Hierarchic Structure

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Abstract

We are concerned with structuration, the ways in which complex structures are built from simpler components, and especially with levels of organisation, primarily from a geometrical point of view, with the objective of developing a generalised crystallography extending from the atomic level, beyond regular crystals, towards biological structures which contain fibres, membranes and other sub-structures, some informational. Some levels may be random, some crystalline and some may be regular in different ways Materials characterised as textures must now be specified in more detail. The quasi-crystal affair has provided the stimulus for a fundamental re-estimation of orthodox crystallography.

“If this is the best of all possible worlds, then what the others may be like hardly bears thinking about” — Voltaire, (Candide), (1758).

Newton, having determined “the motions of the planets, the comets, the Moon and the sea”, was unfortunately unable to determine the remaining structure of the world from the same propositions because: “I suspect that they may all depend upon certain forces by which the particles of bodies, by some causes hitherto unknown, are either mutually impelled towards one another, and cohere in regular figures, or are repelled and recede from one another. Those forces being unknown, philosophers have hitherto attempted the search of Nature in vain; but I hope the principles here laid down will afford some light either to this or some truer method of philosophy”. Preface to the “Principia”, (1687). ¹

“It will be found that everything depends on the composition of the forces with which the particles of matter act upon one another; and from these forces, as a matter of fact, all phenomena of Nature take their origin”. R. J. Bošković. “Theoria Philosophiae Naturalis”, Venice, (1763). (sec. 1.5).

“A number of quite independent lines of argument converge toward the assertion that there is often a limit to the complexity of systems”. Richard Levins, (“Towards a Theoretical Biology”, 3, 73-88, (1970)).

"Physics is simple only when analysed locally". Misner, Thorpe and Wheeler, ("Gravitation")

Quasi-crystals are only part of the revolution in thinking about structure brought about by the computer and computer graphics\(^2\) [8]. I want to direct your attention to the "Generalised Crystallography" first promulgated by J. D. Bernal [2] about 25 years ago. X-ray crystal structure analysis has been so successful that people have forgotten that most materials are not in fact crystalline. In 1959, J. M. Burgers [3], very conscious of the defects in crystals, discussed generally the problems of the emergence of patterns of order. He had almost discovered the Lorenz attractor. Discussions current in the period 1930-1960, for example the important Faraday Society meeting on liquid crystals, can now be picked up again at a new level.

As you will see, I am trying to collect together many rather disparate parts of a general science of spatial structure and, since it is very difficult to organise them into a linear sequence, have begun to use the Hypertext system to handle them as a strongly connected network. This corresponds better, I believe, to the way ideas are organised in the brain than to a linear paper. We can think only about a few items at a time, so that in order to organise our thinking about structures, we have to seize on the key feature of hierarchy [15] [20] [22].

1 The Blind Watchmaker

The Blind Watchmaker [4], besides making life, also makes inanimate materials.

My comments are based on the parable of the two watchmakers presented by H. A. Simon in his book [20] *The Sciences of the Artificial*.

A watch has 100 parts. The first watchmaker assembles each part in turn but on each occasion when he is interrupted, the watch falls to pieces and he has to start again from the beginning. The second watchmaker, subject to the same interruptions, has broken down the construction into 10 subunits each of ten parts. He makes subunits and then assembles 10 of these to make a watch. Needless to say the probability of his completing a watch is enormously greater than that of the first watchmaker. The difference is due to *hierarchisation*. We should also note that the watchmaker has some kind of description of a watch in terms other than a completed watch itself.

Hierarchisation is one of the basic principles of constructing anything. This is well-recognised by protein crystallographers who see primary, secondary, tertiary and quaternary levels of structure in proteins\(^3\). Protein structures are all built of domains in this way. The domains are seen as an "implicit structural consequence of the folding process" [17]. Inorganic structures are hierarchised by seeing coordination polyhedra which are then linked into larger features like, threads, sheets and frameworks. Molecular compounds are usually seen as having only two levels, the molecules themselves, where atoms are linked by covalent

\(^2\)Many colour pictures were shown in the lecture which thus corresponded only roughly with the present text.

\(^3\)primary structure is the sequence of amino-acids; secondary structure is the alpha-helices and beta-sheets produced by favoured angles between units; tertiary structure is the way in which sheets and helices twist and turn to make globules; quaternary structure is the composition of globules to make the whole protein.
bonds which are much stronger than the forces between molecules, and solutions or crystals of molecules.

Liquids and amorphous materials present special problems because it is difficult to find distinct levels.

The key feature is that in a hierarchic structure the rule of composition at each level has to stop and to give way to a new rule for the composition of the next higher level. Orthodox crystals have only one rule. The classical symmetry operations do not explain how to stop. The considerations of facial energy introduced by G. V. Wulf and developed by Donnay and Harker, which explain external morphology, are the next step and after that questions of grain boundaries. N. Rivier [16] has connected grain boundaries with quasi-crystals, suggesting that, between coincidence site lattices and completely general grain boundaries, quasi-crystal interfaces form a preferred type.

2 Levels of organisation

At a given level there may be a particular kind of structure, crystalline, random, textured, curved manifold.

A good example is opal. Here, amorphous hydrated silica forms a random network; layers of such network form spheres like hailstones; these spheres crystallise (to give the layers with spacings in the optical wavelength range so prized in precious opal) [6]; regions of crystalline and non-crystalline opal form rock.

Hans Zocher has demonstrated crystals of crystals with $\beta - \text{FeOOH}$ and tungstic acid crystals. Sadanaga, Takeuchi and Morimoto [18] have shown that minerals with complex crystal structures can be seen as built of fragments of simpler structures. A. D. Wadsley and others have greatly simplified the crystal chemistry of complex oxides with this outlook which has been amply confirmed by high-resolution electron microscopy, which shows simple coordination polyhedra coupled in a complicated ways not easily to be disclosed by X-ray diffraction.

A regular hexagonal packing may occur at the scale of the Giants' Causeway, although through physical processes which are quite different from the process which produce hexagonal graphite. Physical forces are not scale-independent.

3 Is there a limit to complexity?

Hierarchisation also takes place in the inorganic world and complex structures are built out of simpler in several levels [18]. There are limits to complexity. For example we may ask our data bank: what is the largest number of different elements which occurs in a crystal structure (where each atom is in a distinct site without solid solution)? The answer is about seven or eight.

We may ask also about the structures which have the largest unit cells. They are almost all proteins, or polytypes, that is, modulations of simple structures or complex framework silicates, that is, recognisably hierarchic structures. An example is paulingite, which is cubic with a unit cell dimension of about 35Å containing a thousand or so atoms, as many as a protein. How much information is necessary to control the formation of such a structure and where is it or, where are the genes for paulingite? Cellular automata begin to give us a clue.
I guess that there is a natural limit to complexity and to exceed this it is necessary to use special structures for the storage of programme. This is characteristic of life, but we see computers as synthetic systems, and the search is on, under the stimulation of the ideas of Graham Cairns-Smith, for inorganic systems which may store arbitrary information controlling the reproduction of other components. In mixed systems it is usually the biological which control and use the inorganic [13].

4 Cellular Automata

The development of cellular automata [23] has perhaps changed our ideas about complexity in showing the richness of patterns which can appear from very simple local rules. In the bonding of atoms we have local rules, but with a stored programme, more arbitrary rules can be implemented. As an extreme, the Mandelbrot set is an infinitely complex structure emerging from a simple equation, more picturesquely than the digits of \( \pi \) emerge. However, it is a mathematical and not a physical structure and takes no account of the natural objects, such as atoms, which are available for making structures. It has no scale and no upper or lower limits.

In a crystal the same rule takes one an enormous distance but characteristically in biological objects, only a few units can be added at the same level before the rules of combination change.

The decimal numbers used for counting are an example of perfect hierarchisation. After counting from 0 to 9 we must carry one to the next most significant place and begin again. The Turkish army is organised like this. A corporal Onbaşı is (literally) the head of ten; a captain Yüzbaşı is the head of a hundred and a major Bimbaşı is the head of a thousand. As has been known from antiquity this is the way to control large numbers. If they act in unison the amplitudes of their individual efforts add and the total intensity is the square of this (as explained by F. W. Lanchester [11]). If they act as individuals the intensities of their efforts add.

As Simon points out with respect to opening the combination lock of a safe, if we can separate a structure into its hierarchic levels we can analyse its behaviour enormously more easily (and the safe can be cracked).

Nothing is totally separable. EPR (Einstein, Podolsky and Rosen), Bell and Aspect show that everything is connected to everything else, albeit weakly, and that levels in a hierarchy are not completely separable. Equivalence must be replaced by quasi-equivalence.

If the span of levels is large, for example 10, then the levels are more clearly separable than if the span is only two.

One of the most interesting features of the Penrose tiling / Quasi-crystal affair is that hierarchic structures have been introduced into formal crystallography [12]. This begins to resemble the renormalisation procedures which connect interatomic forces with whole-body phenomena such as phase transitions.

\(^4\)The ordinary binary numbers are not the best way for machines to count because more than one digit may change at a time leading to uncertainty. The proper counting sequence is the Gray code. e.g. Nature, 340, 514, (17 Aug. 1989).

\(^5\)Just these tactics were used intuitively by Octavian to defeat Mark Anthony in the naval battle of Actium which was fought just off Preveza on 2nd, September 31 BC (2019 years ago) — One of the decisive battles of the world.

\(^6\)Richard Feynman has told you how to do it.
4.1 Logical paradoxes

The problem is that the levels in the hierarchy of inorganic and particularly metallic structures are not very clearly separable. Metals are almost all close-packed assemblies of atoms each with about 12 neighbours. Kauffman writes: "The cyclic behaviour of the network now is equivalent to logical contradiction in its circuitry. We suggest that spontaneous persistent activity in deterministic discrete systems is the equivalent of self-contradiction in the networks" [9]. That is, attempts to resolve frustrations repeat cyclically.

The Burgers vector is a measure of the frustration in a static, spatial structure.

It is also evident that in setting up some of the various logical paradoxes which have confounded philosophers, we are in fact constructing cellular automata of this type. An example of this type of frustrated structure is the card which carries on one side the words "The statement on the other side of this card is true" and on the reverse side the statement "The statement on the other side of this card is false". We should not be surprised if linguistic structure reflects the structure of the real world. Linguistic philosophers have made very heavy weather of such paradoxes.

A similar situation can be found in spin glasses.

5 Two-dimensional structures and differential geometry

The inorganic way to look at a structure is to consider the network of interactions between point atoms, attaching appropriate force functions to pairwise or multiple interactions.

A more biological way is to regard atoms as force centres and to allocate domains to them. The simplest type of domain is the Voronoi polyhedron and many more complex criteria for boundaries can be devised. The next simplest is the radical plane division and, with Coulomb forces between charged ions, equipotential surfaces appear dividing up space. Surfaces are structures orthogonal to lines of force.

The characteristic shapes of protein and other molecules which interact with great specificity are largely discussed in terms of potential contours. These are of intense interest in drug design.

Further up in scale we can consider membranes made of lipid bilayers which define inside and outside and give the possibility of cell structure characteristic of living systems. These membranes are effectively two-dimensional manifolds [19]. We may consider surface tension either as energy per unit area over the surface or as force per unit length along the perimeter. Similarly, we may find the integrated Gaussian curvature either by summing over the area or along the perimeter.

Membranes may have elastic properties. F. C. Frank [5] has given general expressions for the energy of a nematic which can be used for long molecules in membranes. Membranes, in addition to bend, splay and twist energies, may have some of the properties of a rigid medium. Gradually we can introduce force considerations into the geometry. Surface tension

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7There was an interesting study by Stuart Kauffman in 1969, in which he showed that cellular automata with high coordination numbers had much longer cycles of repetition than those with low coordination numbers (2).

8Such a frustrated structure is the basis of the children's game where an odd number of children stand in pairs in a ring. The odd one out stands in front of a pair and the rear member of the pair is displaced.
(and more complex expressions for energy) represent a way of carrying the interatomic forces seen at one level, to the handling of structures at the level of membranes where the individual atoms are not resolved.

Two-dimensional structures are of particular interest because, living as we do in three-dimensional space we have one more dimension and can sit back and see the two-dimensional structures embedded in the three. The topology of two-dimensional manifolds can be described exactly by specifying how many handles they have. We can make physical models or projections of non-Euclidean two-dimensional manifolds.

Examples of two-dimensional structures are the curved silicate layers which form cylinders of imogolite and the curved layers of graphite in similar tubules. Graphite layers also may form doubly curved sheets (of positive Gaussian curvature) as Kroto [10] has demonstrated. Certain kinds of sea-weed form sheets with negative Gaussian curvature and various lipids form layers which are periodic minimal surfaces with negative Gaussian curvature. Many protein components form tubes or spherulites rather than crystals. One example of tubes is furnished by a degradation product of haemoglobin.

The mathematics of differential geometry have now entered crystallography in a serious way as may be seen, for example, in the work of Stephen Hyde [1], Sten Andersson, and their colleagues.

6 Three-dimensional manifolds

Three and higher dimensional manifolds are much more complicated. It is not even know whether there is an algorithm for deciding whether two three-dimensional manifolds are the same. The 3-D surface of a 4-D hypersphere provides a useful model of a space of non-Euclidean metric and has been much used.

In addition to real structures, manifolds of various dimensions occur in the description of variables, such as angles, in mechanical systems [21]. We see some promise here for the use of such space for handling the space of the six parameters necessary for the general docking problem (of fitting two molecules together).

7 Textures

The study of textures is now at an interesting stage of relating statistical features to the inter-relationships of the individual units. There are many studies of the symmetries of textures regarded statistically, but detailed structures, based on the interactions of individual particles, are only just emerging. An intermediate stage has been the analysis of singularities for which reference to the book by Maurice Kléman may be made.

Even the simplest textures, those of strained polycrystalline metals, are only just becoming understood [7].

7.1 Centro-symmetry

Centro-symmetry is, in a sense, one of the most general textures. A crystal structure, which has a centre of symmetry, has only centro-symmetrical (cosine) terms in its Fourier transform. The measured amplitudes of the Fourier components thus correspond to phases
of either 0 or $\pi$, and the structure is far more readily solved by the direct methods of Karle and Hauptmann, since the information to be restored is far less $^9$.

8 Conclusion

I regret that I will never be able to apprehend the astonishing book on Gravitation [14] by Misner, Thorpe and Wheeler. However, dipping into it, from back to front, I suspect that it contains many clues as to how solid state physics might develop, working from local order outwards, instead of from infinite perfect crystals as the primary structures. They start from the proposition that "physics is simple only when analysed locally". Newton himself concentrated on the more accessible aspects of the universe and regrettably did not look closely into the consequences of interatomic forces. I recommend more knowledgeable people to try whether they can extract more. I think that we are in a fascinating period where the prevailing paradigm is thinking about the structure of matter is no longer the X-ray crystal structure analysis of orthodox crystallography but has moved to cellular automata and the consequences of the computer in all its generality [8] as a tool, as a concept and as a parallel to living processes.

References


$^9$The German Enigma coding machine was made centro-symmetrical for administrative convenience (coding and decoding procedures were then the same) — in a setting where typing in A gave B, then typing in B would return A. This weakness led to an entry to the decypherment by M. Rejewski, J. Różycki and H. Zygalski, and later others at Bletchley Park, which perhaps ultimately led to military defeat of the Axis. Probably no crystallographers were involved on either side. Enigma, Władysław Kosaczuk, (in Polish, 1976)(in English, 1984)


