DEDUCTION OF QUASILATTICE WITH FIVE-FOLD SYMMETRY AND PARTICLE FRACTAL STRUCTURE MODEL

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ABSTRACT

In the structure of quasicrystal, the coordination icosahedron has long ordering but no translation ordering. The author dealt with the building principle of quasicrystal and thought that two principles played a certain role in the quasicrystal structure, i.e. the icosahedron principle and the golden mean principle. We obtained the most simple structure model of quasicrystals, and could explain all details of the high-resolution electron microscopic image of the Al-Mn quasicrystal based on the two principles. The author's model has the characteristic of fractal structure, therefore, we call it the particle fractal structure madel. The author has made a systematic deduction of quasicrystal point group, forms, possible type of quasicrystal lattice.

Key words: quasicrystal, icosahedron principle, golden mean principle, fivefold symmetry, fractal structure model.

I. INTRODUCTION

D. Schectman[1] and his colleagues discovered a fivefold symmetric diffraction pattern in micro-size grains of Mn-Al alloy. This led to the discovery of the icosahedral phase.

Some structure explanations have been given to the new phase. D. Schectman et al. suggested that the way in which the coordinational icosahedra connected was by sharing.edges and arranging disorderly. This explanation is unsatisfactory. Levine and Steinhard[2] tried to explain the structure with the 3-dimensional penrose pattern and P. Kramer and R. Neri[3] thought that of the pattern was given by the

* This was the final paper about quasicrystal that the late Prof. Peng Zhi-zhong, a famous mineralogist and crystallographer, had planned to write. After his death on March 31, 1986, we organized the writing of the paper according to his wish. The paper is a summary of his work on quasicrystal. Participants in the discussions about the draft of this paper were Shen Jing-chuan, Shu Jin-fu, Lu Qi, Wang Su and Shi Ni-cheng. Finally, the paper was written by Shi Ni-cheng, Lu Qi and Wang Su and translated into English by Wang Su. All figures were plotted by Lu Ya-li.

projection of a superlattice in high-dimensional space. Nelso, however, sought for an explanation by the periodic accumulation of icosahedra in curved space. Hiraga et al.[4] have put forward a 3-dimensional model of 12-icosahedra. In the present paper the results of study of crystallography on quasicrystals, including the deduction of the point groups and forms with fivefold symmetry, icosahedron principle and golden mean principle which used to explain the structures of quasicrystals and particle fractal structure model, will be reported, which had been systematicly studied by the author since 1985[5–9]. These results were introduced to the First Chinese Symposium of quasicrystal.

II. Point Groups and Forms With Fivefold Symmetry

In order to set up the system of structural crystallography and geometric crystallography of quasicrystal the point groups and forms with fivefold symmetry have been deduced. The following 14 point groups have been deduced by using the rule of combining symmetry elements.

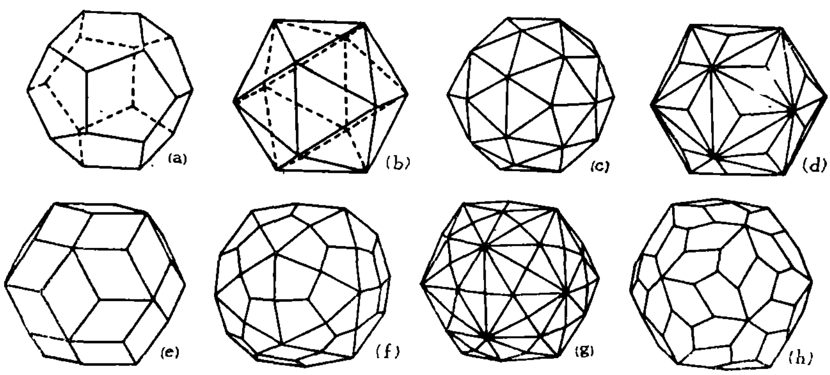


Fig. 1. Eight single forms with five rotational symmetry in isometric system.

Isometric system: $Y = 235$, $Y_h = m\bar{3}\bar{5}$.

Decagonal system: $C_{10} = 10$, $C_{10v} = 10\ m$, $C_{10h} = 10/m$, $C_{10i} = \overline{10}(5/m)$, $D_{5h} = \overline{10}\ m(=5/mmm)$. $D_{10} = 102$, $D_{10h} = 10/mmm$.

Pentagonal system: $C_5 = 5$, $C_{5v} = 5\ m$, $D_5 = 52$, $C_{5i} = \bar{5}$, $D_{5d} = \bar{5}\ m$.

The forms belonging to each point group above are shown in Table 1.

Twenty-four forms are introduced here. We should point out that not all of these are new ones. Some of them had been mentioned in some works on symmetry[10,11] and some of forms belonging to polyhedral geometry[12]. The author just introduced them into crystallography. Fig. 1 shows 8 new introduced forms in the isometric system.

III. Icosahedron Principle and Golden Mean Principle

Two principles are found to play an important rule in quasicrystal structure, i.e. icosahedron principle and golden mean principle.

Table 1

Forms in New Introduced Point Groups

(a) Isometric System

	$Y = 235$	$Y_h = m\bar{3}\bar{5}$
1	orthopentagonal dodecahedron	orthopentagonaldo decahedron
2	icosahedron	icosahedron
3	rhmbic tricenihedron	rhombic tricenihedron
4	trigonal trisicosahedron	trigonal trisicosahedron
5	tetragonal trisicosahedron	tetragonal trisicosahedron
6	pentadodecahedron	pentadodecahedron
7	pentagonal trisicosahedron	hexicosahedron

(b) Pentagonal System

	$C_5 = 5$	$C_{5v} = 5m$	52	$C_{5i} = \bar{5}$	$D_{5d} = \bar{5}m$
1	pedion	pedion	pinacoid	pinacoid	pinacoid
2	pentagonal prism	decagonal prism	pentagonal prism	decagonal prism	decagonal prism
3	pentagonal prism	pentagonal prism	decagonal prism	decagonal prism	decagonal prism
4	pentagonal prism	dipentagonal prism	dipentagonal prism	decagonal prism	dipentagonal prism
5	pentagonal pyramid	decagonal prism	pentagonal bipyramid	pentagonal reverse bipyramid	didecagonal bipyramid
6	pentagonal pyramid	pentagonal prism	pentagonal reverse bipyramid	pentagonal reverse bipyramid	pentagonal reverse bipyramid
7	pentagonal pyramid	dipentagonal prism	pentagonal trapezohedron	pentagonal reverse bipyramid	dipentagonal scalenohedron

(c) Decagonal System

	$C_{10} = 10$	$C_{10i} = \overline{10}$	$C_{10v} = 10m$	$D_{5h} = \overline{10}m$ (5/mmm)	$C_{10} = 10, 2$	$C_{10h} = 10/m$	$D_{10h} = 10/mmm$
1	pedion	pinacoid	pedion	pinacoid	pinacoid	pinacoid	pinacoid
2	decagonal prism	pentagonal prism	decagonal prism	pentagonal prism	decagonal prism	decagonal prism	decagonal prism
3	decagonal prism	pentagonal prism	decagonal prism	decagonal prism	decagonal prism	decagonal prism	decagonal prism
4	decagonal prism	pentagonal prism	didecagonal prism	dipentagonal bipyramid	didecagonal prism	decagonal prism	didecagonal prism
5	decagonal pyramid	pentagonal bipyramid	decagonal pyramid	pentagonal bipyramid	decagonal bipyramid	decagonal bipyramid	decagonal bipyramid
6	decagonal pyramid	pentagonal pyramid	decagonal bipyramid	decagonal bipyramid	decagonal bipyramid	decagonal bipyramid	decagonal bipyramid
7	decagonal pyramid	pentagonal pyramid	dipentagonal bipyramid	didecagonal bipyramid	decagonal trapezohedron	decagonal bipyramid	didecagonal bipyramid

1. *Icosahedron Principle*

The icosahedron principle means that the icosahedron coordination is the most

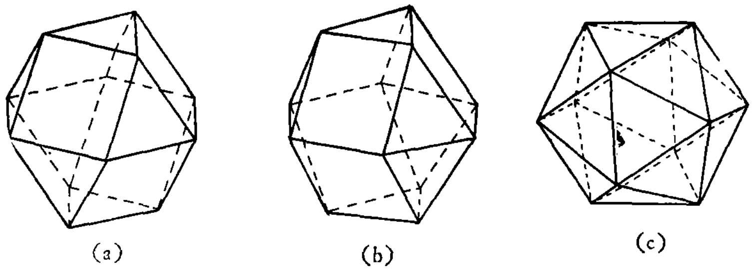


Fig. 2. Coordination polyhadron with $N = 12$.
(a) Cuboctahedron coordination, (b) hexagonal closest packing coordination, (c) icosahedron coordination.

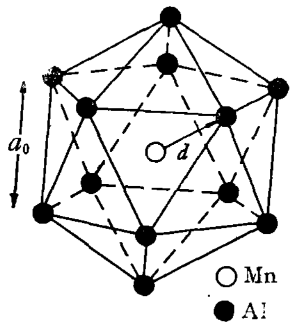


Fig. 3. Icosahedron coordination of $Al_{12}Mn$ in Al-Mn quasicrystal (according to Hirage et al.[4]).

favourable in energy in the system composed of few atoms of similar sizes. It is common knowlege that there are three basic patterns of coordination polyhedron whose coordination number is 12. Fig. 2(a) shows cuboctahedron coordination usually found in the structure of cubic closest packing, in which the corner-corner distances are equal but the corner-centre-corner angles are unequal. The angle distribution is 90°, 60°, 90°, 60°. Fig. 2(b) gives the hexagonal closest packing coordination. In this kind of coordination polyhedron the coordination atoms are unequivalent and can be separated into two classes: one is similar to the atoms in cuboctahedron, the other has the angle distribution of 90°, 90°, 60°, 60°. The two groups of atoms are unevenly distributed. They are not as stable as those of a cuboctahedron in terms of energy. Fig. 2(c) shows the icosahedron coordination, in which each atom in its corner is equivalent and all corner-centre-corner angles are 60°. This kind of coordination is the most stable in energy, so the icosahedron is the ideal coordination form for an isolated 12-coordination. Only for the geometric reason can icosahedra not be connected to form the space lattice structure. That is why we rarely see icosahedron coordination in crystal. Even if it exist, it will appear with a deformed pattern. The potassium's coordination in the structure of $K_3[Co(NO_2)_6]$ is just the case.

Hiraga et al. observed the icosahedron coordination in Al-Mn alloy quasicrystal from HREM image. The icosahedron coordination is the basic structural unit, therefore, the alloy is called the icosahedron phase. The radii of the metal atoms of Al and Mn are 1.432 Å and 1.366 Å respectively and their ratio (r_{Mn}/r_{Al}) is 1.048, which is the best value to form 12-coordination of icosahedron in which Mn is located at the centre, Al at the 12 vertexes. The distance of Mn—Al is 2.80 Å and that of Al—Al is 3.0 Å, which is close to Al—Al = 2.60 Å and Al—Mn = 2.61 Å in the equiponderant phases of Al-Mn alloy (Fig. 3).

A. Mackey and other crystallographers noted that when crystallites begin to grow around gas atoms the first few shells of atoms often have a tendency to take the coordination of an icosahedron. And J. Maddox pointed out that the precipitated metal formed by sublimated gases can form 5-fold symmetry.

As to the biological structure, Crick (1959) considered that the spherical virus tends to have icosahedron symmetry. As shown in Fig. 4 the centre of a cancer virus is nucleic acid surrounded by 42 protein couples (at vertexes and 30 edges in the icosahedron coordination). The example mentioned above that icosahedron coordination is the most favorable in energy. The icosahedron principle is not only applicable to the atom and molecule range but also applicable to bigger ranges, such as the gathering of spherical particles and the growing of cells.

It can be concluded that the icosahedron principle is not a guess but a objective law which dominates some processes of organization, especially in the beginning of the organization from a disordered state. As to crystal, in order to form lattice structure the icosahedron prinnciple has to be given up. The quasicrystal with fivefold symmetry can keep the principle up to micro-size grains. In biosphere the icosaheron principle is implemented fully and applied widely.

2. *Golden Mean Principle*

It is known that golden mean $(\sqrt{5}+1)/2 = 1.618$ is an irrational number and its reciprocal $(\sqrt{5}-1)/2 = 0.618$ is called golden section. The dimensions of

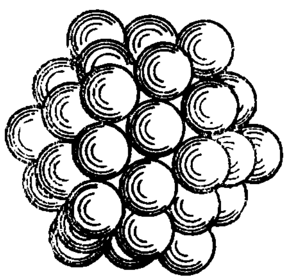


Fig. 4. Virus of cancer (from Belov, N. V. [13]).

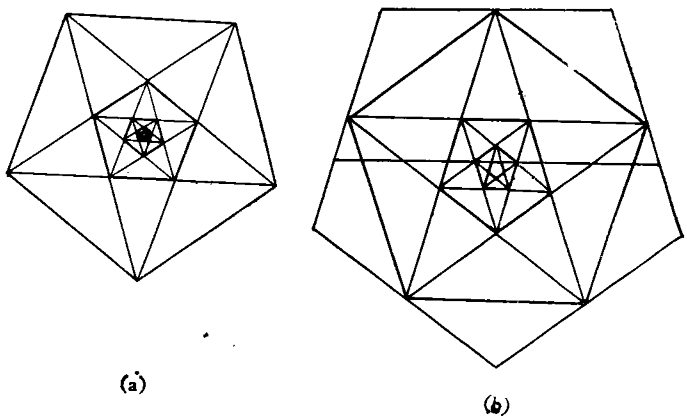


Fig. 5. Pentagon represents the golden mean principle.
(a) Alternation of pentagon and pentacle.
(b) Fibonacci sequence from pentagon.

buildings, the growing of plants and the breeding of animals are often in the ratio of golden mean. In other words, golden mean is a rational number in nature. We simply call all these mentioned above the golden mean principle.

In order to prove that the golden mean principle plays an important role in the structure of quasocrystal, first we should see pentagon in which all cross points of each line are located at the position of the golden section point. The cross lines of alternating vertexes of a pentagon construct a pentacle and a smaller pentagon is formed in the centre. Keep on doing so as shown in Fig. 5. The side of each pentagon and its neighboring of pentacle are in the ratio of golden mean. It is well worth notice that the side increases in the ratio of 1.618^2.

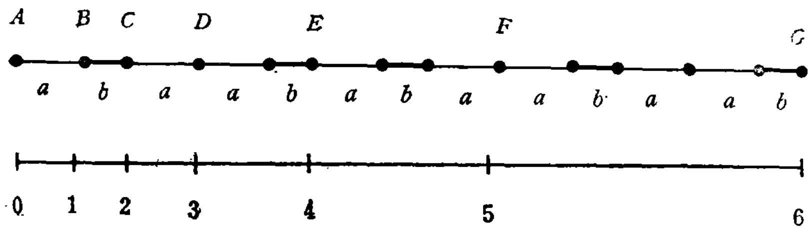


Fig. 6. Fibonacci seqrence.

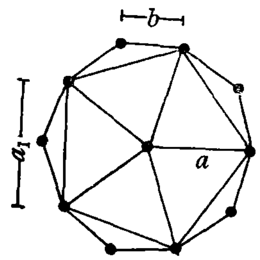


Fig. 7. Relation of a, b to a_1 in a_1 icosahedron on projection along fivefold axis.

Secondly we introduce Fibonacci sequence. F_n represents Fibonacci number which has the following: $F_0 = F_1 = 1$, $F_n = F_{n-1} + F_{n-2}$ ($n \geqslant 2$, positive round number), and when $n \to \infty$, $F_{n+1}/F_n = 1.618$. So the increase of F_n is in the ratio of golden mean. The Fibonacci sequence can be deduced from a pentagon (Fig.5). Fig. 6 displays the Fibonacci sequence in which there are two basic units, a and b, and $a/b = 1.618$.

As illustrated in Fig. 6 the sequence increased according to the golden mean principle has no translational period but has two definited lengths (a and b) and their radio is golden mean ($b/a = 0.618$). The emergency probability of each unit in the sequence is also golden mean. It is also a kind of periodicity and we call it quasiperiod.

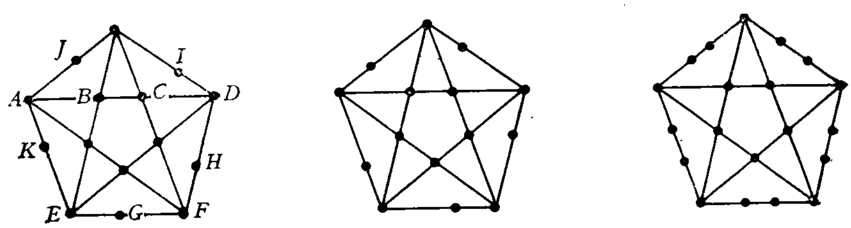
In the projection of the structure of Al-Mn quasicrystal (Fig. 7) along the fivefold

Table 2

Total lengths of T_n

The edge lenghts of the icosahedron (nm)	b_0 0.18	a_0 0.30	b_1 0.48	a_1 0.78	b_2 1.26	a_2 2.04	b_3 3.30	a_3 5.35
Row projection lenghts (T_n) as unit in a and b (nm)	T_{-2} 0.15	T_{-1} 0.26	$T_0(b)$ 0.41	$T_1(a)$ 0.66	T_2 1.07	T_3 1.74	T_4 2.81	T_5 4.55

axis the definite distances in the main rows are 0.66 nm(a_0) and 0.41 nm(b_0). The relation of a, b to a_1 (from Hiraga model) is shown in Fig. 7. They are deduced from $a_1 = 0.78$ nm($a_1/a_0 = 1.1755$, $a/b = 1.618$) measured from the high resolution image of Al-Mn quasicrystal. On the basis of the two units the step length T_n can be calculated (Table 2). T_n also increases in the ratio of golden mean.



(a) (b) (c)

Fig. 8. Deduction of quasicrystal lattice.
(a) A kind of deduction of quasicrystal lattice.
(b) Another way of deducing quasicrystal lattice.
(c) Superposition of two ways.

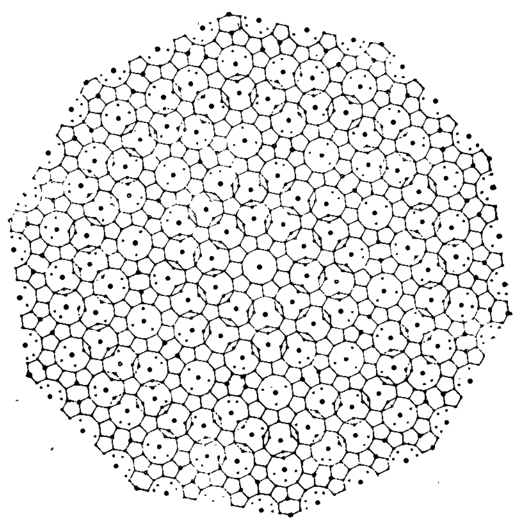


Fig. 9. Ideal quasicrystal lattice from fivefold axis.

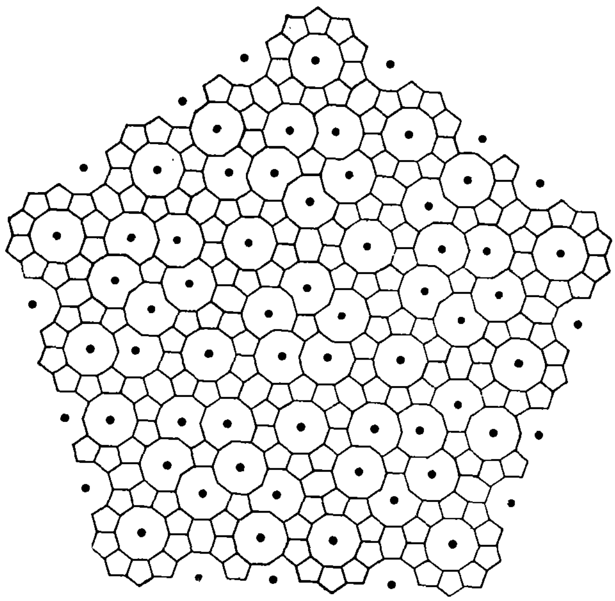


Fig. 10. The projection of pentagonal quasilattice.

The edge length of icosahedron increases in the ratio of golden mean. It is similar to penrose tiling in which the side ratios are also the golden mean[14].

IV. Deduction of Quasilattice With Fivefold Symmetry

The ideal quasilattics are deduced as follows.

We begin from the pentagon displayed in Fig. 8. Every point in it represents a coordination icosahedron. The *ABCD* row can be expanded according to Fibonacci sequence. We add a point *G* on the *EF* row which is parellel to *ABCD* and $EG = a$, $GF = b$. According to the symmetry *H*, *I*, *J*, *K* are deduced, and each row accords with Fibonacci sequence. In this way the whole lattice can be deduced. In the above way *n* for 2 μm-size grain of quasicrystal is 16 golden mean period. The whole grain is equal to an a_8 icosahedron.

We can distinguish the quasilattic type from original geometric patterns. 4 kinds of quasilattice are deduced:

Isometric system: (1) icosahedron quasicrystal (Fig. 9).

(2) orthopentagonal dodecahedron quasilattice (Fig. 9).

Pentagonal system: (3) pentagonal quasilattice (Fig. 10).

Decagonal system: (4) decagon quasilattice (Fig. 11).

For the 4 kind of quasilattice the one shown in Fig. 10 is deduced from the pentagon. The icosahedron quasilattices deduced from pentagonal axis and orthopentagonal dodecahedron quasilattice are equal in projection (Fig. 9).

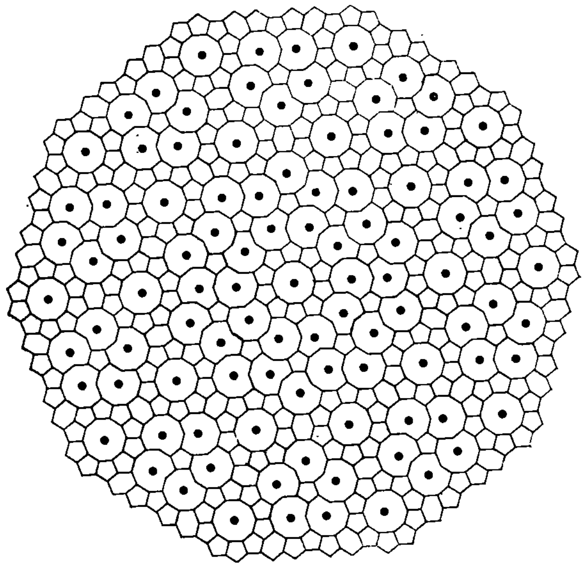


Fig. 11. The projection of decagonal quasilattice.

V. Particle Fractal Structure Model

Though Hiraga's model can manifest m35 symmetry and explain the high resolution image of Al-Mn quasicrystal, it has defects, e.g. in the model one of the Al—Al distances for each Al atom is just 1.854 Å which is too short to be realized. The problem would not be solved unless the model is completely changed. Moreover analysing Mossbauer spectrum L. Swartzendruber verified that in the structure (containing a little Fe) there are two kinds of Mn in the ratio of 1.60 ± 0.2 which is close to golden mean (1.618). Hiraga's model cannot explain the phenomena mentioned above. Another defect, which makes the model untenable is that an a_{-1} icosahedron is only composed of 12 a_0 icosahedron (Fig. 12), but in fact is not the case. When the model was made, we observed a big empty hole in the centre of the a_1 icosahedron. Actually the empty hole is another kind of the a_1 icosahedron with the edge length $b_0 = a_0/1.618$. There is an empty hole $b_{n-1} = a_{n-1}/1.618$ in an icosahedron. When the quasicrystal grain is big enough to 2 μm, it is certainly imposible that there is a hole as enough as 0.6—0.7μm in its centre. All these holes are illogical. So the Hiraga's model needs to be modified.

Based on the icosahedron principle, the golden mean principle, quasilattices deduced from the two principles and absorbing the rational parts of the quasicrystal models submitted by former scientists, we present a quasicrystal model, named as the particle fractal structure model.

In contrast with the crystal lattice, we understand the fractal structure as the structure whose period is varied. The periodic length can vary from 0 to ∞ on the axis and there is a fixed ratio between the neighbouring periodic lentghs. The ratio in the quasicrystal is 1.618 (or 0.618). This situation can appear in 1, 2, 3 dimen-

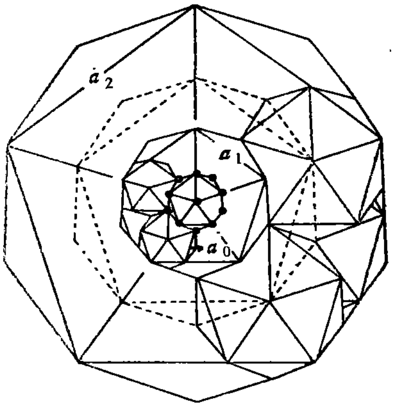


Fig. 12. Icosahedron (from Hiraga et al.[4]).

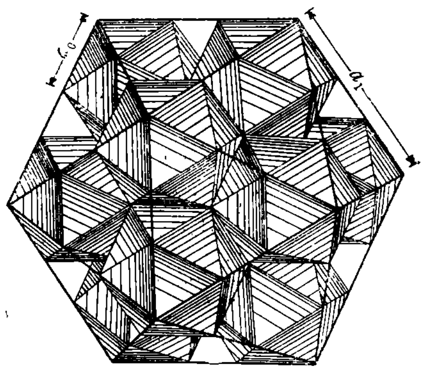


Fig. 13. a_1 icosahedron (Hiraga's model).

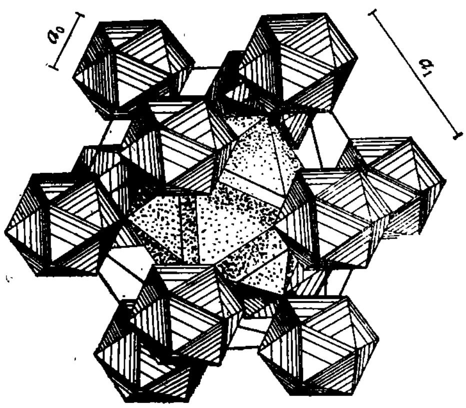


Fig. 14. a_1 icosahedron at the centre of the ideal Al-Mn quasicrystal, b_1 icosahedron at the center, a_0 icosahedron $Al_{12}Mn$ at the vertex (only Mn_I is shown).

sions. From the centre to the outside of the pattern, pentagon and pentacle appear alternatively and their size increases in the ratio of golden mean. This periodicity is the quasiperiodicity mentioned above or golden mean periodicity. This kind of preiodicity is also the selfsimilarity.

Fig. 5 shows that a quasicrystal grows from a nucleus of icosahedron. Applying the golden mean principle we deduced all grades of an icosahedron by modifying Hiraga's model. Another coordination polyhedron must be produced because icosahedron cannot fill up the whole space. The two kinds of polyhedron are in the ratio of golden mean in size. Because of being in fivefold symmetry the icosahedron coordinations must keep identical in orientation and the crystallite grows in the step of integer power of golden mean. The coordinational polyhedra are arranged in Fibonacci sequence. Fig. 14 shows the $Al_{12}Mn(I)$ coordination polyhedron in the structure of quasicrystal.

To sum up, the structure of quasicrystal with fivefold symmetry is a particle structure which is based on icosahedron coordination and is a fractal structure with golden mean similarity scale fractor, which has quasiperiodicity. This is the first discovery of the fractal structure in the scale of atoms in nature, which is of important significance.

According to the structure model of quasicrystal, Fig. 9 can be explained as follows.

The small black points represent the positions Mn(I) in the Al_{12} Mn(I) icosahedron and big black points the duplicated Mn. In the figure the projected distance of Mn—Mn is 0.41 nm, but the actual one is 0.78 nm which is larger than 0.48 nm, the Mn—Mn distance when the icosahedron is connected with common edges and is also larger than $2d(= 2 \times 0.28 = 0.56$ nm), the distance when the icosahedron is connected with common apexes. In the structure the Al_{12}Mn(I) icosahedron are separated each other, neither connected with common edges nor connected with common apexes (Fig. 14). This is the essential difference between our model and the others'. In the structure there is an Al-Mn(II) coordination polyhedron which is connected with Al_{12}Mn(I) icosahedron with common apexes. b_1 icosahedron and a_0 icosahedron are connected in the way of common apexes (Al), forming a_1 icosahedron $(a_1 = 0.78)$ shown in Fig. 14. In the projection a_1 icosahedron is a decagonal ring. Al_{12}Mn(I) are located in the apexes of a_1 icosahedron. This is the essential difference of a_1 icosahedron between our model and Hiraga's (Fig. 13). In our structure Al—Al distance is very rational. All of Al—Al distance are equal to 0.30 nm within Al_{12}Mn icosahedron and between them.

Another difference between our model and Hiraga's is that different grades of icosahedron in Hiraga's have different centres while in our model all grades of the icosahedron have a common centre from which the edges of the icosahedron are increased with T_n in the ratio of 1.618^n. The radius of the particle (T_n) is 9.095 nm, and the corresponding edge of the icosahedron is 10.69 nm, which is situated between a_2 and a_3 icosahedrons.

It can be observed from the projection that the pattern is invariable when it is expanded or shrunk 1.618^n time! This is just the character of fractal.

VI. Conclusion

Many theoretical and actual models of quasicrystal have been published by Hiraga[4], L. A. Burgill[14] et al. These models are all based on icosahedron connected by common edges. In this paper the particle fractal model is suggested on the basis of the icosahedron principle and the golden mean principle. In the model Al_{12}Mn icosahedron are not connected by common edges, but by common vextexes, which is different from all other models.

The things worthy of note are D. Levine and P. J. Steinhardt's work[2]. They Fouriertransformed a 2-dimensional Penrose pattern and got a tenfold symmetry pattern which is amazingly similar to the quasicrystal's diffraction pattern (Fig. 1, in D. Levine's paper) and to the centre part of Fig. 9 (in this paper)! The result deduced from Fibonacci sequence is a positive lattice and D. Levine and P. J. Steinhardt's pattern is a reciprocal lattice. This illustrated that our pattern is another form of Penrose tiling. The concept of quasicrystal should have a wide meaning and should not be limited to the materials with fivefold symmetry. The selfsimilarity scale factor should not be limited to golden mean. By combining quasicrystal with fractal we can understand the material world at an advanced level.

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