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A new method for generation of quasi-periodic structures with *n* fold axes: Application to five and seven folds*

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Abstract. A new geometrical method for generating aperiodic lattices for *n*-fold noncrystallographic axes is described. The method is based on the self-similarity principle. It makes use of the principles of gnomons to divide the basic triangle of a regular polygon of $2n$ sides to appropriate isosceles triangles and to generate a minimum set of rhombi required to fill that polygon. The method is applicable to any *n*-fold noncrystallographic axis.

It is first shown how these regular polygons can be obtained and how these can be used to generate aperiodic structures. In particular, the application of this method to the cases of five-fold and seven-fold axes is discussed. The present method indicates that the recursion rule used by others earlier is a restricted one and that several aperiodic lattices with five fold symmetry could be generated. It is also shown how a limited array of approximately square cells with large dimensions could be detected in a quasi lattice and these are compared with the unit cell dimensions of $MnAl_6$ suggested by Pauling.

In addition, the recursion rule for sub-dividing the three basic rhombi of seven-fold structure was obtained and the aperiodic lattice thus generated is also shown.

Keywords. Quasi-aperiodic 2D lattices; seven-fold axes; five-fold axes.

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1. Introduction

The recent discovery that rapidly cooled alloys of Al with Mn and other transition metals give electron diffraction patterns having the crystallographically forbidden 5-fold symmetry (Shechtman *et al* 1984) has created an exciting interest among the condensed matter physicists, material science researchers, metallurgists and others in the past few months. During this period, more than fifty papers have appeared dealing with both experimental and theoretical aspects of these alloys. In most of these papers, the experimental results have been interpreted as due to "quasi" crystalline nature of the alloys (Levine and Steinhardt 1984; Levine *et al* 1985; Elser 1985; Heiney 1985). Another interpretation is that the experimental results are due to twinning of crystals of these alloys (Field and Fraser 1984; Pauling 1985).

It is believed that the quasi-crystal structure is an aperiodic array of atoms but nevertheless, gives rise to Bragg-like peaks in the diffraction patterns. Calculations of

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Fourier transforms (FT) of structures with icosahedral symmetry and comparison with observed electron diffraction patterns have been recently reported (Duneau and Katz 1985). In particular, Duneau and Katz have shown from the projection data that the computed FT's agree reasonably well with the electron diffraction patterns taken along with 5, 3, and 2 fold axes of a triacontahedron. Other studies on quasi crystals include symmetry properties or an explanation of the origins of the quasi phase in terms of Landau theory (Bak 1985; Levine *et al* 1985; Mermin and Trorian 1985). However, no detailed structure determination of these alloys has been reported so far.

Moreover, it is also believed that the earlier proposals based on $Al_{86}Mn_{14}$ stoichiometry seem to be incorrect since ideal quasi composition is now taken to be near $Al_{80}Mn_{20}$.

Powder X-ray diffraction pattern of the Mn-Al alloy could not be indexed on any Bravais lattice earlier (Shechtman *et al* 1984). However, it was indicated that this powder pattern could be indexed on a Penrose lattice, with a cell edge corresponding to a rhombohedron $a_R = 4{\cdot}60$ A (4 atoms per cell according to the density). All the aperiodic structures based on the icosahedral symmetry have long range orientational order but no translational symmetry.

Recently, however, Pauling (1985) strongly argued that the icosahedral symmetry found in the Mn-Al alloy and in other transitional metal alloys is due to the directed multiple twinning of cubic crystals. Pauling suggested that the alloy can form metastable cubic crystals with a large cubic edge of approximately 26·7 A containing about 1120 atoms or more. The structure could have an ordered multiple growth such that twenty of them roughly tetrahedral in shape produce an aggregate with an approximate icosahedral symmetry. Pauling also showed that the X-ray powder lines could be indexed on the above cubic cell edge arrived at from chemical arguments. He had suggested a growth profile of a twenty-fold twin of the Mn-Al alloy with a pentagonal dodecahedron as a seed. Pauling, however, has not analyzed the characteristic 5-fold electron diffraction pattern of these alloys. Recently, it has also been argued that a macroscopic twinning of the alloy as suggested by Pauling has not been observed in any of the electron microscope studies that have been carried out so far (Shechtman and Blech 1985).

The purpose of the present paper is to develop a geometrical procedure for generating aperiodic lattices in two dimensions for *n*-fold noncrystallographic axes. The application of this procedure to 5-fold and 7-fold structures in two dimensions is also given. It is also shown how a limited array of approximately square cells with large dimensions could be detected in a quasi lattice in two dimensions.

2. The present method

Several projection methods such as strip method and generalized dual method (Socolar *et al* 1985) have been recently developed to generate aperiodic lattices. These are essentially projections of a hypercube of 6 dimensions on to 3,2 and other dimensions. The first attempt to generate an aperiodic structure was made by Penrose (1974) based on the now well-known Kite and Dart methods used for tiling a floor. Subsequently, Mackay (1981, 1982) generated aperiodic lattices in two dimensions using a set of recursion rules. This was further extended by him to aperiodic structures in three dimensions. In the following, a method for generating aperiodic structures using simple geometrical considerations will be described. The method was based on the assumption that the alloys of Al-Mn and other transitional metals comprise of interacting polyhedra such as zonohedra. Here, we consider a zonohedron that can be inscribed in a sphere. In two dimensions, a zonohedron projects as a regular polygon with appropriate edges that can be inscribed in a circle. The interactions in three dimensions of zonohedra in spheres reduce to in two dimensions, interactions of regular polygons inscribed in circles. The method discussed here shows how these regular polygons of $2n$ edges could be generated and how these could be used to generate aperiodic structures in two dimensions.

2.1 *Gnomons*

The method described here is based on the self-similarity principle. It is well known that a gnomon (Euclid 1908) is defined as any figure which when added to any other figure whatsoever leaves the resultant figure similar to the original. Thus, in any triangle one part is a gnomon to the other. In figure 1a, BCD is a gnomon to ABD. In figure 1b, which is an isosceles triangle with a vertex angle A = 36° (the angle relevant to the present investigation), CD divides equally the base angle at C. Again BCD (Q) and ADC (P) are gnomons. In this case, the lengths BC, CD and AD are equal. Gnomons which are also relevant to the present investigation are rhombi and two such rhombi ABCD and BEDF are shown in figure 2. The above principle of dividing a triangle into gnomons has been successfully employed to generate rhombi which are again gnomons and to fill the two-dimensional space aperiodically with a n-fold noncrystallographic axis such as 5, 7 etc.

A regular polygon of $2n$ edges could be divided into $2n$ isosceles triangles. The isosceles triangle of a regular polygon is referred to as a basic triangle. In figure 3, the basic triangle (drawn in thick lines) of a regular decagon has been divided into two parts so as to generate two rhombi, one acute and the other obtuse. The decagon is thus made up of 10 rhombi, 5 acute(RI) and 5 obtuse(RII) types having a 5-fold axis at the centre. Referring to figure 1b which is the basic triangle for $n = 5$, it is clear that the acute(RI) and the obtuse(RII) rhombi of figure 3 have the angles at the vertices as follows RI (36°, 144°) and RII (72°, 108°). This will also be referred to either as RI(5) and RII(5) or as RI(α, 4α) and RII(2α, 3α) where $\alpha = 36°$. Similarly, the basic triangle for $n = 7$ corresponding to a regular 14-gon can be divided in the same manner so as to generate rhombi and fill up the space in two dimensions. Figure 4 shows the basic triangle (drawn in thick lines) for $n = 7$ and this can be divided into three parts, R, S and T as shown and the corresponding polygon with 14 edges has been made up of three sets of rhombi, RI(7), RII(7) and RIII(7) with angles at vertices: RI(α, 6α), RII (2α, 5α), RIII (3α,

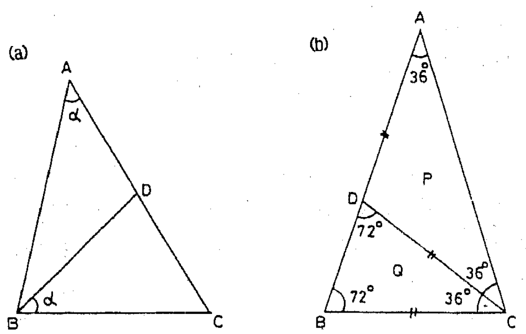


Figure 1a. In triangles ABC, ABD and BCD are gnomons. **1b**: Isosceles triangle with vertex angle A = 36°. P and Q are gnomons. Note that BCD and ADC are also isosceles triangles.

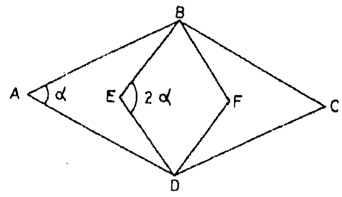


Figure 2. Two rhombi as gnomons. Here the lengths AE and FC (not joined) are equal to the sides of the rhombus BEDF.

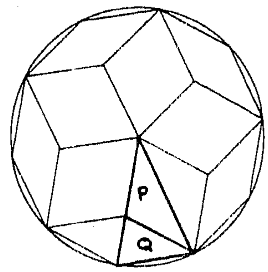


Figure 3. The basic isosceles triangle of a decagon (thick lines) is divided into two isosceles triangles P and Q. Note that there will be 10 such isosceles triangles leading to 5 acute and 5 obtuse types of rhombi.

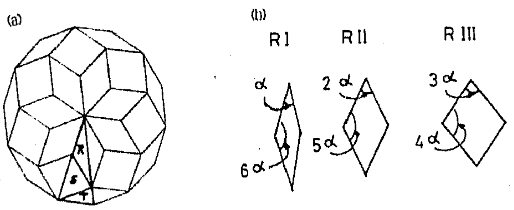


Figure 4a. The basic isosceles triangle for $n = 7$ fold axis (thick lines) is divided into three isosceles triangles R, S and T. These triangles generate the three rhombi of 4b. **4b.** A set of three rhombi RI(7), RII(7) and RIII(7) along with the corresponding angles at the vertices that are required to fill two dimensional space aperiodically with $n = 7$ fold axis.

4α) where α is 180/7 = 25·714°. Similarly for $n = 6$ (not shown here), the basic triangle of a dodecagon can be divided to generate three sets of rhombi with vertex angles as RI(α, 5α), RII(2α, 4α), and RIII(3α, 3α), where α is 180/6 = 30°. The table lists the appropriate sets of rhombi required to fill polygons with $n = 4$, $n = 5$, $n = 6$, $n = 7$, $n = 8$ and $n = 9$ axes of symmetry.

Thus for a regular decagon two rhombi, for a regular dodecagon and for 14-gons three rhombi and for regular 16 and 18 gons four rhombi with appropriate vertex angles as shown in the table are required. It is obvious that the basic triangle for any value of n (axis of symmetry) can be divided appropriately to generate a finite set of rhombi to fill the corresponding polygon.

2.2 *Generation of aperiodic lattices for a $n = 5$ fold noncrystallographic axis*

For a $n = 5$ axis of symmetry, there are four arrangements of the two rhombi, RI(5) and RII(5) of the table that are possible and fill the decagons. The four arrangements are shown in figure 5. These are called the 4 patterns $A(5)$, $B(5)$, $C(5)$ and $D(5)$ of the decagon. Minor variations in the arrangements of the three rhombi, which form a hexagon within the decagon, such as interchange of two RI(acute) rhombi with one RII(obtuse) rhombi are possible (see caption to figure 5). The possible arrangements of these four patterns in the two dimensions lead to a number of aperiodic lattices and these are described below.

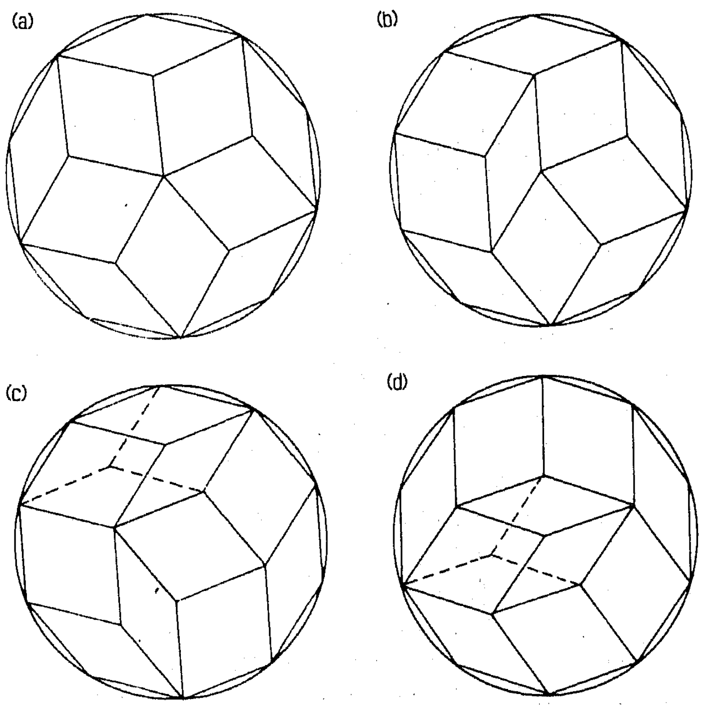
Two decagons inscribed in circles can share one, two or three edges (see figure 6). And for $n = 5$, the total number of shared edges at any intersection of three or more circles should necessarily be five. Figure 7 shows the sharing of edges by a set of intersecting circles. Here the circle at the centre intersects with five, six or seven other circles surrounding it. It is readily seen that at any intersection of any three circles, the shared

Table 1. Minimum sets of rhombi required to fill two-dimensional space for a few noncrystallographic axes of symmetry.

n*	RI $(\alpha, \pi\text{-}\alpha)$	RII $(2\alpha, \pi\text{-}2\alpha)$	RIII $(3\alpha, \pi\text{-}3\alpha)$	RIV $(4\alpha, \pi\text{-}4\alpha)$	Value of α
4	$(\alpha, 3\alpha)$	$(2\alpha, 2\alpha)$	—	—	$\frac{\pi}{4} = 45°$
5	$(\alpha, 4\alpha)$	$(2\alpha, 3\alpha)$	—	—	$\frac{\pi}{5} = 36°$
6	$(\alpha, 5\alpha)$	$(2\alpha, 4\alpha)$	$(3\alpha, 3\alpha)$	—	$\frac{\pi}{6} = 30°$
7	$(\alpha, 6\alpha)$	$(2\alpha, 5\alpha)$	$(3\alpha, 4\alpha)$	—	$\frac{\pi}{7} = 25{\cdot}71°$
8	$(\alpha, 7\alpha)$	$(2\alpha, 6\alpha)$	$(3\alpha, 5\alpha)$	$(4\alpha, 4\alpha)$	$\frac{\pi}{8} = 22{\cdot}5°$
9	$(\alpha, 8\alpha)$	$(2\alpha, 7\alpha)$	$(3\alpha, 6\alpha)$	$(4\alpha, 5\alpha)$	$\frac{\pi}{9} = 20°$

The minimum sets of rhombi required to fill two dimensional space for other values of n, can be readily obtained. Aperiodic lattices are not obtained for $n = 2$ and 3 axes of symmetry. Both periodic and aperiodic (with the sets of rhombi given above) lattices are obtained for $n = 4$ and 6.

* As the polygon is taken to have $2n$ edges, there exists in each polygon a $2n$ fold axis of symmetry. Note that for both n and $2n$ values, therefore, the same set of rhombi can be used for generating aperiodic lattices. Hence the minimum set required in each case corresponds to the maximum $2n$ fold axis.



Figures 5a, b, c & d. The four arrangements of the two rhombi, RI(5) and RII(5) to form 4 decagons. The arrangements are referred to as patterns $A(5)$, $B(5)$, $C(5)$ and $D(5)$ in the text. The alternative arrangement of the two acute and one obtuse rhombi within the decagon is also shown by dashed lines.

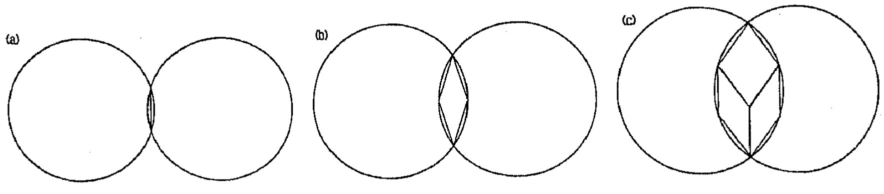
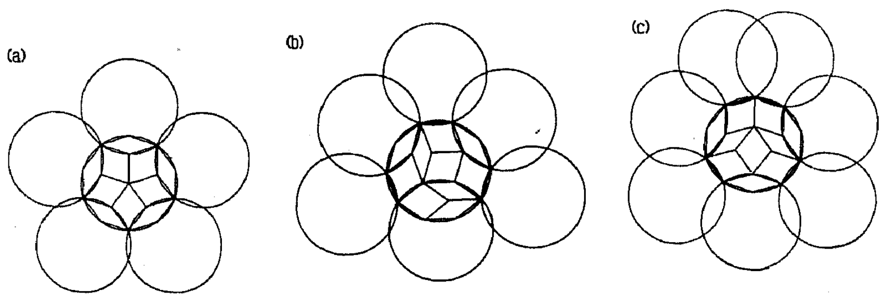


Figure 6. Two decagons inscribed in circles can share (**a**) one, (**b**) two and (**c**) three edges. Only the edges of the decagons shared are shown.



Figures 7a, b & c. Sharing of edges by a set of intersecting circles; (**a**) circle at the centre with the decagon corresponding to pattern $A(5)$ intersects with 5 other surrounding circles. The shared edges are 2,2 and 1. Note that the outer circles share only one edge with each other. (**b**) a circle at the centre with the decagon corresponding to pattern $C(5)$ intersects with 6 other surrounding circles. The shared edges are 1,2 and 3. Note that the outer circles share two edges or one edge with each other. (**c**) a circle at the centre with the decagon corresponding to pattern $C(5)$ intersects with 7 other surrounding circles. The shared edges are 1,2 and 2. Note that the outer circles share two or three edges with each other.

edges of the three circles are either 2,2,1 or 3,1,1 leading to a total of 5 shared edges. For intersection of more than three circles other possible combination of shared edges will be, 4, −1,1, and 3,3, −1 not shown in the above figure (see caption to figure 9). Again the total number of shared edges being equal to 5 only.

Thus, starting from any one of the 4 patterns of the decagons inscribed in a circle a series of intersecting circles (decagons) can be obtained and thus fill the two dimensional space, following the general rule that at any intersection of 3 circles or more, the total number of shared edges being equal to 5. Figure 8 shows a lattice generated using this principle. In generating this lattice patterns $A(5)$ and $C(5)$ have been used. This lattice is identical to the Penrose lattice of Mackay. This clearly demonstrates that this procedure of generating aperiodic lattice is valid. It also indicates that the recursion rule used by Mackay and others is a special case of a more general rule and that several other aperiodic lattices can be generated. That this is so can be appreciated from figure 9 which shows only the intersecting circles of the previous figure without the decagons inside the circles. It is obvious that any combination of the four patterns can be used to fill the circles and the intersections so as to generate a number of lattices with different arrangements for the two rhombi RI(5) and RII(5). So also other types of arrangements for the intersecting circles other than what has been shown in figure 9 can be generated and these filled with decagons using any combination of four patterns and obtain different lattices. A pattern thus generated for an arbitrary arrangement of the two rhombi is shown in figure 10.

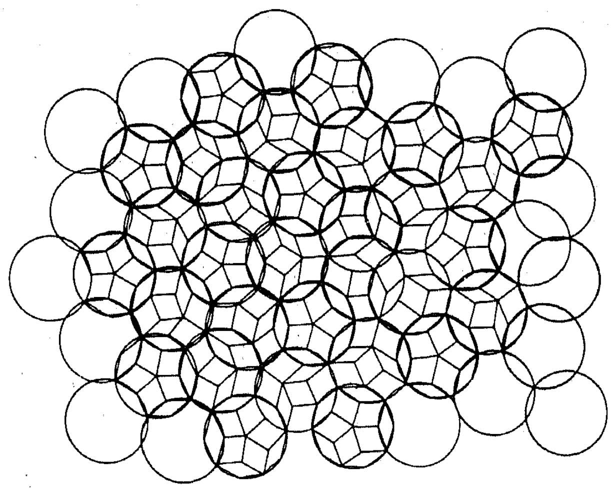


Figure 8. Aperiodic lattice generated using the procedure described in this paper. This lattice is identical to one described by Mackay and others.

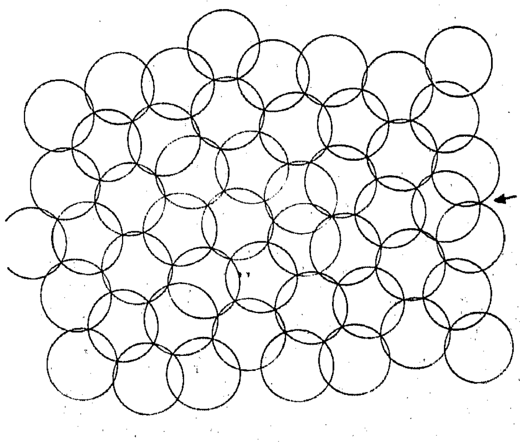


Figure 9. Same as figure 8 but with only the intersecting circles and without the decagons inside. The arrow is the intersection of four circles wherein the shared edge of two circles lies within the third circles. The fourth intersecting circle is not drawn. The third circle shares 3 edges with the other two circles. Thus, the total number of shared edges to be reckoned as $3+3-1=5$. This is referred to in the text as $3,3,-1$.

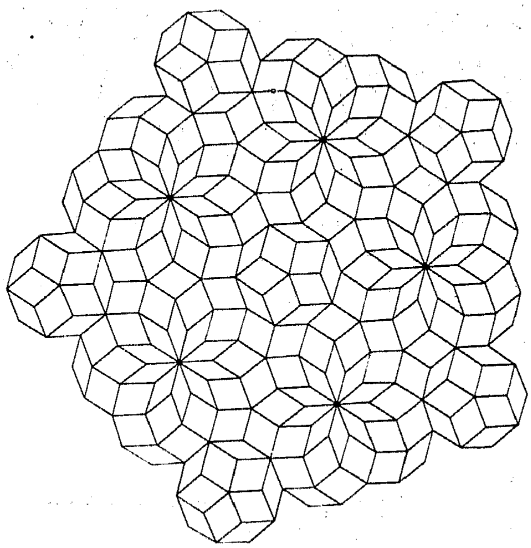


Figure 10. An aperiodic pattern generated for an arbitrary arrangement of the two rhombi RI(5) and RII(5). The origin is a true 5-fold axis.

An aperiodic lattice should necessarily have only one true *n*-fold axis. This can be at the centre of the diagram as shown in figure 11 or at infinity as has been taken by others, in which case, the true *n* fold axis will not be there in the two-dimensional diagram and only local *n*-fold site symmetry will be present. In figure 11 one can readily recognize a series of hexagons with vertices having local 5-fold symmetry. Far away from the centre (the origin of the true 5 fold) these hexagons although aperiodic appear to be arranged in a regular fashion. This aspect is emphasised in figure 12 wherein only the hexagons are drawn without the decagons inside for clarity. Away from the centre these hexagons are periodically arranged in a restricted fashion. Most interestingly it turned out that one can obtain approximate square cells as shown. Further away from the two square cells shown, a series of approximate square cells are therefore generated. Thus, approximately a square lattice (with one cell edge larger than the other edge by 2·5 %) with a large cell dimension such as 30 A can be obtained, away from the true *n* fold axis. If still larger (approximately) square cells are used, the above percentage of difference in

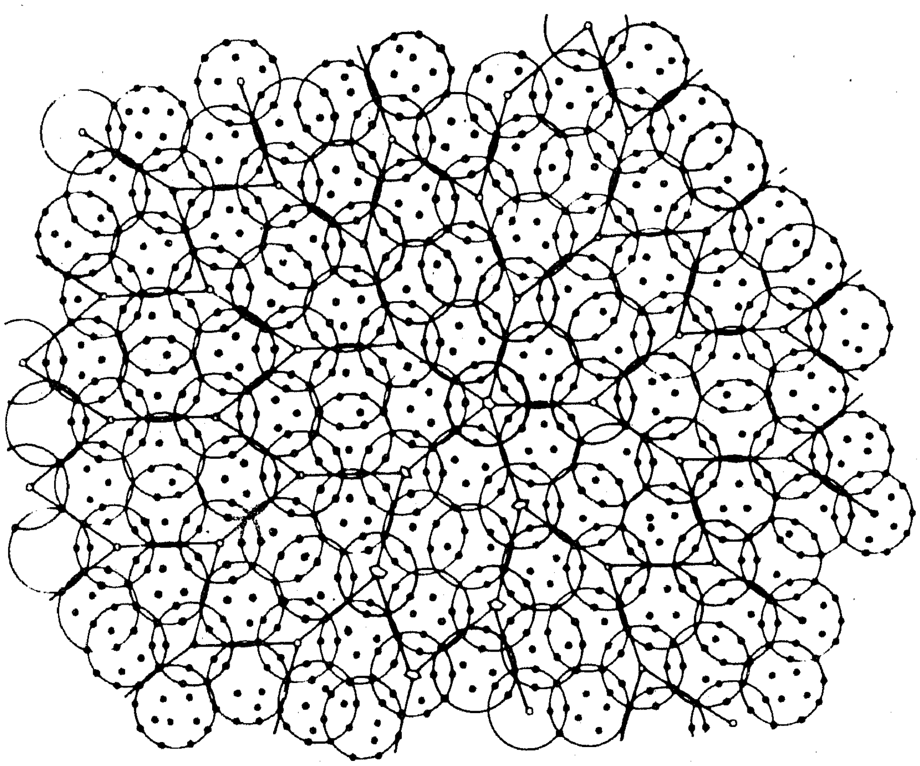


Figure 11. Aperiodic lattice with a true 5-fold axis at the centre. The vertices of the rhombi of the intersecting decagons are shown as dots. Note that hexagons are formed by joining the centres of local 5 fold site symmetry. Note that the hexagons appear parallel at the edges of the figure.

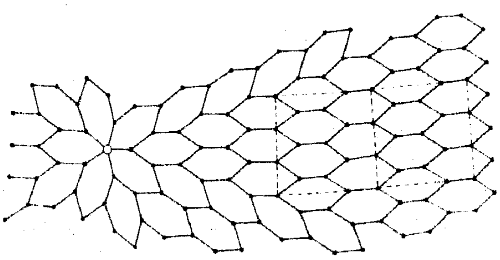


Figure 12. This is an extended pattern of figure 11 but with only the hexagon drawn without the decagons inside. At the right hand side the approximate square cells obtained are also shown. One side of this cell is larger than the other by 2·5 %.

the cell edges can still be reduced. Taking the ionic radii of Al and Mn ions as given by Pauling the unit cell shown in figure 12 coincides with the unit cell given by Pauling for $MnAl_6$ alloy within an error of 2·5 %. It may be noticed that the contents of the hexagons are the decagons in two dimensions and these decagons could then be the two dimensional projection of triacontahedra rather than pentagonal dodecahedra, as suggested by Pauling.

Thus, it turns out that aperiodic structures with one true 5-fold axis could be generated and fill the space in two dimensions. In one such aperiodic structure unit cells which could be approximated to square cells with large dimension (of the order of 30 A) and with a difference of 2·5 % or less in the cell edges could be generated. When extended to 3 dimensions, this could be approximated to cubic cells. Further work has to be done to clearly demonstrate whether the basic structure is a triacontahedron or a pentagonal dodecahedron.

2.3 *Generation of aperiodic lattices for a $n = 7$ fold non-crystallographic axis*

The geometrical approach was then applied to $n = 7$ fold noncrystallographic axis for generation of aperiodic structures in two dimensions. The basic triangle for generating the three rhombi is shown already in figure 4. The figure is a regular polygon of 14 sides which is filled by 21 rhombi (7 × 3) and with the use of RI, RII, RIII types of rhombi for $n = 7$ given in the table. Following the procedure adopted for the $n = 5$ fold, several regular polygons with 14-sides could be generated using the set of three rhombi. Different arrangements of the three rhombi RI(7), RII(7) and RIII(7) are possible to fill the 14-gons. Calling these arrangements as patterns of the 14-gons, as before, a number of aperiodic lattices could be generated. These regular polygons of 14-sides inscribed in circles could be shown to share 1,2,3 etc edges and starting from any one of the patterns of the 14-gons a series of intersecting circles could be obtained and thus fill

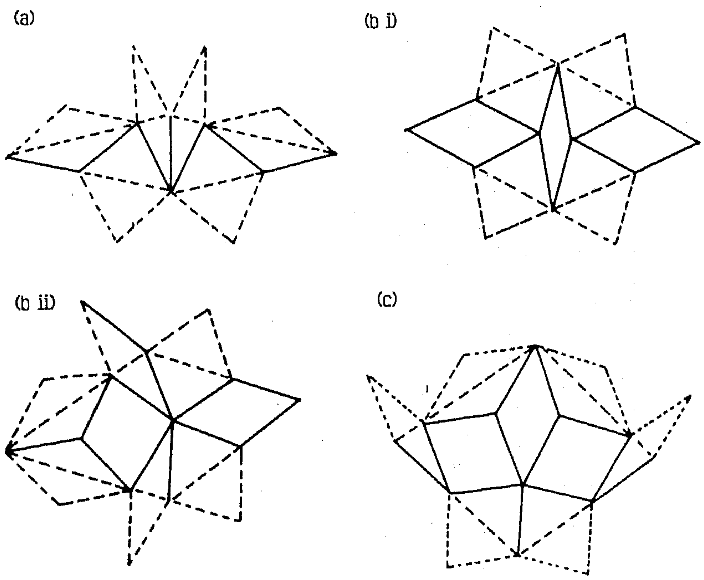


Figure 13. Recursion rule for sub-dividing the three rhombi of table 1 corresponding to $n = 7$ to generate structures in two dimensions. (**a**) Rhombus RI(7) goes to one RI(7), one RII(7) and one RIII(7) in the next generation. (**b**) Two variations of sub-dividing rhombus II. In both, one RII goes to one RI, two RII and two RIII. (**c**) In the above one RIII(7) goes to one RI(7), two RII(7) and three RIII(7). Variation of subdividing rhombi are there in (a) and (c) also but not shown here.

the two-dimensional space, following the general rule that at any intersection of three circles or more, the total number of shared edges should be equal to 7.

It was also found that a recursion rule for sub-dividing the three basic rhombi RI(7), RII(7) and RIII(7) could be obtained and hence aperiodic lattices generated. The recursion rule for sub-dividing the three rhombi is shown in figure 13. Here, one RI(7) goes to one RI(7), one RII(7) and one RIII(7) in the next generation as shown in figure 13a. Similarly, one RII(7) goes to one RI(7), two RII(7) and two RIII(7) (figure 13b). Again, one RIII(7) goes to one RI(7), two RII(7) and three RIII(7) (figure 13c). Nonpoles in one generation become the poles in the next. A 7-fold aperiodic lattice generated following the above recursion rule is shown in figure 14. The origin has a true 7-fold axis and the diagram illustrates the heptagonal arrangement. Several polygons of 14 sides can be seen. The symmetry properties, other features of this diagram and the extension of this to the three dimensions are being investigated and will be reported

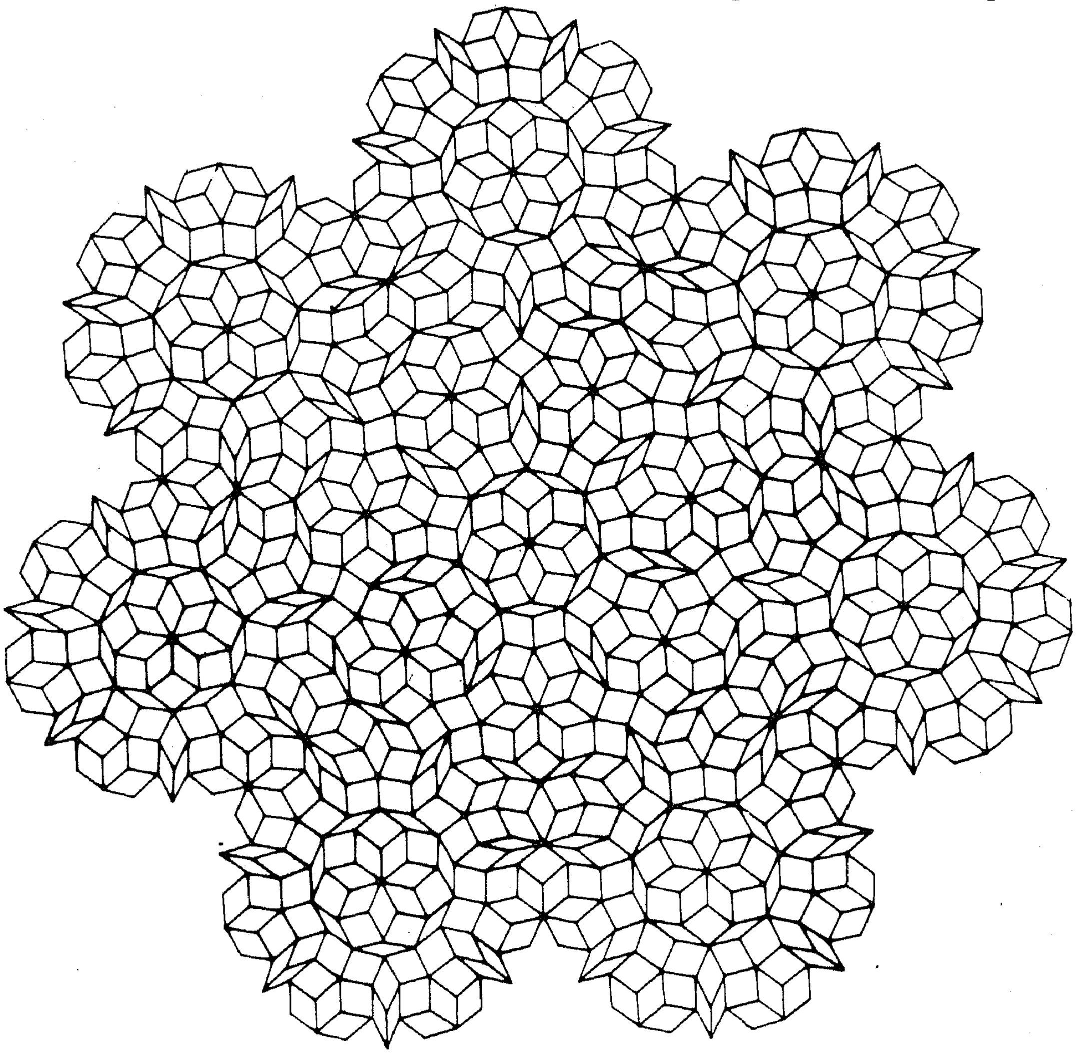


Figure 14. A 7-fold aperiodic lattice generated following the recursion rule shown in Fig. 13. The origin is a true 7 fold axis. The local 7 fold site symmetry can be readily seen. The diagram illustrates an heptagonal arrangement. Note several polygons of 14 sides. The recursion rule (Fig. 13b(2)) is used at the origin. To illustrate, the other rule (Fig. 13b(1)) has been applied at three vertices of the heptagon. This application results in the rotation of the 14-gons at these vertices by $\pi/7°$ and hence appear nonequivalent about the origin. For exact equivalence, these 14-gons are to be rotated by the above angle about these vertices.

elsewhere. So also application of the method described in this paper to other noncrystallographic axes other than 5 and 7 will be described elsewhere.

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