majority party in present-day crystallography, the people the Russians call »structurshchiki«, are not really concerned with crystals at all. They are interested primarily in how the sub-units, the molecules, are bonded internally and the forces, assuming that they are much weaker than the internal forces, which keep the molecules together as a crystal, are of subsidiary interest. (With the possibility of topochemical reactions in the solid by which one can achieve syntheses impossible in solution, the attitude of chemists is rapidly changing from this view. If the structure analyst could obtain the transform of a molecule by taking a single molecule and allowing it to scatter X-rays, then he would be more than satisfied. For him a crystal has only two functions. Firstly, it acts as a scattering amplifier, a kind of passive laser multiplying by N the amplitude of the wave scattered by one molecule Secondly, it resists damage by the radiation, in that molecules which become casualties make their contributions, now deformed, not to the discrete spots which are being measured, and which are due to the rank and file as yet unwounded, but to the background . On the other hand, the other party, whom the Russians may call the »apparatchiki«, belong completely to their instruments and are not always very interested even in crystals. I do not support either of these parties. Crystallography is, I maintain, concerned with regular structure. Regular means, governed by rules, which it is our task to discover. As Marcus Aurelius (121-180) commented: »The world is either the effect of cause or of chance. If the latter it is a world for all that, that is to say, it is a regular and beautiful structure«. That is, he recognised both determinate and stochastic operations.[5]

 The task of crystallography, then, is to answer the following questions :

(1) What is the structure ? What rules describe its form (statistically) ? .

(2) How does it work? (Dynamic aspects). (Function). (Thermal vibrations - behaviour for small displacements). (Chemical reactivity - behaviour for larger perturbations).

(3) Where did it come from and where will it go? How was it formed ? (Growth and evolution). (Morphogenesis)..

(4) How does it appear as part of the system of nature and how can it be controlled or adapted? (Application to the purposes of man - technology).

(5) The methodology and techniques by which these tasks may be accomplished.

Naturally we cannot investigate all aspects of everything, but we should not be too modest.

METHODS

Let us look first at the methods for investigating structure. I see three main categories:

(1) Spectroscopy

 This is analysis, taking the sub-units separately and looking at properties which are not greatly changed by aggregation. We have typically, mass spectrometry, optical, UV, IR spectrometry, Moessbauer, X-ray, n.m.r., e.s.r., Raman spectrometry, etc. We learn little directly about the overall structure, except through the perturbations which the sub-units suffer by incorporation. There are also, of course, forms of spectroscopy which examine frequencies, either spatial or

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[5]   The idea of regularity conformity with a rule, was hypostatised by the ancient Celtic Church into the Saint Regulus or Saint Rule who was supposed to be the founder of the university town of S t Andrews.

temporal, which are characteristic of the aggregate as a whole. The scattering of X-rays, neutrons and phonons comes into this category, but these will be considered later from a more modern viewpoint.

(2) Logical methods

For example, we can infer much ab out the structure of dichloro-benzene if we know that there are exactly three stereo-isomers. If we add hydrogen sulphide to the solution of an ion and get a black precipitate then we are well on the way to identifying it. Such logical methods are better known as chemistry.

(3) Generalised microscopy

This means looking directly to get an image of the structure, since most structures examined are primarily spatial. Imaging is the most powerful method because it enables us to use directly the computer we have in our heads gaining access through the input peripherals known as eyes. This computer is furnished with programmes of extreme subtlety steadily improved since the birth of the species and of the individual. Understanding its requirements we can match its input and output impedances to our apparatus in the most satisfactory way and use the apparatus as extensions of our human faculties, just as we use cups, knives or chopsticks, without thinking much about it, as temporary prostheses for our hands. Using appropriate detectors we can employ, for example, photons to show electron density, neutrons to show nuclear density and spin, electrons to show electric potential, phonons to show mechanical discontinuities (from tapping the wheels of a railway wagon to detect cracks to using sound waves to find nuclear submarines). There are all kinds of variations, but the salient characteristic is the diffraction or scattering of radiation, the selective processing of the diffracted beams, and the re-synthesis of the diffraction pattern to get an image of real space again, changed by magnification or in some other way (Figure 1).
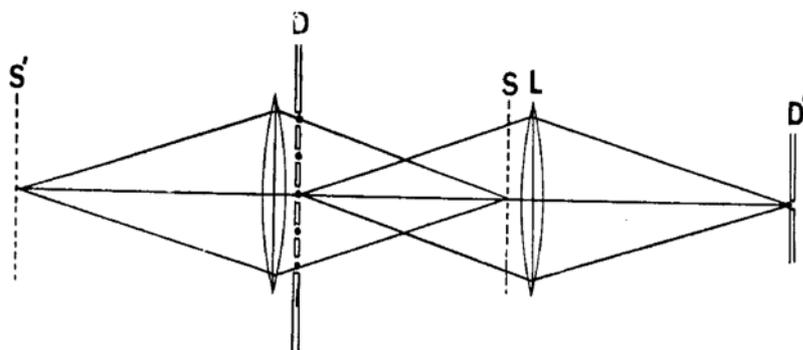

Figure 1. Optical diffractometer illustrating generalised microscopy, D' is point source of monochromatic light. L is the lens bringing the diffraction pattern of the object S to a focus in D, the Fourier transform plane, in which parts of the transform are selected to continue to re-form the image of S at S'.

Even pictures already taken can be re-processed for presentation in more acceptable form. For example, the Apollo-13 astronauts took a vital photograph of their damaged spacecraft, but it was out of focus. It proved possible, however, working in transform space, to deconvolute the image and to separate out the picture in spite of the phase distortions introduced by defocussing. Considering information theory we see that one picture .is indeed worth more than the proverbial thousand words.[6]

The stereoscan microscope is an example of a most admirable instrument, well-matched to human capabilities, because in many modes of operation it provides in a two-dimensional picture patterns

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[6]   The process by which we involuntarily focus our eyes on an object at a distance and then re-focus them when we look at a nearby book is very remarkable. 1 think that it is basically the same as that used in modern structure analysis techniques for improving the resolution of protein structure determinations by extending the region for which the phases of the reflections are known. It is based on knowing what we are looking at, so that the observer supplies some of the information.

of light and shade which can be interpreted by the brain, using a lifetime's experience, as three-dimensional structure.

THE DESCRIPTION OF STRUCTURE

The next problem is the description of structure and we will concentrate on this aspect. To describe a regular crystal structure is quite simple. Using a suitable system of co-ordinates an asymmetric unit of pattern is described. This is then repeated by the operation of the symmetry group until all of space is filled. There are only 230 such symmetry groups and they are fully defined in the International Tables for X-ray Crystallography. However, Eric Temple Bell, the historian of mathematics, once wrote: »The cow-boys have a way of trussing up a steer or a pugnacious bronco which fixes the brute so that it can neither move nor think. This is hog-tie, and it is what Euclid did to geometry«. I think that this is now what the International Tables (standing as the expression of classical crystallography) are doing to inhibit the development of crystallography. Let me give an example. The traditional description of crystal structure applies only to infinite crystals, whereas every natural crystal has defects, even if the only defect is the existence of a surface. We would really like a system which would enable us to describe all kinds of structure, a range stretching from almost perfect crystals, through disordered crystals, to structures like glasses and liquids where any reference to a pre-existing crystal lattice is meaningless. Suppose we have a particle of 55 atoms of gold (Figure 2). This has a definite structure, which consists of three icosahedral layers. The symmetry of the particle is 5m3m. Both experimental and theoretical studies confirm that this arrangement is stable with respect to a face-centred-cubic packing. If more layers are added, then the f. c. c. structure eventually becomes the more stable and a transformation occurs. The cluster has a definite structure, which is not that of a crystal, nor that of a molecule; it is not one of the space-groups (or even point groups) listed in the International Tables; it is not a twin (although it could be described in terms of twinning); I suggest that this kind of structure deserves a special name and propose that it should be called a crystalloid. (This word existed earlier in chemistry, but is now in disuse.) Clearly finite particles such as those of the viruses, the spherical icosahedral particles of for example, poliomyelitis virus and the rod-shaped particles of TMV, come into this category.[7]
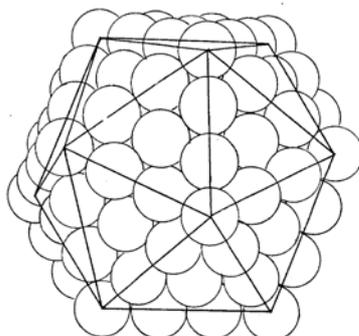

Figure 2. A »crystalloid« a 147 atom icosahedral cluster consisting of four shells (with 1 12, 42 and 92 spheres) as observed in microcrystals of gold.

I want, by this example, to assert that regular structures (those describable by simple rules) need not be crystalline. Regular structures may be stable or metastable. Particles such as the icosahedral poliovirus particles may themselves crystallise. Many of the elaborate alloy structures investigated

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[7] I would define a crystalloid as follows: A crystalloid is a configuration of a finite number of identical sub-units, assembled in a true free-energy minimum in a unique, regular and reproducible way into a cluster which is not recognisable as being a crystallite. A crystallite is a small crystal, distorted by the presence of a surface, but of recognisably the same structure as that which an infinite region of the same material would adopt. Crystalloids may contain exact or approximate symmetry operations which are not allowed in the 230 space groups.

by Sten Samson contain icosahedra and their derivatives and, I think, crystallography in general has not really come to terms with this problem of local symmetry. It is not really enough to say that the icosahedra, and the space group in which they occur, have common sub-groups. It is clear that, more and more, quantitative measures of symmetry are required. With the development of methods for measuring complexity it is now becoming clear how this may be done. A crystal is not infinite, so that it must lie in asymmetric surroundings. Usually, if the crystal is of 'reasonable' size this effect will be negligible. But, if .the crystal is only ten unit cells each way, half the unit cells will have access to the outer surface and this may make them very different from the others (by for example, the adsorption of water). Similarly the internal world of the atom has its own symmetry. Consequently the symmetry of a crystal is not absolute but only approximate. We ought to be able to say quantitatively how approximate. There is good progress in describing the small distortions of regular polyhedra but proper quantisation is only just beginning. The problem is a very general one and is closely related to the way in which we look at complex structures by decomposing them into sub-structures. If the sub-structures did not interact to form the structure, then they would not be perturbed and vice versa. If the sub-structures interacted very strongly then they could no longer be distinguished as sub-structures and the matrix which represents the interactions .between them could not even approximately be decomposed into nearly block-diagonal form. Completely block-diagonal form would represent non-interaction. Inasmuch as w e can see a large structure as built up of sub-unit-s, each of which is itself built up of sub-units, the structure is hierarchical. As early as 1788, Buffon said: »all t-he work of the crystallographers serves only to demonstrate that there is only variety even here, where they suppose uniformity . . that in nature there is nothing absolute, nothing perfectly regular« (Histoire Naturelle des Mineraux, 111, 433, Paris, 1732-8).

A superstructure cannot be completely predicted from a knowledge of its sub-units, so that each structure must be investigated at each level of complexity.

TAXONOMY OF STRUCTURES

Let us look at crystal and other structures from the point of view of the arbitrary information which they may contain. This means that we must deal with metastable structures, since a perfect crystal tells us nothing. It is simply at the absolute minimum of the free energy surface in N-dimensions. We are interested in those cases where a structure is trapped in a local minimum in a metastable state. This is arbitrary information, since we need to specify which local minimum it is in. In 1944 Schroedinger referred to the hereditary substance as an »aperiodic crystal«. Now we can look on the DNA sequence as carrying two bits of information per. pair of nucleotides. The molecular weight required to carry one bit of information, keeping it reasonably safe against disturbances of the order of KT, and taking it through read-in and read-out processes, is about 100. Dislocations and 'disordered' layer sequences represent information which tells us something about the history of a piece of material.

Working upwards in the levels of complexity we can see, I believe, a progressive segregation of the information-carrying structure, until in a living organism we can see the fully-developed system where enough information to regenerate the whole structure is carried in each cell. Since seeding a solution with a crystal will enable a meta-stable crystal to reproduce, we have a similar process at the lowest level. The process of crystal growth in this way is only significant as an information carrier when the crystal structure is metastable and causes the production of a form which is thus arbitrary to the extent of one or two bits. If the nucleation by any type of nucleus produces the stable form then no arbitrary information is passed on. G. D. Woodward and W. C. McCrone (J. Appl. Cryst., 8, 342, 1975) give some fascinating examples of substances where for years the meta-stable form was always produced, but where once the truly stable form has appeared, nuclei from it were propagated everywhere as dust and nobody could make the metastable form any more. (The examples ere amplicillin, ethylene diamine tartrate and some sugars.) In this case the information-carrying structure is not differentiated from the structure itself.

In silicon carbide we can perhaps see the rudiments of an information-carrying process, in that we could perhaps have an arbitrary sequence of layers (either hexagonal or cubic in their surroundings). If the crystal were then dislocated with a Burgers vector perpendicular to the layers and of the same length as the sequence we wished to transmit, then material added to the surface

would crystallise in the required sequence and the crystal would reproduce itself. We will look at various structures and ask where arbitrary information is stored :in them.

THE GENERATION OF FORM

The most obvious question to ask, when .faced with structures, is how are they formed, that is, what are the rules which lead to the organisation of matter into these forms. This is the problem of morphogenesis and, particularly since the clarification of the role of DNA in heredity, its study has received a great impetus.

C H. Waddington (»Towards a Theoretical Boology«, Edinburgh University Press) has suggested a number of basic categories and I will elaborate on his list :

(a) *Unit generated forms*

Assemblies of components which may be particles, fibres or sheets and of one or more kinds. Crystals clearly fall into this category.

(b) *Instruction-generated forms*

Here the instructions or programme for the assembly of the structure are written in some meta-language. (A meta-language is a language used for speaking about language itself.) The natural system of the translation of DNA into protein and the hypothetical models of self-reproducing automata suggested by von Neumann fall into this category. I will return to the question of translation processes later In the natural case, DNA can be translated into protein but the influence of protein on DNA is only through the process of natural selection. We could imagine cases in which there were two languages and a structure in either one could be described in terms of the other.

(c) *Template generated forms*

In the case of crystals, epitaxy and topotaxy fall into this category. There are two cases: either a substrate produces a structure like itself, or it produces a structure complementary to itself. The latter case, for reproduction, would imply a series of operations which would occur in a closed loop. As a special case we have the 'crystal engineering' of Schmidt where spatially controlled chemical reactions are used (J. H. Thomas, Phil. Trans. Roy. Soc., 277, 1974).

(d) *Condition-generated forms*

These are forms, like crystals perhaps, which occur by the operation of some global minimisation principle. Plateau's problem concerning the shapes of soap films is a matter of finding surfaces of minimum area. (We might note that there may be several solutions of different energies depending on the topologies allowed.)

To Waddington's list we might add two other items :

(e) *Standing waves*

When a wave process is confined by boundary conditions, patterns result. The model for this is Chladni's plate. The shapes of atoms, and thus the requirements of chemical bonding, result from the eigenfunctions of the wave equation with spherical symmetry.

(f) *Cellular automata*

The last process (is more mathematical or computer science or informational than physical in nature and can be given the heading of cellular automata or, perhaps better, local programming. I wish to develop the last concept of local programming.

LOCAL PROGRAMMING

The idea of crystal or other forms appearing as the result of standing waves in an enclosure of definite shape is a wave description of structure. The concept of cellular .automata is really one coming from the field of mathematical games and is a point-by -point particulate description.[8]

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[8] Most physical processes can be described, with more or less difficulty in either a wave notation or in a particle notation. Which you use depends partly on the scale (the dimensions of the components compared with the characteristic wavelengths). In crystallography we are accustomed to this duality and can describe most of the phenomena of crystals either in real or in Fourier transform space. We are thus natural Manicheans in a largely

You may know the »life game«, invented by J. H. Conway in Cambridge, which has used up enormous quantities of computer time. We have a number of pieces, like those in draughts, set out on an infinite chessboard. A set of transition rules describes how the pattern at one moment changes into the pattern at the succeeding moment. A piece may have neighbours on any of the eight adjacent squares. If it has two or three neighbours it survives for the next generation; if it has less than two neighbours it dies of loneliness; if it has more than three neighbours it dies of overcrowding. If there is a vacant square with exactly three neighbours, then a birth takes place there. These simple rules produce an astonishing richness of behaviour.

If we think of the growth of a real crystal from the vapour, we can recognise that we already use similar rules. If a molecule alights on a flat surface it re-evaporates. if it is close to the shelter of a step it may migrate into the re-entrant angle of the step and remain; if it can get into a corner, then it moves to one and so on.

It seems probable that the problem of the snowflake, which caused Kepler to formulate his concept of the 'facultas formatrix' may be solved in this way. Stanislaw Ulam, a mathematician who has for more than a decade produced patterns by various types of cellular automata, has given very simple rules which in three-dimensions give arrangements very like dendritic crystals (R. G. Schrandt and S. Ulam, in »Essays on Cellular Automata« ed. A. W Burke, University of Illinois Press, 1970, pp. 232-243.).[9]

We want to provide a notation which will take us all the way from the structures of simple crystals like NaCl to the bacteriophage which is constructed in a recognisably mechanical way by complex paths of self-assembly.

The extension to the International Tables which I would propose is by way of providing, instead of the simple rules which describe the operation of glide planes, screw-axes, etc., more complicated snatches of programame which contain conditions, that is, statements such as »IF...... THEN . . . . . .«. These might bring a closer resemblance to real crystals which have apparently arbitrary growth features.[10]

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orthodox world. The Manicheans of Iran believed, quite reasonably, that God and the Devil were equally effective forces in the world and that each should be respected. They thus had the idea of dynamic equilibrium between two tendencies and might be said to be the forerunners in using a dialectical method of analysing phenomena. The Bogomils of Bosnia were instrumental in bringing this heresy to Western Europe and earned much obloquy through it. The great South Slav intellectual, Matija Vlacic (152-1575) continued this dialectical attitude. Normally, when we are able to describe a phenomenon in either wave or particle terms, then we can say that we have a reasonable understanding of it.

In crystallography a current field of interest is the development of a wave (that is, lattice vibration) description of thermal transformations in crystals where a particle-by-particle description is only a first approximation. In the dialectical process of attempting to describe the same thing in two different languages we deepen our understanding of it.

Since we are being ideological; 1 should mention that the traditional »Dialectics of Nature« of Friedrich Engels (about 1870-80) seems to carry over quite well into modern terms. We may equate his three major principles as follows:

(a) the unity of opposites wave/particle duality. inseparability of the observer and the observed in relativity and Heisenberg' s principle, etc;

(b) the negation of the negation the principle of Le Chatelier (1888);

(c) the transformation of quantity into quality the general theory of phase transformations, the Ising model, the general theory of catastrophes, etc. Asking of a physical system where these principles were to be seen would indeed lead to fundamental insights. Engels, who kept up to date with the controversies of the day, would have had interesting comments on modern molecular biology, systems theory, etc.

[9] For some time 1 believed Tolansky's theory of the snowflake because of its elegance, but now 1 must transfer my allegiance to the cellular automaton theory. Tolansky's theory was that a snowflake behaved as a Chladni plate and that water molecules only settled at the nodes. If a molecule could attach itself somewhere it produced a nodal system, like touching the vibrating place with a finger and thus more water molecules could condense on all the symmetrically related places. It seems that this is not the correct model but in fact, that the local vapour pressure controls the growth.

[10] There are two ways of considering the relationship of the asymmetric unit of pattern to the unit cell.

(a) W e can ask under what operations is the completed pattern invariant? This corresponds to the use of the diagram of the interlocking symmetry elements which make up the group. It is not very helpful in generating the pattern.

(b) W e can begin with the asymmetric unit and can operate on it with each of the symmetry operations in turn and generate new parts of the pattern continuing until no new parts appear. This process is not too easy to systematise for a computer and demands tests to see when the pattern is complete and whether any particular part has already been generated. It is clear however, that the second viewpoint is the one which is most readily developed as it embodies a
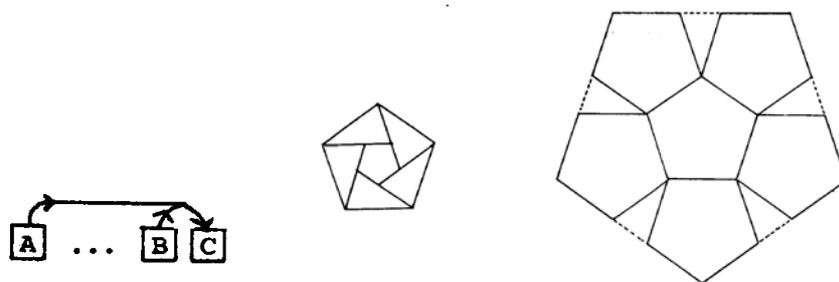
Let us look at examples of local programming in 1, 2 and 3 dimensions. We wish to show the complexity which can result from simple transition rules; beyond those of the International Tables. Consider a linear sequence of binary digits (1 or 0). The value of the next cell C is given by a matrix of values depending on :the two previous values of the cells A and B.

(In this case the value in the matrix is the difference mod 2 of the two arguments.)

This is the programme. B is the cell immediately preceding C and A is the cell (N-l) digits earlier. The sequence of N cells A.. . B serves as the seed for the process. For example; if N = 3 and we begin with the seed 111 we obtain the sequence 111/0100111/0100111/ . . . of period 7. For other values of N the periodicities are

N period(s)

2 3
3 7
4 15
5 21, 7, 3 (where there are several periods; the one encountered depends on the value of the seed)
6 63
7 127
8 63, 63, 63,63, 3
9 73......
10 889, 7,. . . ..

Figure 3. Assembly of higher order pentagon from six pentagons plus five triangles, the latter obtained by dissecting a seventh pentagon.

The object is to demonstrate that a simple rule, involving only local short-range relationships; can produce a complex long-period structure. It is possible that the sequences in polytypes such as SiC

kind of local programme. Conditions such as REPEAT THE MOTIF X BY THE PLANE M IF THE RESULTANT MOTIF X DOES NOT COME NEARER THAN A DIST ANCE D TO A PART ALREADY GENERATED are not greatly different from what we already use, particularly if we are considering the packing of actual molecules. This second approach corresponds to the use of the table of equivalent positions which may be read: IF (X, Y, Z) THEN ALSO (X + 1/2, -Y, Z) etc.

might be explained by such a notation, but I have not yet been successful. Here ternary digits might be necessary.

F or a long time I have been interested in the occurrence of structures with five-fold symmetry which lie outside the compass of the International Tables. I have tried a number of experiments in generating pattern's having simple rules but which were not crystalline but were nevertheless regular. In particular, I have tried to produce a pattern illustrating a hierarchic structure produced by a recursive programme.

Most elementary texts state (that you cannot tile a floor with regular pentagonal tiles. This is, of course, correct but suppose we try to do :it by making the minimum number of changes to the rules. If we put six pentagons together we get a larger pentagon, except for five small isosceles triangles which need to be filled in. There are two rather interesting ways in which this might be done. The first is to take a seventh pentagon and cut it up (as shown in **Figure 3**) to obtain the required five triangles plus a smaller pentagon. Thus from seven pentagons we can make one regular pentagon which is bigger by a linear factor of Φ (where Φ is the golden number 1.618033989. .) and one which is smaller by the same factor. We have the relation $\Phi^4 + \Phi^{-4} = 7$. Thus we can continue the same process until the bigger tile overlaps the area we wish to cover and the smaller tile all but disappears (**Figure 4**).

For producing a similar pattern in a different way, using only two different component pieces A and B (see **Figure 5**), we can proceed to give rules as follows:

.
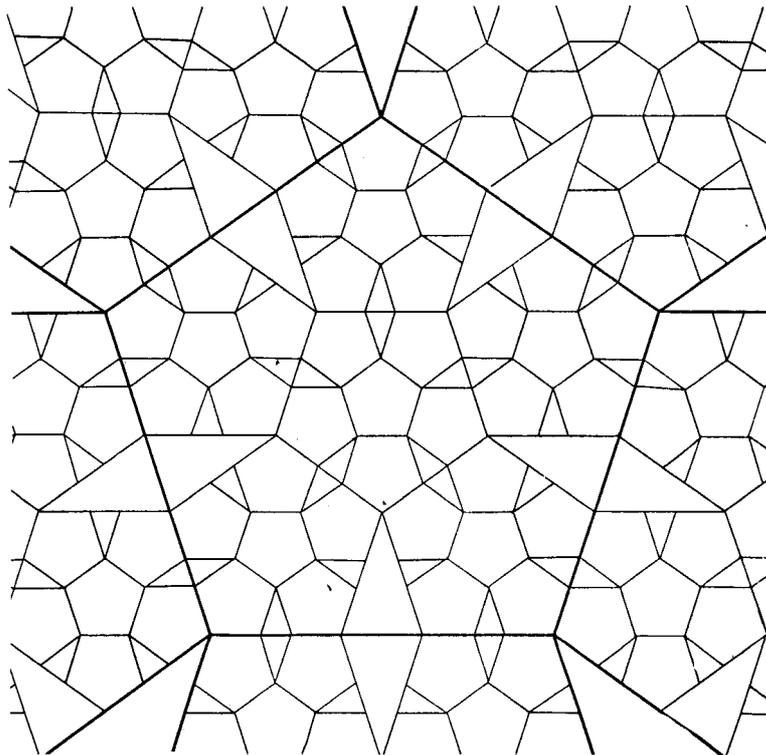
First order pentagons are made from 3A + B as shown in **Figure 3**.
First order triangles are made from B components.
Second order pentagons are made from six first order pentagons plus five first order triangles.
Second order components B are made from A + B as shown.

Figure 4. Hierarchic structure produced by the packing of pentagons according to a recursive rule.

Second order components A are made from 2A + B as shown. Rules must be given for the orientation of .the pieces relative to each other working outward from an initial point. The limiting composition of the pattern is A B: it is an example of a non-crystalline regular structure hierarchical

in nature, involving only two types of units. The transform of the pattern is surprisingly non-crystalline in appearance.

As examples of three-dimensional figures produced by a local programme, we may again cite the work of S. Ulam, but illustrate the style with a two-dimensional dendritic pattern (**Figure 6).**
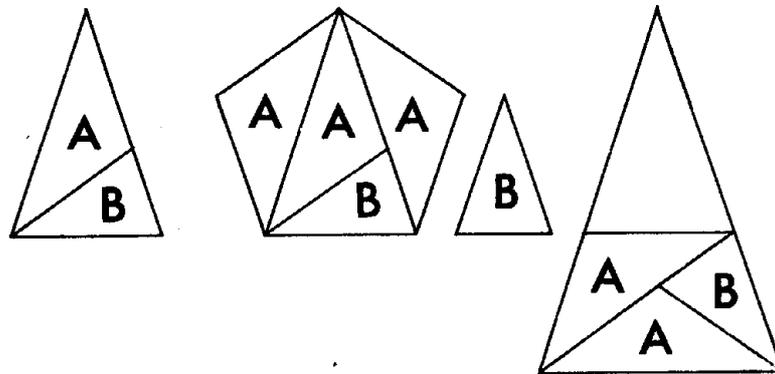

Figure 5. Exemplification of the recursive rules for building pentagons and triangles from pieces of the two types of isosceles triangles A and B. At each stage the linear dimensions are multiplied by $\Phi^2 = 2.618....$

THE DESCRIPTION OF STRUCTURE

Having seen examples of rule-generated structures we can proceed to the more general discussion of
the rule of descriptions. The first problem *given rules (= local programme), generate the structure*
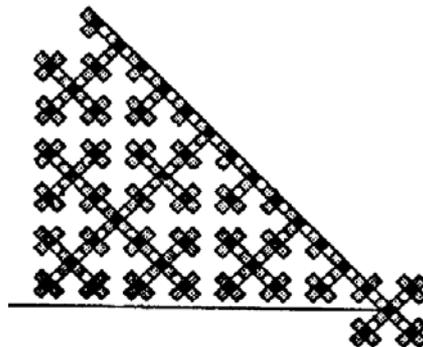is

Figure 6. Asymmetric unit (one eighth) of the plane pattern generated by a simple cellular automaton (local programme of transition rules).

not difficult. In a material structure the rules are written in the language of geometry and interatomic forces. The statements are executed using the energy of the thermal vibrations flowing through the system. The structure shakes down to the lowest free energy minimum which is accessible, bearing in mind the history of the changes in the system. In a phase change like freezing and the production of, for example, a snow crystal, thermodynamics is not enough we must know the exact details of every component. Thermodynamics tells us only when a phase change (in a social as well as in a physical system) is energetically possible. It can say nothing about the actual details of nucleation - this depends on microstructure at the level below that at which we may be

observing. We can write varied programmes and see whether the results bear any resemblance to physical (or social or biological) systems which we may need to understand.

The inverse problem is given a structure, recover the programme which gave rise to it and is clearly much more difficult. It brings us up against an absolutely fundamental difficulty which is the size of factorial N. If we have N objects or symbols, they can be combined in something like N! ways. R. J. Boscovich realised this (»Theory of Natural Philosophy«, 1763. Art. 541, 552). We wish to find why a particular combination occurs but, with more than a few components, it becomes impossible, even in principle, to examine more than a few of the possibilities. If we have a protein chain of 100 amino-acids, the possible sequences are of the order of 100! Voltaire commented that »if this is the best of all possible worlds, what the others may be like does not bear thinking about«, but we cannot think about more than a minute fraction of them. In order to describe a structure, since we cannot do it exhaustively, we have to use all our cunning and even then may fail to see the order evident in our structure. If so, we must characterise the structure as random or sui generis or unique in its complexity.

Gregory Chaitin (Sci. Amer., May 1975, . 47-52) has recently made an important contribution to the clarification of our ideas of randomness and complexity. In summary, suppose we consider a sequence 0f N binary digits, N being perhaps 100. Then if we have a sequence 00111001110011100111 . . we can say it is non-random and can describe it as (00111)20 the same group of five digits repeated 20 times. In binary 20 is 10100 so that we could use the symbol (00111)10100. This has described our 100 digits by 10. Clearly the sequence is non-random and Chaitin's contribution is that he gives a quantitative description of randomness or disorder or complexity. If a sequence of N digits requires about N .digits for its description then it is random, or has a complexity of N. The degree to which condensation of the description is possible represents the degree of order. According to this criterion crystals are very highly ordered and the International Tables furnish condensed descriptions.

Chaitin's description does not remove the human element from the study of structures - it introduces it explicitly.We might recall a definition of (human) intelligence given by H. A. Fatmi and R. W. Young (Nature 228 October 3 1970, p. 97). » Intelligence is that faculty of mind, by which order is perceived in a situation previously considered disordered«. (It is immediately clear why diffraction, producing as it does the Fourier transform of an object, which shows up periodicities, is an extremely powerful method .of finding the presence of certain kinds of order). The more intelligent .you are, the shorter is the description (in terms of generative programme) which you can give, but there are limits. The descriptions of elaborate concepts which may simplify the description must themselves be included in the description. My critique of the International Tables is that they describe only the first set of rather simple rules for generating or describing structures. Chaitin restores the role of intelligence in crystallography. The International Tables have been a factor in inhibiting the growth of crystallography because they allow only one kind of description. They are clearly quite unsuitable for dealing with biological structures; but even for crystals they are inadequate. They cannot deal with dislocations, defects, twinning and the like and, in for example the case of niobium oxides, of almost continuously variable composition. Very high resolution microscopy, where one can almost see individual atoms, has shown the essential complexity of most materials. The transform .of defects appears mostly between the lattice spots and thus has been largely neglected by the structure analysts.

THE TRANSLATION OF DESCRIPTIONS

We then see:
(a) a structure can be described in a meta-language;
(b) a structure and it's description are of similar complexities.
(c) the meta-language description of a material structure .is also
material and must be written in atoms (or in waves of some kind);
(d) a description is not a complete description of all conceivable features of a structure, but only of those considered significant;

e) if a structure is highly redundant or symmetrical, then it can have a concise description which, in terms of number of atoms needed for the materialisation of the description, may be small compared with the structure itself.

We may now consider a complex quasi-physical process, which I will call translation, by analogy with the process of translation between natural languages (and not referring to the simple geometrical translation or displacement). Translation is a transformation in which .the introduction of some external information :is necessary. The simplest example is the description of a molecule in terms of Cartesian coordinates. An alternative description is to use the matrix of all the interatomic distances [dij]. This can always be calculated uniquely. We can also reverse this transformation, but not quite perfectly, in that we have to supply from our external experience, the information as to the cheirality of the molecule (whether it is left or right-handed). The next simplest example is the process of projection a three-dimensional object is projected on to a two-dimensional plane. This can always be done, but the matrix which describes the transformation is singular and has no inverse. The reverse transformation, the restoration of a three-dimensional object from a two-dimensional projection is thus impossible without external information. It is thus a translation rather than a transformation.[11]

It is a common psychological observation .that when the observer has the wrong preconceptions about the three-dimensional object which he sees only in projection, the physical data is misinterpreted to force it to make sense in accordance with the preconceptions. This demonstrates the interpreter' s contribution to the process of 'translation' .

Let us take some further examples of the translation of connected pairs of descriptions of structures. These give some answer to the chicken .and the egg paradox. As Samuel Butler pointed out, the chicken is the egg's way of making another egg so that we might regard ourselves as a complex epiphenomenon associated with the polymerisation of DNA. Obviously the .two systems each describing the other in meta-language are both necessary in that it is the alternation from one to the other which enables the joint system to evolve.

A simple analogy is a small dynamic system. For it to oscillate, that is, to show behaviour, it is necessary for the energy to be stored in two forms and .to be interconvertible. Normally we have kinetic energy and potential energy. The order of the differential equation describing the motion is the same as the number of the different modes of storing the energy.

Examples of pairs of systems are as follows:
(1) Physical structure - description on paper (blueprint)
(2) Spatial structure - brain mapping of this structure
(3) Brain structure - language
(4) Language - written representation
(5) Language A - language B
(6) Organism: protein (phenotype) - DNA (genotype)

Translation is done under the constraint that at each stage it should make sense (that is the translator or observer forces it to make sense). If it fails to make sense the particular individuals concerned may fail to be reproduced and may take no further part in the history of the system as a whale. The resulting products are selected by evolution and may be a little better or worse after the process of translation and re-translation than before. This gives the possibility of the evolution of !the whole system through the perpetuation of the changes.

Although the process is described in anthropomorphic terms, the same process, in vestigial form, can be seen in the growth of crystals where dislocation and mistakes occur affecting the subsequent growth history. In these cases the information content of the structure has not separated from the structure itself.

To summarise:

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[11] * If we have an equation [A] x =h it has a solution x =$[A^{-1}]$ h. One can, however, construct a generalised inverse [Ag] by iterating the expression [A] $[S_{k+1}]$ = [A] $[S_k]$/[2I - AS_k] where $[S_{k+1}]$ converges to the generalised inverse [Ag] : The solution is then x = [AgJ h + [1 - AAg]z where z is arbitrary.

(1) We have experimental methods of seeing how sub-units (typically atoms) are assembled into more complex structures (typically real crystals). These make up traditional crystallography.

(2) A fruitful question is to ask how complex structures are to be described. A description is a representation of a structure in a meta-language. (As a building may be represented by a blue-print.)

(3) A description of a structure comprises a computer programme and a body of information about the mechanics of the system.
A generalised machine may carry out the manufacture of the structure from its description.

(4) The description of a series of digits can be a shorter series of digits (if the structure described is redundant or symmetrical). Analogously this can b e materialised so that the description of a structure of atoms can be another set of atoms.

(5) A description is never complete. It describes what are considered to be the essential features of the structure. A description can be reversed to regenerate the structure (with greater or lesser fidelity).

(6) In passing from one description to another, or from a description to the structure and back, variations creep in. They give rise to the possibility of the evolution of the structure by natural selection.

(7) A structure assembles itself as each component takes its position according to local rules. For simple structures the rules are a local micro-programme determined by the neighbouring units previously assembled.

(8) For more complex structures we can single out components especially concerned with the assembly of other components. The concept of template becomes more elaborate until it becomes almost a description of the structure in a meta-language.

(9) We can then see compound structures where the total structure contains its own description as a sub-structure. This is first clearly exemplified in the bacteriophage which contains its own description in its DNA.

(10) This process can then be contained recursively so that a structure plus its description in physical form can be described in a higher-level meta-language, and the two can be combined to give a higher-order assembly, which can itself be described in a further meta-language, and so on.

(11) Such a scheme allow s the emergence of higher properties, such as the eventual appearance of mind-like behaviour. Roughly speaking, mind and matter are related to each other as a language is related to the structure it describes.

(12) In this way we can connect the structure of NaCl with that of the bacteriophage (which represents a materialisation of von Neumann's self-reproducing automaton). A continuity between living and dead structures can be discerned.

(13) We consider all structures to exist in a flux of energy so that we are not concerned with absolute stability but only with meta-stable states.

(14) A particulate description has been used but probably the whole argument could be transposed into wave terms.

CONCLUSIONS

I believe that most of the historic objectives of crystallography are now accomplished. That is, there are now successful methods for finding the structures of more and more complicated systems. In attacking the International Tables, what we may call classical crystallography, I do not wish to disparage in any way what the structure analysts and the apparatus people are doing. I want to suggest that if crystallography is to continue its successful progress, as a recognisably coherent subject, we need to formulate new objectives and a new ideology. I would like to see crystallography become a general science of structure. I believe that with the special insights which we have acquired into the geometrical structure of matter we are in a good position to participate in the next offensive which is to explain and control the behaviour of more and more complicated systems, most of which are not crystals. The controlling effect of geometrical structure on what happens at the molecular level is absolutely paramount. I consider that big discoveries in the way matter organises itself to produce and maintain life are coming and that we can contribute greatly to them. If you asked me for a distant objective for the new crystallography I would suggest the

following question for your consideration : "How does matter think ? ". However, the next step in this direction is to discern order in simpler structures hitherto considered too complex or disordered for comprehension. The way to this is by systematic experiment, but we must have a preliminary ideological framework in which the experiments can be formulated and analysed. I have ventured to speculate on this, and I am most grateful to the Yugoslav Crystallographic Centre for having afforded me this opportunity for putting my views.