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EDITORIAL

Generalizing crystallography: a tribute to Alan L. Mackay at 90

Istvan Hargittai[1]

Abstract Alan L. Mackay, one of the rare generalists of our time, was a disciple and follower of J. Desmond Bernal. Mackay has contributed decisively to the development of the science of structures and taught generations to look at the broader picture when determining crystal and molecular structures. He was constantly seeking coherence and regularities in observations and in thought experiments and was aiming at creating concepts on the basis of those regularities. His inquiries prompted him to predict the existence of regular but not periodic crystal structures that are known today as quasicrystals.

Keywords Alan L. Mackay · J. Desmond Bernal · Generalized crystallography · Quasicrystals · Mackay icosahedron · Birkbeck College

In our preoccupation with finding out how atoms are arranged in space, we are in danger of losing sight of the whole picture.
Alan L. Mackay [1]

Crystallography is not just a scientific specialty, but is a way of life.
Alan L. Mackay [2]

Introduction

My first encounters with Alan L. Mackay (Fig. 1) were in the scientific literature. We met in person for the first time in 1981 in Ottawa during the Congress of the International Union of Crystallography. It was not a glorious occasion: I went up to him, introduced myself, we exchanged a few words; he then turned and left. I was surprised when a few weeks later I received a gracious letter from him that he was happy having made my acquaintance and urged me to visit him whenever I had an opportunity. A great interaction developed, including weeks of stays in each other's homes in Budapest and in London. We organized his first visit to Budapest in September 1982 and he gave three lectures on that occasion at the University of Budapest, including two on various aspects of five-fold symmetry. He said, among other things, that we should be aware of the possibility of extended structures of five-fold symmetry, although these were forbidden by the rules of classical crystallography. If we thought them impossible, they might go by us unnoticed and unrecognized.

By the time Mackay was delivering his talks on five-fold symmetry and issuing his warning about extended structures of five-fold symmetry, and without Mackay knowing about it, Dan Shechtman had already observed the first such extended structures—soon they became known as quasicrystals. Mackay did not merely think and speak about such structures, but he had published papers discussing them, complete with a simulated electron diffraction pattern. When I was listening to Mackay speaking about five-fold symmetry in September 1982, and, increasingly, in hindsight, I felt as if I were present at creation.

The universal importance of five-fold symmetry should not be exaggerated at the expense of other symmetries. However, because classical crystallography exiled it from its considerations as non-crystallographic symmetry, its come-back was all the more spectacular. It was remarkable that two outstanding discoveries in the mid-1980s, both in the science of materials, were related to five-fold

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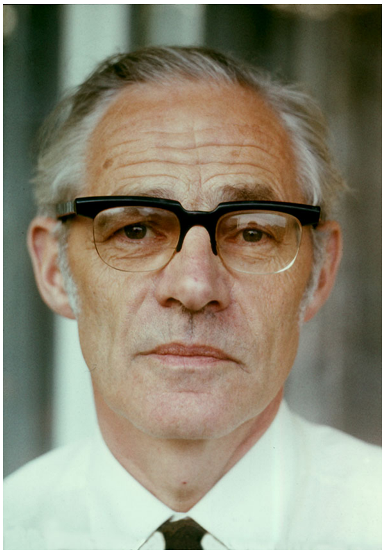

Fig. 1 Alan L. Mackay in 1982 in Budapest (photograph by I. Hargittai)

symmetry. These were the fullerenes and the quasicrystals. Quoting Mackay [3]:

> The main significance of five-fold symmetry for science is that it furnishes us with an explicit example of frustration, which has proved a most fertile concept in the physics of condensed matter. … Neither we nor nature can have everything simultaneously—not all things are possible … We have only the freedom of necessity. 'Nature must obey necessity' as Shakespeare (*Julius Caesar IV:iii*), Democritus, Monod, Bernal, and many others have also recognized. Science probes the limits of necessity and, in the case of five-fold symmetry, has found a corridor that leads us to a new territory.

The beginning

Alan Lindsay Mackay was born on September 6, 1926, in Wolverhampton, England. Both his parents were born in Glasgow. They were physicians, and lived in Wolverhampton in the English Midlands. Alan's father served as an infantry officer in World War I before he became a doctor and as a second in charge of a military field hospital in the Middle East in World War II. Alan's parents ran their own practice in the late 1920s and 1930s, which they sold in 1938. They then became consultants and, especially Alan's mother, served the community in various other capacities dictated by her social conscience. There was always professional talk at their table during their meals, which Alan found exciting. It was also understood that what he heard there could not be retailed outside their home. There were brothers and sisters who eventually dispersed to Australia and America.

Alan started his formal education in a small private school at the age of five, continuing at the Wolverhampton Grammar School from 1935 to 1940. He had to pass an entrance examination at the age of eight to get into this school. His school years overlapped with the Second World War. At the age of thirteen, he was a messenger in the Auxiliary Fire Service. From 1940, he was sent to a boarding school—Oundle School—after he passed another entrance examination. There was talk of a possible German invasion. Alan stayed at Oundle until 1944 and received there an excellent science education. There were difficulties in life during the war, but not in education. His teachers had first class degrees in science and mathematics—teaching was a sought-after profession during and after the Depression. Just to characterize the level of instruction, his chemistry teacher one day demonstrated periodic chemical reactions, today called oscillating or Belousov–Zhabotinsky oscillating reactions. The concentrations of reactants and products undergo periodic changes in such a reaction and they offer a spectacular view if the participants have colors. No such reactions could occur under equilibrium conditions, but they can occur far from the equilibrium. Even twenty years later, Belousov found it difficult to get his manuscript describing such reactions accepted for publication.

School instruction included many demonstrations of experiments and an emphasis on practical applications of knowledge. Alan was a good student and was awarded various scholarships, which eased the financial burden on his parents. But there was never any doubt that he should study regardless of whether or not there were scholarships available. Alan developed an independent mind from an early age and he refers to this as that he was becoming an "internal immigrant."

Early on Alan had acquired a skeptical attitude and later he himself thought about the influences that must have moved him in this direction. He did remember one incident, when he was about five or six, and he told a girl of the same age that her parents had been lying to her over the nature of Father Christmas. In Alan's words, "I was very surprised to find how annoyed people were. It was like Gandhi's or H.G. Wells' experiments with truth. I discovered that you should not believe everything that grown-ups tell you nor say what you actually think. … The tradition of my ancestors was to listen to what authority said and keep their doubts to themselves" [4].

This intellectual disposition of being an internal immigrant was probably strengthened by a predicament of gradually increased difficulty of hearing, which started becoming noticeable from 1955. On the other hand, Alan developed exceptional reading skills in at least half a dozen languages—he has been a voracious reader. He travelled a great deal, especially in Eastern Europe and from 1961, in Asia, including Japan, China, and Korea as well as India.

Start of a profession

Alan L. Mackay (Fig. 2) had earned excellent credentials and in October 1944, he went to Cambridge with a scholarship for the famed Trinity College. He focused on physics and chemistry and studied also electronics, mineralogy, and mathematics. Sir Lawrence Bragg was one of his professors along with other famous scientists, such as the physical chemist and later Nobel laureate R.G. W. Norrish, the physical chemist Frederick Dainton, the inorganic chemist H.J. Emeleus, and others. He won the Percy Pemberton Prize and graduated in 1947.

In the summer of 1947, Alan went with a group of students to Yugoslavia to help build a railway and he has been actively interested in politics ever since. After graduation, in the years 1947–1949, Alan worked in the crystallography laboratory of Philips Electrical Ltd., and, while working for Philips, he earned his BSc degree in physics in 1948 as an external student. He decided to study for his PhD and he joined Birkbeck College of London University, and he has stayed at Birkbeck for the rest of his professional life. First he was there part-time, from 1949, in the crystallography laboratory of J. Desmond Bernal (1901–1971), later moving to full time. He defended his PhD thesis and was awarded the degree in 1951.

Mackay learned Russian in summer school, and there, he met Sheila, his future wife (Fig. 3). They married in 1951 and by 1961 they had three children, two boys and a girl, and moved to their home in North London where they stayed ever since.

Already by then, Alan's interests were broad and he published more broadly than would someone with a narrow specialization. This did not help his promotion in the university ranks. In this he followed his mentor's example although he learned also from Bernal that for his career broad interests counted as a disadvantage. Alan would be awarded his DSc degree in crystallography and studies of science in 1986. He was appointed Professor of Crystallography in the same year and became Professor Emeritus in 1991. In 1988, he was elected Fellow of the Royal Society (FRS).

Bernal's example was an inspiration for Alan ever since he had chosen Bernal's book, *The Social Function of Science*, as his prize for winning a competition in Cambridge. It would be difficult to imagine an environment more conducive to developing a generalist approach to science, and, in fact, to life, than Bernal's circle. Bernal was nicknamed "Sage" for he was supposed to know everything worth knowing. In the 1930s, Bernal was a member of the Club for Theoretical Biology, along with Joseph Needham, C. H. Waddington, and others. They dealt with such questions as the application of X-ray crystallography and other physical techniques to solving problems in biology. Already in the mid-1930s, Bernal had shone X-rays onto protein molecules and the fact that he could record interference patterns led him to believe that the structures of such large biological systems could be solved on the atomic level. Bernal was good in delegating tasks and he delegated the structure determination of large biological molecules to such disciples as the future Nobel laureates Dorothy Hodgkin, Max Perutz, and Aaron Klug. Bernal served as science advisor at the highest level during World

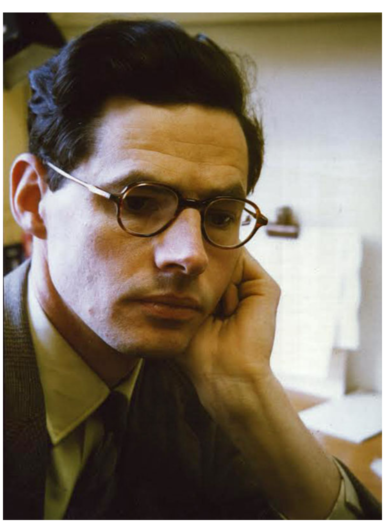

Fig. 2 The young Alan L. Mackay (courtesy of Robert H. Mackay)

Fig. 3 Alan and Sheila Mackay around 2000 in front of their home in London (photograph by I. Hargittai)

War II. After the war, his communist politics and friendship with the Soviet Union were a serious impediment to his obtaining support for building up a research center that would have been adequate for implementing his far-reaching research ideas.

J. Desmond Bernal (Fig. 4) collected around him an excellent group of scientists in mathematics and computing, in the theory and experiment of X-ray crystallography, physical chemistry, both inorganic and organic structures, and his laboratory ran a skilled workshop. A stream of international visitors complemented his staff. Scientists like Norbert Wiener, Linus Pauling, André Lwoff, and H.S.M. Coxeter came and so did representatives of world culture, like Picasso and Paul Robeson. Bernal's associates felt they were "living in the center of the universe" [2]. Mackay realized from the start how privileged it was to be part of Bernal's circle of his closest associates. The combination of scientific, social, and political activities appealed to Mackay's own inclinations.

In 1956, Bernal invited Mackay to accompany him to Moscow. Bernal gave lectures on the origin of life at Aleksander I. Oparin's institute. Mackay had the opportunity to meet such giants of Soviet science as Petr L. Kapitza, Lev D. Landau, Igor E. Tamm and Vladimir A. Fock (of Hartree-Fock fame). Bernal and Mackay visited the Institute of Crystallography of the Soviet Academy of Sciences and met its director, Alexey V. Shubnikov and Shubnikov's co-workers, among them Boris K. Vainshtein and Zinovii G. Pinsker (Fig. 5). Mackay had already begun building up an international network of friends, especially with crystallographers at international meetings, and his interactions with the Moscow crystallographers were especially active. In 1962, he spent five months at the Institute of Crystallography in Moscow. Scientifically it was not a very fruitful stay, but for getting to know many colleagues and Soviet life, in a more realistic way than from propaganda materials, it was.

Research

Mackay's first research project was the structure analysis of a particular modification of solid calcium phosphate used in fluorescent tubes, which was of interest to Philips. The company had an array of various projects involving X-ray crystallography related to practical applications. When Mackay moved to Birkbeck College, he continued doing research on inorganic materials. He joined the section whose major concern was the properties of cement. When Bernal was at a committee of the Ministry of Works, he volunteered that he could find out why cement sets, and a whole research project developed from this assertion.

Icosahedral structures became the focus of Mackay's interest rather early. He had already met with the structure of beta-tungsten at Philips. Then, he found some interesting old papers at Birkbeck, evidence that there had been interest in these structures at the College before Mackay. Bernal also considered the icosahedral arrangement rather early, because it would prevent crystallization, and he thought that icosahedral coordination might give some clues to understanding the structure of liquids. Mackay was also aware of Pauling's interest in icosahedral structures. When Bernal was to go to Budapest to give a talk at the meeting honoring Zoltan Gyulai's 70th birthday, he asked Mackay to draw the figures. Bernal's talk was about the

Fig. 4 J. Desmond Bernal about 1960 in London (photograph by and courtesy of Alan L. Mackay)

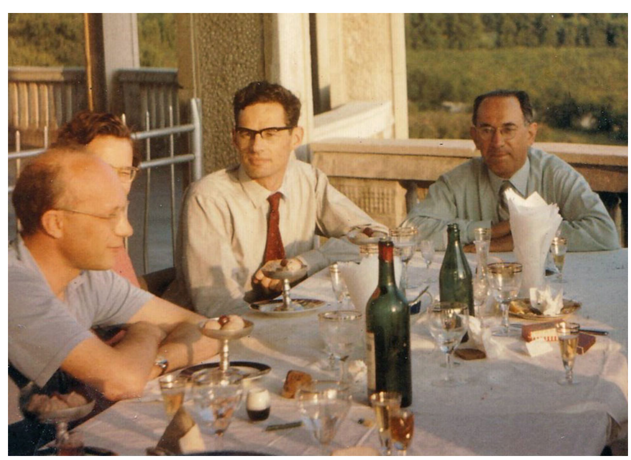

Fig. 5 Alan L. Mackay (in the middle) in the company of Boris K. Vainshtein (left) and Zinovii G. Pinsker (right) in 1962 in Moscow (courtesy of Alan L. Mackay)

symmetry in solids and liquids. It was a most comprehensive presentation [5].

The icosahedral arrangement of atoms is interesting because it could also be a step in the progression from the isolated molecule to an extended structure. When a second icosahedral shell surrounds an icosahedron of 12 spheres about a sphere in the center, the size of this second shell is exactly twice the size of the first shell [6]. This second shell contains 42 spheres and lies over the first so that spheres are in contact along the five-fold axis. Further layers can be added in the same fashion.

The third layer is shown in the Figure and this is known as the Mackay polyhedron (Fig. 6) or Mackay icosahedron—an example of icosahedral packing of equal spheres. The layers of spheres succeed each other in cubic close packing sequence on each triangular face. Each sphere which is not on an edge or vertex touches only six neighbors, three above and three below. Each such sphere is separated by a distance of 5% of its radius from its neighbors in the plane of the face of the icosahedron. The whole assembly can be distorted to cubic close packing in the form of a cuboctahedron. The Mackay icosahedron has "made tremendous impact on particle, cluster, intermetallics, and quasicrystal researchers…," [7] according to the late K.H. Kuo, the doyen of Chinese crystallographers. Kuo identified two basic concepts in Mackay's paper. One was the icosahedral shell structure consisting of concentric icosahedra displaying five-fold rotational symmetry. This structure occurs frequently and not only in various clusters, but also in intermetallic compounds and quasicrystals. The other concept, according to Kuo, was the hierarchic icosahedral structures due to the presence of a stacking fault in the face-centered-cubic packing of the successive triangular faces in the icosahedral shell structure.

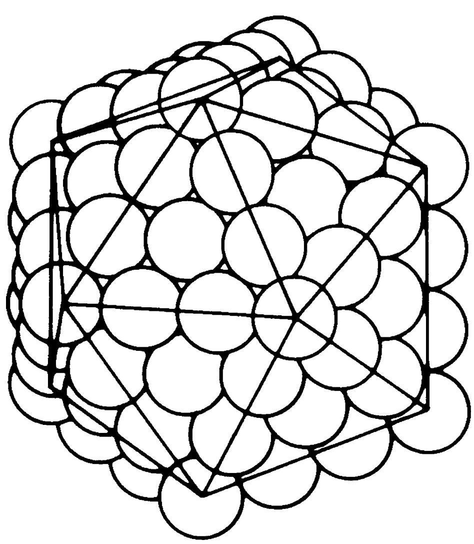

Fig. 6 The "Mackay polyhedron" emerging from the icosahedral packing of equal spheres. Only the third shell is visible (courtesy of Alan L. Mackay [6])

Mackay questioned dogmas wherever and whenever he met them. This was especially so in the case of crystallography where the classical rules had worked so well but eventually proved increasingly to be limiting the scope of structures the subject embraced. Those rules limited the inclusion of novel kinds of structures that kept emerging as well as structures that had been abandoned by crystallographers; but the need arose to include them in a broader system. There was an obvious deficiency when the theoretical constraints of crystal symmetry were confronted with real crystals in that crystals are not infinite. The approach to discussing crystal symmetry used to be to think of the formation of a crystal through insertion of individual atoms or groups of atoms into the three-dimensional framework of symmetry elements, whereas in reality—as Mackay liked to point out—the symmetry elements emerge as a consequence of the structure being formed through the local interactions between individual atoms or other building elements. The concept of crystal symmetry itself became a target of Mackay's inquiry and he creatively deepened and expanded its meaning. When I asked him if he would like to select one of his papers for inclusion in the current special collection of articles, he chose the one titled "Crystal Symmetry"[8] reproduced in the "Appendix".

Mackay compiled a list of concepts in two versions, showing the transition from the classical to the modern (Table 1). He has refined his list over the years, but the 1981 one demonstrates from a 35-year perspective how forward-looking his ideas were.

This list appeared in a paper, which Mackay titled *De nive quinquangula* (on the pentagonal snowflake), which was a direct reference to Johannes Kepler's treatise on the six-cornered snowflake [10].

There were several threads in Alan's career that were rapidly coming together. In his words [11]:

> I used to do science abstracts—for ten years I abstracted all the Russian papers on crystallography—and I remember abstracting a paper on the incommensurate arrangements of spins in iron oxides, in hematite. The period of the helical magnetic spin is not the same as the crystallographic period. So incommensurate structures were current before that time. Even much longer before that I thought of a simple thing about printing wall paper. Suppose your wall paper is simply printed from a roller. But suppose you are printing two motifs from two rollers of different diameter. Then you get a non-repeating pattern. I wasn't able to think of producing an aperiodic two-dimensional pattern in this way. I was only aware of the possibility of one-

Table 1 Mackay's compilation of classical versus modern concepts in 1981 (courtesy of Alan L. Mackay [9])

Classical concepts	Modern concepts
Absolute identity of components	Substitution and nonstoichiometry
Absolute identity of the environment of each unit	Quasi-identity and quasiequivalence
Operations of infinite range	Local elements of symmetry of finite range
"Euclidean" space elements (Plane sheets, straight lines)	Curved space elements. Membranes, micelles, helices. Higher structures by curvature of lower structures
Unique dominant minimum in free energy configuration space	One of many quasi-equivalent states; metastability recording arbitrary information (pathway); progressive segregation and specialization of information structure
Infinite number of units. Crystals	Finite numbers of units. Clusters; "crystalloids"
Assembly by incremental growth (one unit at a time)	Assembly by intervention of other components ("crystallise" enzyme). Information-controlled assembly. Hierarchic assembly
Single level of organization (with large span of level)	Hierarchy of levels of organization. Small span of each level
Repetition according to symmetry operations	Repetition according to program. Cellular automata
Crystallographic symmetry operations	General symmetry operations (equal "program statements")
Assembly by a single pathway in configuration space	Assembly by branched lines in configuration space. Bifurcations guided by "information", i.e., low-energy events of the hierarchy below

> dimensional incommensurate patterns. I was really interested in hierarchic patterns and not in aperiodicity as such. It came directly from Bernal's suggestions and the polio virus project. I produced a hierarchic pattern, a hierarchic packing of pentagons. Then in 1974 I was getting some help in computing from Judith Daniels at the University College Computing Centre and, incidentally, showed her these patterns. She said that Roger Penrose had something like them. So I made an appointment with Roger Penrose [(Fig.7)] and Robert, my son, and I went to see Penrose in Oxford, and he showed us the jigsaw puzzle, with the kits and darts and so on. Basically his concern was with forcing aperiodicity, and my concern was with hierarchic structures. It turned out to be very similar.

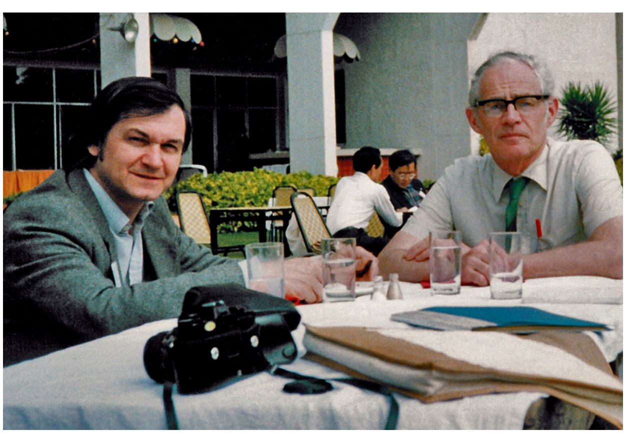

Fig. 7 Roger Penrose and Alan L. Mackay (courtesy of Alan L. Mackay)

In the paper about the pentagonal snowflake, Mackay, à la Penrose, built up a regular, but non-periodic (he called it then "noncrystalline") structure from regular pentagons in a plane (Fig. 8).

It starts with a regular pentagon of given size, which we may call the zeroth-order pentagon. Six of these pentagons are combined to form a larger regular pentagon, the first-order pentagon. There are triangular gaps in this pentagon and Mackay filled these gaps with pieces from cutting up a seventh zeroth-order pentagon. This cutting up yielded five triangles and a smaller regular pentagon as left-over, which is the pentagon of the order of -1. This design is repeated on an ever increasing scale.

After the meeting with Penrose, Alan's son Robert went back to his university at York where he was studying computer science and plotted a tiling on his pen-plotter (Fig. 9). We could call what he plotted a Mackay tiling as it was different from the standard Penrose kites and darts. Robert (Fig. 10) started from

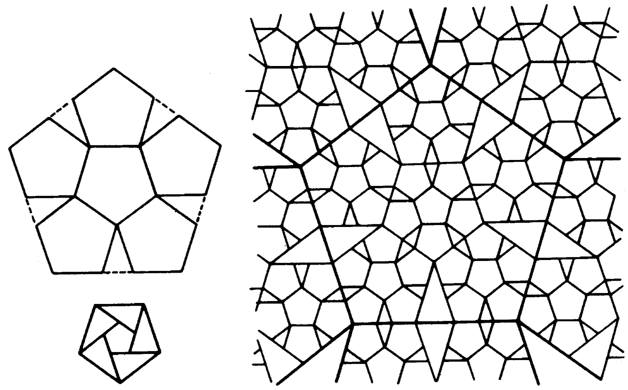

Fig. 8 Tiling with regular pentagons (courtesy of Alan L. Mackay [9])

pentagons of a certain size and as he kept going to larger and larger pentagons, he built up a pentagonal snowflake. Mackay included Robert's design in his paper on pentagonal snowflakes to give his considerations added emphasis.

Mackay was getting ready to make significant predictions concerning the possibilities of real three-dimensional structures with five-fold symmetry. At one point, he got the idea of producing a simulated diffraction pattern of the Penrose tiling [11]:

> First I just drew the Penrose type pattern and sent it to George Harburn in Cardiff who was a colleague of Charles Taylor who had a good optical diffractometer. I had stuck it into a laser beam here but you need a precise adjustment. You can do many beautiful things with the optical diffractometer that you can't see in the computer, with very fine detail; it is amazing. Then George Harburn made a second version which instead of consisting of lines, had dots; thus the diffraction pattern was not dominated by the streaks from the lines ([11], p 154)

Mackay wrote up and published his paper in which he communicated a simulated diffraction pattern (Fig. 11) [12].

It is remarkable, how, once again in a broader context, he was considering the characteristics of the pattern and the diffraction it generated [11]:

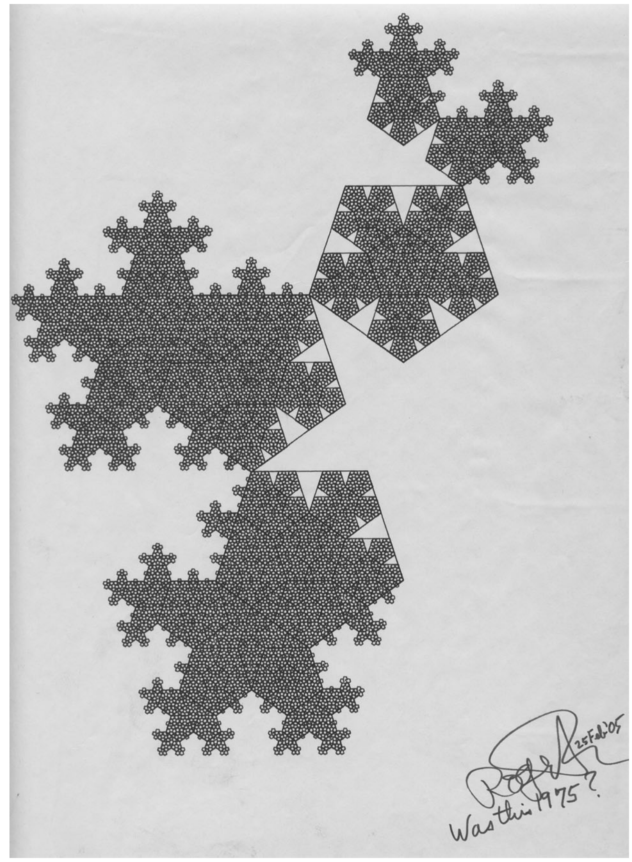

Fig. 9 Robert H. Mackay's computer drawing of the formation of a "pentagonal snowflake" in 1975 [9] autographed by Roger Penrose in 2005 (courtesy of Robert H. Mackay)

Fig. 10 Alan L. Mackay and Robert H. Mackay in April 2016 in London (photograph by and courtesy of Magdolna Hargittai)

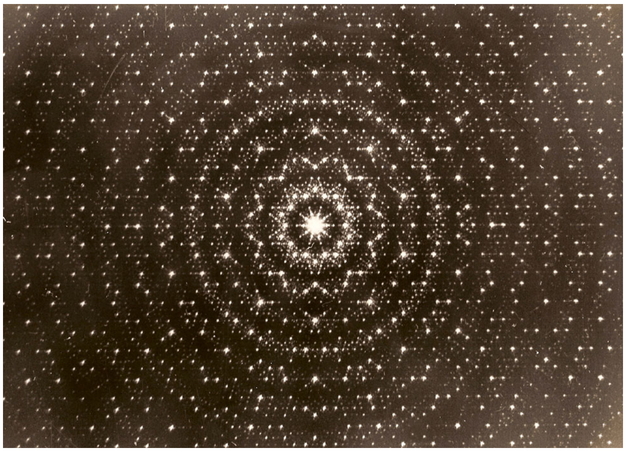

Fig. 11 Mackay's simulated "electron diffraction" pattern of a three-dimensional Penrose tiling (courtesy of Alan L. Mackay) [12]

> I had also a theory about collagen, and had some patterns bearing on that. The theory was that collagen fibers are connected with the Fibonacci spiral. If you draw a Fibonacci spiral of circles along the spiral, then locally the pattern keeps changing between square packing and hexagonal close packing. This corresponds closely to the diffraction you infer from collagen fibers. Richard Welberry in Canberra, Australia, had a still better optical diffractometer and took some very good diffraction pictures from the Fibonacci spiral. Then [the botanist] Eriksson in Philadelphia showed that the diffraction pattern of the Fibonacci spiral was self-similar to the Fibonacci spiral itself. … This may point to a connection between phyllotaxis—the scattered leaf arrangement about stems—and internal structure on the atomic level" ([11], p 155).

Alan's story is a brilliant example of the importance of pursuing a lot of lines in research and look for their possible convergence. In this, Mackay followed Bernal's philosophy of asking a thousand questions rather than just one, because this way the probability of finding answers is greatly enhanced. Along the way, Mackay documented his findings. This was useful, because after the publication of Shechtman's experimental observation of quasicrystals in November 1984 [13], theoretical/modeling papers followed in rapid succession [14]. It could have been easy to distort the real succession of events related to the circumstances of the discovery. Indeed, one-sided reports did appear. For example, an account in one of the January issues of *The New York Times* stressed the priority of theoretical work, but failed to mention Mackay's modeling and simulation studies and even downplayed the experimental discovery itself. This prompted me to send in a "Letter to the Editor" in which I described Mackay's contributions, explicitly citing his two publications (*Physica* 1982, 114A:609–613 and *Soviet Physics Crystallography* 1981, 26:517–522). As far as I know the letter was not printed but it is well documented ([11], pp 171–172).

Mackay recognized the potential practical applications of quasicrystals early on. He thought that Shechtman's discovery may very well be more important than it had been believed. He recognized that the low thermal conductivity of quasicrystals may be utilized for nonstick frying pans, turbine blades, in internal combustion engines, and so on. A suitable technology might be able to create quasicrystal surfaces by glazing metal with a laser. He foresaw great economic potential in the discovery.

Alan told me about this when I asked him about Shechtman's possible Nobel Prize, back in 1994. He had an interesting line of thought about the different kinds of Nobel Prize as he saw them. He characterized Shechtman's discovery as when someone turns over a stone and finds something truly important, maybe like superconductivity or the scanning tunneling microscope or the Mössbauer effect. There isn't an enormous amount of work but someone was in the right place at the right time, and recognized what he's found. In 1994, Mackay thought that Shechtman's Nobel Prize would come in this category.

The only reservation Mackay had in evaluating the importance of the discovery of quasicrystals was that it may have appeared more significant than it really was. He thought that the too restrictive definitions of classical crystallography lent a pivotal character to the discovery. Had the definitions of classical crystallography been broader and more inclusive, there would have been no need to bring about a paradigm change. However, as it happened, the discovery of quasicrystals did prove to be pivotal and it did bring a paradigm change about.

Mackay had truly predicted the existence of regular but non-periodic structures that Dan Shechtman (Fig. 12) then observed in his experiments. It would have been a wonderful sequence of events had Shechtman and others known about Mackay's prediction and have embarked on looking for such structures and found them. The search for extended structures with five-fold symmetry had been going on for centuries and involved excellent minds, such as Johannes Kepler and Albrecht Dürer. Roger Penrose came up with such a pattern in two dimensions and Mackay crucially extended it to the third dimension, and urged experimentalists to be on the lookout for such structures. Nobody took up his challenge and when Shechtman made his observations, he was not aware of Mackay's predictions. Eventually though all these lines came together. In 2010, the American Physical Society awarded the Oliver Buckley Prize to Alan Mackay, jointly with Dov Levine and Paul Steinhardt for their contributions to the

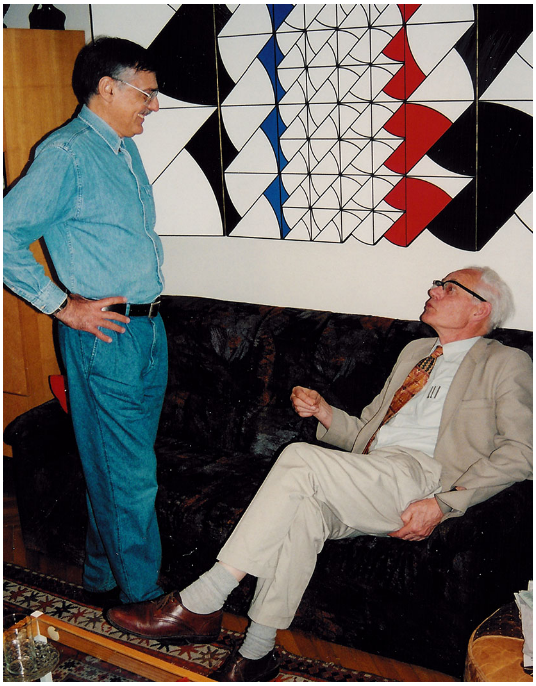

Fig. 12 Alan Mackay and Dan Shechtman in 1995 in the author's home in Budapest (photograph by I. Hargittai)

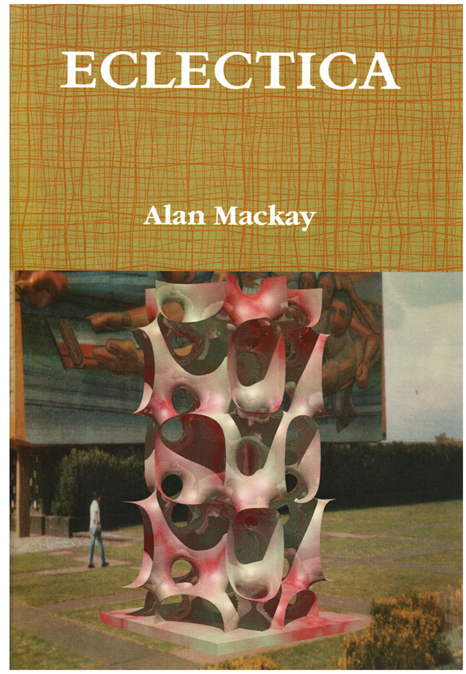

Fig. 13 The cover of Mackay's *Eclectica*. The art is a computer-creation by Alan L. Mackay, one in a long series of images inspired by his studies of minimal surfaces (courtesy of Alan L. Mackay [15])

quasicrystal discovery. The next year Shechtman received the Nobel Prize in Chemistry.

Summing up

Alan does not mind the adjective once applied to him by a colleague, "the well-known eclectic," and chose this word for the title of a selection of his writings, *Eclectica*, self-published for personal use in a handsome volume in 2009 (Fig. 13) [15]. In it, he reproduces many of his published papers and communicates a number of unpublished works as well. The volume is a rich source of information and ideas and here we will merely dip into it for a few selected entries to illustrate its scope and depth.

Appropriately the volume begins with a discussion of copyright—one of Mackay's pet projects. He has been an advocate of protecting the rights of scientist authors to their own intellectual productions versus the publishing companies. One of the solutions he found promising was for professional societies to start their own electronic journals with open access that would be supported by authors' fees. Currently the open access approach is gaining ground rapidly, but there may be a great divide between authors who can and those who cannot afford the often hefty fees for having their manuscript published in open access venues.

As we have seen above, the discovery of the Mackay polyhedron and his prediction of the structures today called quasicrystals, did not happen in isolation. Mackay had long been interested in structures that fell beyond the rigorous and confined system of classical crystallography. He has published at least three reviews under the title "Generalized Crystallography," the latest in 2002 [16]. He defined the aim of generalized crystallography as "to understand the properties of matter, inert and living, at our human scale, in terms of the arrangement and operation of atoms." He recognized the pioneering role of X-ray crystal structure analysis in this quest, but noted that as the array of techniques has become vast, it might be advisable to replace the term crystallography by *structural chemistry*. He also realized though that terms that had been embedded long in scientific literature would be hard to displace. This may be so unless the new term is glued to a fad as, for example, in the case of nanoscience and nanotechnology.

Concerning the pioneering role of X-ray crystallography, Mackay has written about the phenomenon of when a pioneering field becomes a brake on further progress. This happened with classical crystallography whose rigid system hindered the recognition of those structures that fall beyond this classical system. In short, its success became a barrier to progress. Of course, for this, blame should not be assigned to those who originally worked out the system, but it is our task to overcome the barriers that have been erected by the developments since. This kind of success turning into a brake is not unique to classical crystallography. When insulin was discovered for treating diabetes it was a great triumph of the biomedical sciences. It has then been gradually recognized that the availability of this successful treatment, which is not a cure, might have diverted efforts and resources from continuing a quest for the cure of diabetes. Another example from the science of structures was the resistance to recognizing other techniques against the background of the enormously successful X-ray diffraction making it harder for electron crystallography and for neutron crystallography to become accepted and spread [17]. However, Mackay's teachings on generalized crystallography fell onto fertile ground; suffice it to mention a couple of additional contributions to the volume of *Structural Chemistry* dedicated to his 75th anniversary [18, 19].

Mackay's impact on the structural science community is hard to measure, but the impression is that it will be long lasting. He has impacted us through his writing and through personal interactions. In this connection it is notable that he adapted himself easily to local conditions on the occasion of his many visits. When he spent a longer period at the Institute of Crystallography in Moscow, he developed the habit of carrying a shopping bag with him. This was not only because the shops did not give out such bags to carry away their goods; but even more because one never knew what purchase might suddenly become available. After his return to London, he did not find it easy to give up the habit of having his shopping bag at readiness. Although his stay at the Institute of Crystallography in Moscow did not produce scientific results, his interactions with the Azerbaijani crystallographer Khudu Mamedov (1927–1988) greatly helped Mamedov to become well known in the West. Mamedov prepared periodic drawings that were reminiscent of Escher's patterns, but he used historical/cultural motifs from his region. Thus he created a unique interrelationship between art and science. Mamedov, perhaps in Mackay's style, used the term "crystallographic" in a broad sense. Mackay dedicated a talk to Mamedov's memory in 1991, "Form and pattern in Azerbaijani civilization," and its text is reproduced in *Eclectica*.

Mackay (Fig. 14) and Bernal co-authored a presentation entitled "Towards a science of science" for the 11th International Congress for the History of Science in Warsaw in 1965. They outlined what Science of Science was, why it was needed and the methods of their inquiry. Their program included practical recommendations, such as the establishment of departments of the history of science and the need for looking at science as a whole rather than always taking up merely its specificities. Further, they called for establishing the profession of science critic similarly to that of literary critic, and called for

Fig. 14 Alan L. Mackay in 2011 in his study among many of his computer-generated drawings (photograph by I. Hargittai)

international cooperation as recognition of science as a world-wide activity. They also suggested experimental work in order to find the best means of science training and the like. They emphasized the importance of learning about non-European cultures where emphases were different from European cultures as illustrated, for example, by a lower priority for written records, but a higher one for master-pupil relationships. This joint Mackay-Bernal presentation has been reproduced in a number of publications and in a number of languages, yet it is not easily accessible. Hence, it is very useful to have it in *Eclectica*. Mackay co-edited a volume on this topic and the idea of science of science permeated his activities throughout his entire career [20].

In the early 1980s Mackay ran a column called "Anecdotal evidence" in the journal *The Sciences* and the entries are reproduced in *Eclectica*. It suited him eminently, bringing together seemingly disparate ideas and facts. Even the titles reveal some aspects of his approach, such as "Science and Travel," "Rhyme and Reason," "How to write a best seller," "Mackay's *Michelin*," "Molecules and Moores" (referring to Henry Moore), "Message in a Bottle," and suchlike. The column served the readers of this unusual periodical well, but its editors liked to smooth over his often unorthodox style of writing; apparently the flavor of Mackay's writing was a little too much for them.

The *Eclectica* volume is concluded by a list of Mackay's work, including scientific publications (176 entries), miscellaneous publications (130), and book reviews (46). There is then a list of 30 unpublished papers, and 10 entries which he calls "indirect material," and those publications by others in which he figures, including the special issue in *Structural Chemistry* in 2002 dedicated to him [21].

Legacy

The full story of the quasicrystal discovery has yet to be written. At this point, I am offering my thoughts concerning only a tiny aspect of this story, viz. the demeanor of its principal protagonists with respect to the loneliness of the scientific discoverer. With justifiable simplification, there were three protagonists in this story. Alan L. Mackay predicted the existence of quasicrystals. Dan Shechtman discovered them in his experiments. Dov Levine (a graduate student, then) and Paul J. Steinhardt (Levine's professor) coined the name quasicrystals and offered a theoretical interpretation of the structure of this new kind of matter. I have had opportunities of discussing the circumstances of their discoveries in person with Mackay, Shechtman, and Levine. I interacted with Steinhardt only via e-mail exchanges.

Alan L. Mackay's (Fig. 15) demeanor has been such that he was looking consciously for dismantling dogmas and scientific taboos. In doing so, he realized the indefensibility of the dogma of classical crystallography with respect to the prohibition of five-fold symmetry in extended structures. Once he recognized this, he voiced it in his publications and in his presentations. He did not have second thoughts about making a stand and risking his reputation. He did this at a time when there was a reduction of personnel at British universities and he could have been retired prematurely. In 1982, he was 56 years old, had been a Reader in Crystallography at Birkbeck College for quite some time. It would only be in 1986 that he was awarded a personal chair as Professor of Crystallography and was elected FRS in 1988. It seems that the loneliness of the scientific discoverer was his natural mode of existence.

Fig. 15 Alan L. Mackay and Istvan Hargittai in April 2016 in London (photograph by and courtesy of Magdolna Hargittai)

In contrast, Dan Shechtman was not looking to do anything revolutionary. His interest in alloys was in finding compositions for improved practical applications. However, he possessed a good deal of curiosity and this was his driving force at his pre-discovery stage. This curiosity made him embark on testing metal compositions that could have not been expected to offer improved, or any, applications. Once he made the discovery and realized that it was revolutionary, he grew to the challenge and his stubborn nature helped him to see it through to general acceptance. In doing so, he invited the disapproval, even wrath, of the powers that be in science, for example that of the greatest chemist of his time. Shechtman conducted himself with dignified determination in his loneliness, but he was not enjoying it and welcomed any easing of this loneliness. He felt relief and gratitude when Ilan Blech joined him in co-authoring the first paper in which Shechtman—half-heartedly and half-buried among other materials—mentioned his discovery. When he was finally preparing the manuscript that reported unambiguously his discovery, he was happy and grateful that he found three co-authors who helped him formulate what he wanted to say and who eased his loneliness of the scientific discoverer.

Levine and Steinhardt were ready to publish their interpretation of the quasicrystal structure as soon as they had learned about the paper reporting its experimental observation. At this point, they did not have to face the loneliness of the scientific discoverer, because that burden had already fallen onto Shechtman, let alone Mackay. Had Levine and Steinhardt come out with their theoretical model before the experimental discovery, they might have felt the most acute loneliness, possibly even ridicule. From the immediacy of their publication following Shechtman's, we may suppose that they might have made their theoretical discovery some time before. Levine might have written a good thesis even on the basis of a failed model, but for Steinhardt, the risk would have been considerable and possibly sufficient to damage his reputation. He was a Professor of Physics at the University of Philadelphia, then; later on, at Princeton University. Steinhardt (not Levine) had the choice of taking the risk and face the loneliness of the discoverer or wait and see whether there might be a safer opportunity to strike out.

All this is my supposition only, but I see consistency with it in how things played out during those years in the second half of the 1980s.

The adjectives "consistent" and "rational" are among the many characterizing Alan L. Mackay, and they shine through the poem he composed recently that sounds like a parting gift:

Atoms and our Vision of the World
There are no gods.
We are alone.
I am thus two-fold alone
but I have the second sight of science.
As my eyes grow dim,
my mind sees the future.
I see a hand writing on the wall -
the wall surrounds a giant alembic -
built to win gas from coal.
The Chinese hand wrote large
the character which stands for entropy.
It questions the solid state of Earth.
Asking my computer, I find the words
"disordered hyperuniformity"
- today's myopic Vision of the World
glimpsed in the microcosm of atoms.
Death came to my wife of more than sixty years
Her flame went out. Her body was cremated -
Atoms to atoms – Lucretius saw truth.
But where is past history now?
Information increases locally from time to time -
but Entropy will win.
A.L.M. 30 August 2015 [22]

This was not the first time Mackay had expressed his views and sentiments through poetry. His published poems often express topics in crystallography and the science of structures [23]. He titled his collection of poems published in 1980 the Floating World after the works of Japanese artists who lived in the latter half of the 18th century and the first half of the 19th.

According to Mackay, "Scientists inhabit a kind of Floating World of their own, a kind of Global Village, in which they have friends, or friends of friends, everywhere. Rather, like members of a religious order, they can go to any laboratory dealing with their field of study and be hospitably received" [24]. Alan and Sheila Mackay certainly practiced this very hospitable attitude toward many members of the international scientific community.

Mackay has been much concerned with the ways to expand the science of structures to embrace systems with varying degrees of regularity. Here intentions and desires that cannot be formulated yet with exactitude can be expressed as a poem [25]:

We cruise through the hydrosphere
Our world is of water, like the sea,
But the molecules more sparsely spread,
Not independent, not touching
But somewhere in between,
Clustering, crystallizing, dispersing
In the delicate balance of radiation
And the adiabatic lapse rate.

Even when he is composing prose, it sometimes sounds like poetry. Consider this example: "Amorphous materials may be shapeless, but they are not without order. Order, like beauty, is in the eyes of the beholder. If you look only with X-ray diffraction eyes, then all you see is translational order, to wit crystals. … [T]here is a wide range of structures, between those of crystals and those of gases, … Other structures need not be failed crystals but are *sui generis*"[26]. (Italics in the original)

Contemplating Alan Mackay's legacy, it is often said that scientific discoveries, however important, are sooner or later overshadowed by new developments in science. So it is happening with Mackay's contributions to crystallography and the science of structures. However, his demeanor as a researcher and scientific discoverer will serve as inspiration for a long time.

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Appendix

Alan L. Mackay, Crystal Symmetry, *Physics Bulletin* November 1976, published by the Institute of Physics.

A L MACKAY

CRYSTAL SYMMETRY

How can we develop a unified way of dealing with structures covering the range from diamond, naphthalene and iron to living objects? Dr Mackay suggests that 'cellular automata', which show aspects of reproduction, growth and evolution, may provide the answer

E T Bell, the historian of mathematics, has castigated the formalism of Euclidean geometry by saying: 'The cowboys 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 the hog-tie and it is what Euclid did to geometry'. The formalism of the *International Tables of Crystallography* is similarly restrictive. The 230 space groups characterize exhaustively all the symmetries possible for infinite lattice structures. However, crystals are not infinite and there are some which do not have regular three-dimensional lattices. The lattice results from the relationships between the parts of the crystal and not vice versa.

The crystal does not form by the insertion of components into a three-dimensional framework of symmetry elements. It arises from the local interactions between individual atoms and the symmetry elements are a consequence. We must observe that the traditional formalism of crystallography handles occurrences of pseudosymmetry and local symmetry elements rather inadequately. Our argument is that a regular structure should mean one generated by simple rules and that the list of rules regarded as simple and 'permissible' should be enlarged. These rules will not necessarily form groups.

With the recent immense progress in examining biomolecules by crystallographic and other imaging methods, the desirability of developing a unified way of dealing with spatial structures (and their changes, growth, evolution and transformations) which will cover the range from diamond, naphthalene and iron, to recognizably living objects (such as bacteriophages) steadily increases. It is argued here that concepts of cellular automata, having their origins in computer theory, and now being elaborated for the purposes of developmental biology, provide a suggestive ideology for recasting crystallographic ideas so that they can be carried forward.

Cellular automata

Cellular automata are known by a variety of names, not exactly synonymous, such as local programme, tessellation automata, abstract computational devices, transition rules, formal languages, etc. They derive from the Turing machine, formulated first in 1936, which was a conceptual device which notionally could read and write binary digits on a paper tape. Turing investigated the conditions under which a finite part of an infinite sequence could be generated by the machine when it was provided with a finite set of instructions in the same form. This led to von Neumann's theory of self-reproducing automata, which was later seen to be realized in the genetic code.

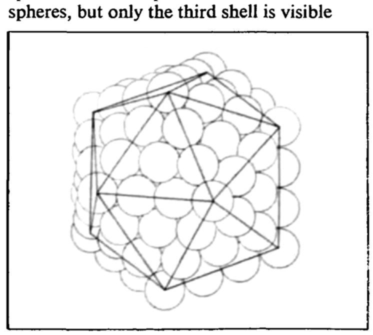
In cellular automata a coordinate system is assumed and the programme is executed at every cell in it simultaneously, and not sequentially as in the single arithmetic unit of the typical computer. The best-known example is J H Conway's game 'Life' where a configuration of crosses is marked in the cells of an infinite chessboard. Transition rules then specify the configuration for the next interval of time. These rules are applied simultaneously to all cells. Picturesquely stated, these rules are: (i) each cell has eight neighbouring cells, (ii) if a cross has more than three neighbouring crosses it dies of overcrowding, (iii) if it has two or three neighbouring crosses it survives for the next generation, (iv) if it has less than two neighbours it dies of loneliness and (v) if an empty cell has exactly three neighbouring crosses then a cross is born there.

Cellular automata, such as Conway's game, show an immense variety of behaviour, suggesting aspects of reproduction, growth and evolution which are encountered in systems of real cells. They stress the kinematic and configurational, rather than the dynamic or energetic features of a system, and deal with information rather than energy. They are thus more apt for the modelling of very complex systems, with large numbers of local minima in the free energy landscape, where the system cannot easily 'find' the lowest minimum. Metastability is enough.

The cellular automaton consists of (i) a space, (ii) an initial configuration, (iii) an alphabet of components, (iv) a grammar of syntactic rules, and (v) a transition scheme for applying these rules. Given the state at one instant, the state at the next can readily be found and thus, by repeated induction, the behaviour of the system in time (or space). The rules may depend only on the state of the one cell to which they are being applied or they may involve the states of other cells in more complex ways. They may be snatches of programme containing, for example, conditional or probabilistic statements.

Even a simple rule can give quite interesting consequences. For example, we can take a linear cellular automaton consisting of a sequence of binary digits. These will be zeros or ones, but could have some physical representation as, for

Figure 1 The icosahedral packing of equal spheres. In all the particle contains 147 spheres, but only the third shell is visible



example, the sequence of layers in some polytype such as silicon carbide. Starting with an initial sequence of four digits, the next digit is generated by comparing the previous digit with the digit three spaces earlier (i.e. skipping over two). If these digits are the same, then the next digit is a zero; if they are different, then the next digit is to be a one. This rule results in a structure of period 15. The same sequence results whatever the seed (the initial four digits). Thus, from the seed 1111 we get 1111/010110010001111/010110010001111/ . . . etc. If we jump over four digits instead of two, and compare the previous digit with the one five spaces earlier, we get a structure of period 63. This application of a simple transition rule shows how long range structure may arise from short range atomic interactions.

We could apply this approach to the description of polytypes, for example, the hexagonal ferrites such as the compound $Ba_{70}Zn_{66}Fe_{444}O_{802}$ which has a period of 1455 Å and where the atomic interactions are still of only the normal range. The unit cell of this compound contains more than 4100 atoms and the structure was solved by identifying the stacking sequence of subunits by electron microscopy. In such cases the high degree of pseudo-symmetry makes the conventional space-group characterization an uninformative description, 'uninformative' meaning 'of low information content'. In fact, *M* and *Y* blocks are recognizable and these are stacked in the sequence $MY_6MY_{10}MY_7MY_{33}$ which gives the very long period. *M* is a five-layer block $BaFe_{12}O_{19}$ and *Y* is a six-layer block $Ba_2Me_2Fe_{12}O_{22}$. Each layer contains four oxygen atoms or three oxygen atoms plus one barium atom per hexagonal cell.

How can we describe the traditional perfect crystal in terms of this model? The asymmetric unit is repeated by the unit cell operations to give a block which is repeated by translations to give a crystal. The order in which the operations are applied is not significant since they form a group. The most general relationship between two identical asymmetric units consists of a rotation followed by a translation. This requires six variables for its description. The relationship is equivalent to a rotation by a particular angle along a helical path. If a series of units are related in the same way then they will all lie on the same helix. The 230 space groups include only those rational helices (2, 3, 4, 6, 2_1, 3_1, 3_2, 4_1, 4_2, 4_3, 6_1, 6_2, 6_3, 6_4, 6_5) which are compatible with three-dimensional lattices. All other helices are excluded, although these can occur in structures finite in one or more dimensions. The same form of generative rule, a helical displacement from one unit to the next, can be used to generate either the regular 230 groups or other structures such as the helical rod of TMV or the icosahedral 532 group of certain viruses. We should note that the rules for making a TMV rod include a statement that construction must be stopped when a specified length of RNA has been incorporated.

a

b

c

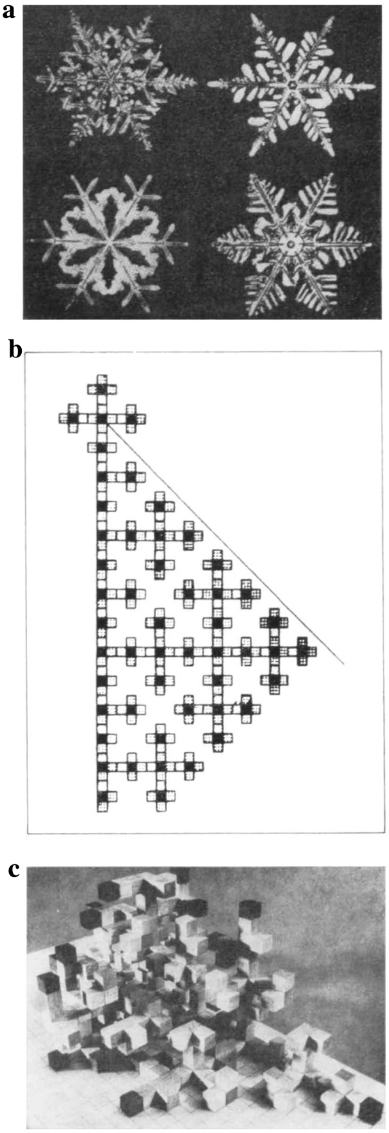


Figure 2 **a** Real snowflakes (from: W A Bentley and W J Humphreys 1962 *Snow Crystals* London: Dover). **b** The asymmetric unit from a two-dimensional pattern produced by S Ulam (Burks A W (ed) 1970 *Essays on Cellular Automata* University of Illinois p223) by simple rules of growth. **c** The asymmetric unit from a three-dimensional pattern produced by R G Schrandt and S Ulam (Burks 1970 p234)

The snowflake

The rules describing the morphology of growing crystals are more like those of the 'Life' game. For example, new molecules may bombard the surface of a growing crystal. If a molecule finds itself alone on the surface, it re-evaporates. If it lands in a corner or in a re-entrant angle then it sticks. If it arrives in the angle of a step, then it moves along until it reaches a re-entrant corner, and so on. Recast in this form there is an evident analogy, but the significance is that a set of simple rules (a little more elaborate than those of the crystallographic symmetry operations) can produce a complex structure. It would seem possible to devise a set which would generate a convincing snowflake. Figure 2 compares a real snowflake with two- and three-dimensional patterns generated by S Ulam, one of the main promoters of the cellular automaton, using a few simple transition rules. These define how one molecule adds on to those which arrived earlier. In deciding whether an arriving molecule sticks or re-evaporates, consideration must be given to the neighbourhood and the notional 'vapour pressure' which it represents in a real ice crystal. (The pressure is higher over convex surfaces.)

Diffraction patterns

We can see how systematic or even local departures from 'perfect' crystallinity can be made implicit in the cellular automata which describe them. We need a way of applying this technique to evidence for such irregularities which has hitherto been overlooked.

Crystals are important for the determination of the structure of molecules because they provide an amplifier which multiplies the scattering of x-rays from a single molecule by the number of molecules in the array, perhaps 10^{15}, and resistance to the damage of single molecules by the viewing radiation. Both emphasize the spots in the diffraction pattern and neglect the background. Molecules which are damaged transfer their scattering contribution to the background as do those which are not repeated with the regular lattice periodicity. It follows that even systematic departures from the regular lattice are likely to be missed or neglected in current techniques of structure analysis. It is, for example, probable that the icosahedral particles of gold, discussed below, have been observed earlier as minor features in diffraction patterns.

In recent years high resolution electron microscopy, most prominently by J M Cowley and his school, has shown the detailed nature of many of the defect structures which may appear in 'nonstoichiometric' compounds (such as niobium oxides). Such defects may in fact be an essential part of the structure but are liable to be neglected because they do not contribute much to the integral spots in the diffraction patterns. It seems likely that there may well be a continuum of structural types connecting the classical crystal structures, through twins, multiple or polysynthetic twins, 'heavily dislocated' structures, and various other disorders, to the definitely noncrystalline but nevertheless 'regular' structures of liquids and glasses.

The aim is to have a notation which carries right across this spectrum, and this may be done by a system of transition rules, some of which may be probabilistic. To extend the definition of crystallinity to mean 'the degree to which identical components in a structure are in similar environments' demands the construction of appropriate quantitative indices of 'similarity of environment'.

Crystalloids

This new approach to crystals, which consists in regarding them as the product of a series of programmed growth steps, could lead to a new classification of imperfect or incomplete crystals.

Consider the very small particles of gold which can be seen with an electron microscope in an evaporated film. Gold is face-centred cubic, but these particles do not have that structure. They are actually icosahedral shells (figure 1). This structure is in no way a distortion or a twin, although formally it could be derived from the FCC structure by either of these descriptions. It has been shown, both experimentally and theoretically to be the most stable configuration for 55 or 147 atoms of gold. Only after growing for several more layers according to this icosahedral rule, does the usual FCC structure become the more stable. Now icosahedral symmetry is not treated in the *International Tables* and crystals are really only defined for infinite repetition. Yet every crystal must be defective, even if the only defect is the surface. However, if a crystal is only ten unit cells cube, about half of the unit cells lie in the surface and thus have environments very different from those of the other half. The physical observation is that very small aggregates need not be crystalline, although they may nevertheless be perfectly structured. It is proposed that the word *crystalloid* should be applied to them. (The word formerly occurred in chemistry but is now in disuse.) Accordingly, we may have revised definitions of the words *crystalloid*, *crystal* and *crystallite:*

Crystalloid: a configuration of atoms (or other identical components) finite in one or more dimensions, in a true free energy minimum, where the units are not related to each other by three lattice operations. It may be possible to describe the arrangement as approximately that of a twinned or dislocated lattice. Under the conditions prevailing, the configuration is more stable than a crystallite with the same components. A crystalloid may be characteristic of a certain number of components and the question of what happens when more are added is not considered.

Crystal: a unit cell consisting of one or more atoms or other components is repeated a large number of times by three noncoplanar translations. Corresponding atoms in each unit cell have almost identical surroundings. The fraction of atoms near the surface is small and the effects of the surface can be neglected.

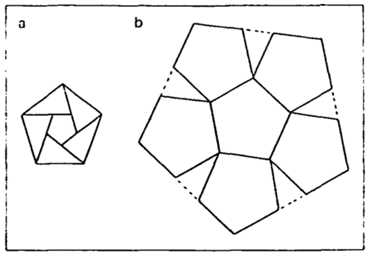
Crystallite: a small crystal where the only defect is the existence of an external surface. The lattice may be deemed to be distorted but it is not dislocated. Crystallites may be further associated into a mosaic block.

Crystallinity is the degree to which corresponding components in a structure are in identical surroundings.

Recursive rules

It is possible to use recursive rules to produce interesting structures which are 'regular' but noncrystalline. Two examples concerned with the packing of pentagons in a plane demonstrate some of the possibilities.

3



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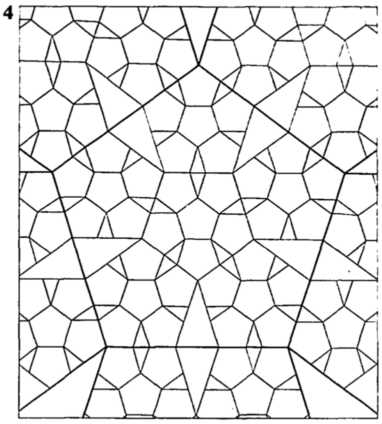

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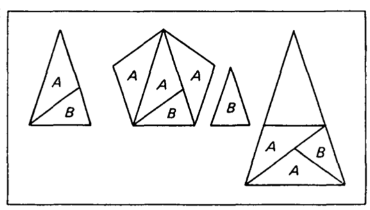

Figure 3 **a** The dissection of a pentagon into five triangles plus a smaller pentagon. **b** The assembly of six pentagons plus five triangles to give a larger pentagon

Figure 4 The hierarchic packing of pentagons

Figure 5 The *A* and *B* units which can be combined to give an infinite nonperiodic hierarchic packing of pentagons

They exhibit two different ways of relaxing the rules of symmetry to comply with the demand to tile a plane with regular pentagons. (There are, of course, a number of ways in which a plane can be tiled with identical, nonregular pentagons.) The first way allows the pentagons to be dissected as shown in figure 3a.

We are given a large supply of pentagonal tiles of order O and asked to cover a plane. This is done hierarchically by combining six zero-order pentagons to make a larger, first-order pentagon (figure 3b) and proceeding recursively. Unfortunately there are five triangular gaps. These are filled by cutting up a seventh pentagon which provides five triangles plus a pentagon of order -1. These constructions are then repeated on an ever-increasing scale (figure 4). The ratio of the linear dimensions of pentagons of successive order is φ^2, where $\varphi = (1+5^{1/2})/2$. This procedure embodies the identities: $\varphi^4+\varphi^{-4}=7$, $\varphi^8+\varphi^{-8}=47$, $\varphi^{12}+\varphi^{-12}=322$, etc. It is continued until the desired area, however large, is covered and the small residual pentagon almost disappears. This procedure is, of course, unlikely to be encountered in nature, since atoms, from which structures must be built, are indivisible.

A similar hierarchic packing of pentagons can, however, be realized without requiring a variety of sizes of components, by using pieces of only two types (*A* and *B* of figure 5). These can be combined together to five pentagons of order O and can also be packed to give the isosceles triangles of all orders which are required for filling in the gaps. We then obtain a tiling of *A* and *B* units which will cover the whole plane. It has many interesting properties, among which are: (i) the composition tends to $A_{\varphi}B$; (ii) there is a unique centre and rules can be given for finding a path to it; (iii) the pattern is an example of a hierarchic structure which looks almost the same at a number of scales if the boundaries of the pentagons are emphasized; (iv) although there is only one centre of true five-fold symmetry, there is an infinite number of centres of local pentagonal symmetry. Relatively short range centres are more densely distributed and longer range centres are more sparsely distributed; (v) circles can be inscribed in the pentagons and in certain interstices, to give a packing of finite density (the density does not tend to zero at an infinite distance from the start). R Penrose has independently produced a very similar pattern.

Hierarchy represents a natural way in which Wilson and Fisher's theory of phase transitions can appear. In this theory the region of interaction increases exponentially until, at the critical point, it comprises the whole specimen.

Attempts to find a three-dimensional analogy of these packings have not been successful, although it has earlier been shown that the packing of 13 icosahedra, each made of 13 spheres, is related to the icosahedral shell packing assumed by gold particles. A three-dimensional packing might be based on the identity $\varphi^3-\varphi^{-3}=4$, etc, but none has been found, the conditions being much more stringent than in two dimensions.

We must ask, as many have asked since Kepler, what the rules which lead to the formation of a snowflake are. We are just beginning to see how the rules for the growth of a tree are written in the genetic code. Is there really any resemblance between these two extremes of complexity? Does it make any sense to look back and ask where the programme for growing a snowflake may be stored? ■

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