



**HAL**  
open science

# Microcrystals and quasicrystals : how to construct microcrystals with mean perfect quasicrystalline symmetry

G. Coddens

► **To cite this version:**

G. Coddens. Microcrystals and quasicrystals : how to construct microcrystals with mean perfect quasicrystalline symmetry. Journal de Physique I, 1991, 1 (4), pp.523-535. <10.1051/jp1:1991108>. <jpa-00246348>

**HAL Id: jpa-00246348**

**<https://hal.science/jpa-00246348v1>**

Submitted on 4 Feb 2008

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



HAL Authorization

Classification

*Physics Abstracts*

61.50E — 61.55H — 64.70E

## Microcrystals and quasicrystals : how to construct microcrystals with mean perfect quasicrystalline symmetry

G. Coddens

Laboratoire Léon Brillouin, Commun CEA-CNRS, F-91191-Gif-sur-Yvette Cedex, France  
Neutron Scattering Project IIKW, University of Antwerp, B-2610-Wilrijk, Belgium

*(Received 15 May 1990, revised 12 December 1990, accepted 13 December 1990)*

**Résumé.** — On démontre comment la construction d’"intermaillage" clarifie la relation entre les quasicristaux et les microcristaux dans les transitions de phase comme celles observées récemment dans les expériences. Les diagrammes de diffraction et la géométrie des domaines sont considérés simultanément et l’éclatement des pics de diffraction dans les transitions de phase peut être calculé. Plusieurs scénarios sont possibles.

**Abstract.** — It is shown how the construction of "intertwining" elucidates the relationship between quasicrystals and microcrystals in phase transitions as observed in recent experiments. Both the diffraction patterns and the domain geometry are studied simultaneously. The splitting of the diffraction peaks in the phase transition can be calculated. Several scenarios are possible.

### 1. Introduction.

Of considerable interest for the study of quasicrystals (QC) [1] is their relation with periodic crystalline structures. Related periodic structures may be a good starting point for QC structure determinations. Various such relationships are discussed in the literature. As a first case in point we may cite crystalline approximants. In the cut-and-projection method [2] crystalline approximants are obtained by a slight tilt of the strip in superspace so as to give it a rational orientation with respect to the superspace lattice [3]. Even Pauling’s [4] controversial questioning of the existence of QC could be classified within this category of relationships, be it that the motivation was entirely different and not appropriate. As a second class of relationships, cases have been reported where QC phases coexist with twins exhibiting orientational correlations with them [5]. The present paper addresses an exciting third class of relationships : recent experimental findings [6] have shown that an icosahedral diffraction pattern can be produced by a polycrystal (microcrystal) built up of periodic rhombohedral or cubic domains. A telling case is the system AlFeCu which for certain stoichiometries has a microcrystalline structure at room temperature and undergoes a phase transition to a perfect QC around 670°C. In order to produce the diffraction patterns observed the microcrystals must have special orientational and phase relations (coher-

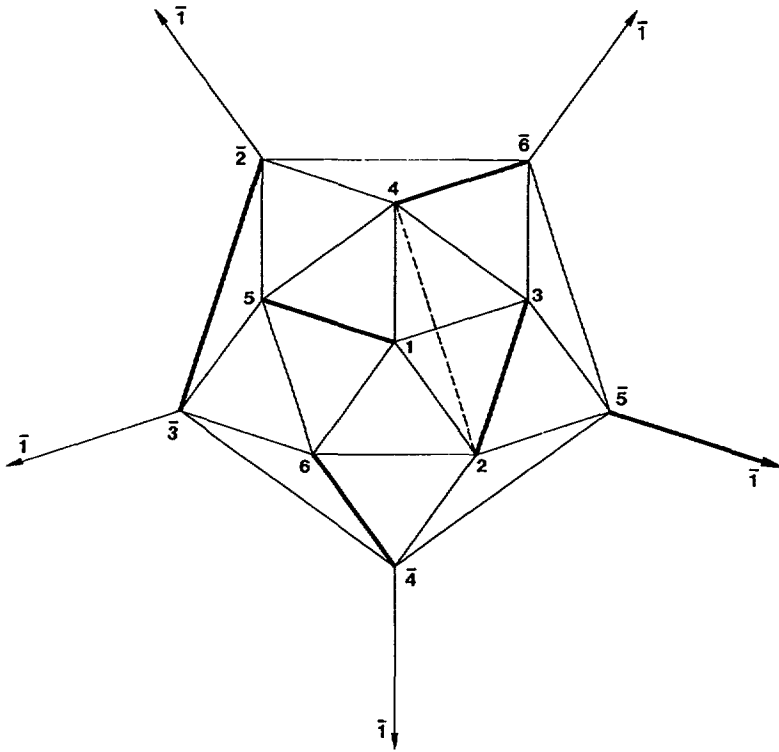


Fig. 1. — Schlegel diagram of the icosahedron allowing to display all its faces in one figure. The triangles 123 etc... correspond to the fat Amman tiles. The thin Amman tiles correspond to triangles of the type 124. The directions of one of the five cubes inscribed are shown.

ent domains). Substantial experimental evidence for the coherence of the domains was reported by Bancel [6]: high resolution electron microscopy shows clearly the domain structure and the X-ray diffraction patterns with overall QC symmetry require a coherence between them. The idea of coherent domains however was first suggested by Denoyer *et al.* [6] but without clue as to how this could be *globally* realised in practice. In fact it is easy to produce such relationships *locally* between domains based on *one* unit cell (if one assumes that there is only one stable phase in the microcrystal) but it remains a problem how to tile *entire* space with mean perfect icosahedral order using only *one* rhombohedral or cubic unit cell. This order can only be understood in an average and not in a rigorously perfect sense since the five-fold symmetry is a forbidden crystallographic symmetry, i.e. we have to expect some errors. More precisely the microdomain arrangements in the figures of Denoyer *et al.* are visually very appealing since all the domain boundaries are perfect twin boundaries without any frustration. But to pursue extending this esthetic ideal to the whole microcrystal as these figures may suggest, reveals itself quickly as a Utopian dream since sooner or later one has to run into a frustration (as illustrated in our Fig. 2) due to the crystallographic forbidden symmetry. In this paper we try to describe a structure that is as realistic as possible. (a) We construct a coherent disposition of domains with the average QC symmetry observed. The coherence extends throughout the whole microcrystal despite possible *apparent* frustration inside certain domain boundaries. (The essential ingredient for a recipe to obtain infinitely long range coherence does not consist in ending up with nice pictures with *unit cell* tilings showing perfect

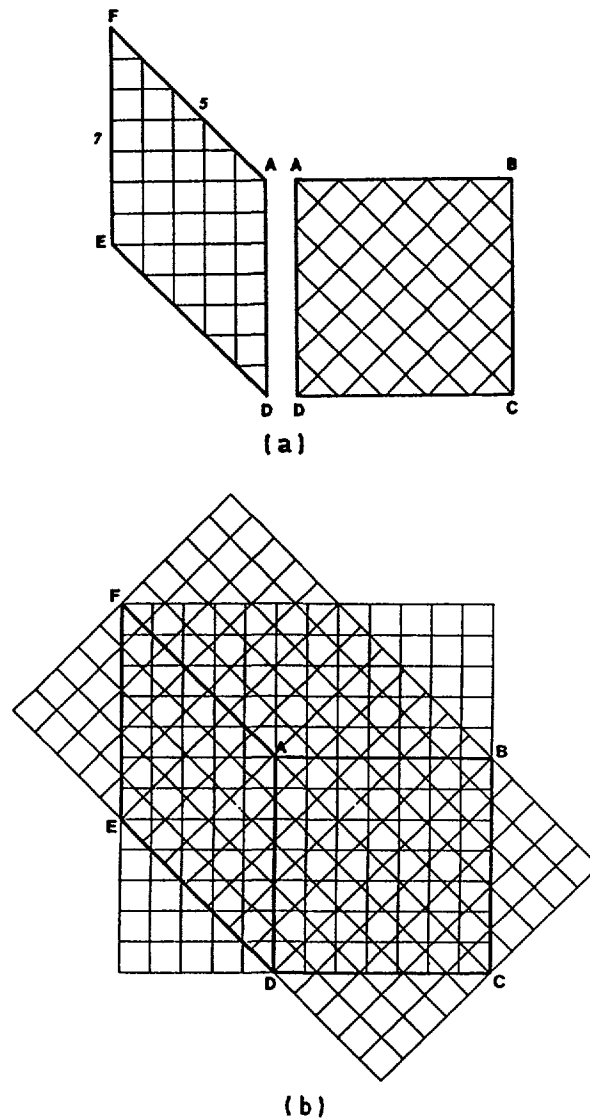


Fig. 2. — Construction of a domain in the case of octagonal symmetry. Bottom : tetragrid; A,B,C,D,E,F are points where four lines from the four families are almost intersecting in a same single point; Top : a corresponding possible choice of domains ABCD and DEFA; putting these domains together produces frustration along AD; the ratio 7:5 is close to  $\sqrt{2}$ .

twin boundaries). (b) In the construction the average lattice planes of the QC (actually a periodic subset of them) become the regular lattice planes of the microcrystal, which looks as a good starting point for a more detailed description of the phase transition. (c) This is also the key feature to understand how the frustration inside the domain boundaries as shown in the top part of figure 2 can be to a large extent only apparent. Typically a unit cell can be a large approximant. This means that the microcrystal is still very close to the bottom part of figure 2 in which a periodic

subset of the average planes, i.e. of the lines of the multigrid, is shown. The complete multigrid extends thus through the boundaries but this is hidden in the representation in terms of unit cells. Instead of truncating the unit cells at the boundaries one could also make them overlap. Due to the underlying presence of the complete multigrid the decorations of the overlapping unit cells will match rather closely. (d) However in the present paper we will not discuss the decorations of the unit cells since we expect this to be case dependent and our aim is rather at pointing out the general principles of the structure. (e) With the important relativation of point *c* in mind we would of course still like to get as close to a perfect drawing as possible, i.e. with as few tiling errors at the domain boundaries as possible, but since eventually such errors cannot be avoided we focused our efforts on minimising the strain in the direction perpendicular to the domain boundaries instead of inside of them. To justify this approach we base ourselves on the following remarks. (1) The construction reduces the dimension of the problem of the strain energy by one. In view of the typical domain sizes, this is already an appreciable reduction. (2) The domain pattern obtained in our reasoning is in turn a quasiperiodic (QP) tiling. This is an important observation which we interpret as follows : there is an intrinsic difficulty in having simultaneously long range order and crystallographic forbidden point group symmetry in a solid. The gist of this difficulty is inexorably substantiated in a QP tiling. Therefore our domain geometry indicates that all the intrinsic difficulties as regards to energy considerations, stability, geometry, etc... of a QC are found back on a larger length scale at the level of the microcrystal. For this reason we think that in our approach we have optimised the energetics of the microcrystal boundaries down to the level of the intrinsic difficulties, whose solution however at present has not yet been fully settled. The present debate on the energetics of QC is if they could be seen as random tilings or rather as deterministic tilings governed by matching rules. Putting domains together in a QP pattern with or without frustration inside the boundaries maps exactly onto those two alternatives.

Since the quasicrystalline order in question is only an average order we will use also the terminology "average quasicrystal" (AQC) for such microcrystals. This should not lead to confusion : an AQC is thus not a QC but a special type of microcrystal. The terminology microtwin has been used occasionally in the literature. We adopt the terminology microcrystal to account for the remarks given above. The solution proposed in the present paper is different from approximating the irrational slope of the strip in superspace by a piecewise linear function entirely constructed with rational slopes, since this would in general yield at least *two* different approximants, i.e. two large unit cells instead of just one. The starting point is the so-called "intertwining" construction from the previous work of the author [7]. In that work it was shown how Penrose type QP tilings constructed with the grid method can also be obtained by taking the mass centre of suitably chosen sets of points belonging to a union of periodic lattices. This construction is related to other constructions based on interpenetration. Spal [8] and Kalugin *et al.* [9] have investigated how one can build a QC by a superposition of modulated crystals. Duneau and Oguey [10] and Godrèche and Oguey [11] obtained average lattices for quasiperiodic (QP) structures by displacive transformations. However in the "intertwining" construction the periodic lattices involved cannot be considered as average lattices from which the QC would be obtained by a modulation since in the construction the number of points changes. Therefore we will not be able to predict how the atoms move in a phase transition from a QC to an AQC.

Throughout this paper we will call domains coherent if (1) they have definite orientational relationships and (2) their respective lattice planes are in phase. The basic idea of our model is to give the microcrystal a domain pattern which is an inflation of the QP Penrose like tiling and to decorate the domains with periodic lattices which are all based on the same single type of unit cell but with different orientations. Any of the orientations necessary to obtain the overall AQC symmetry fits into the inflated domains to a high level of accuracy. The higher the level of inflation used to produce the domain geometry the higher this accuracy gets. This accuracy is a measure of

the strain induced by the mismatch between the periodicities in adjacent domains at their common boundary in the direction perpendicular to this boundary. The construction preserves the phase and orientational relationships between the microdomains up to infinite correlation length. The experimental observation that in certain cases a small splitting of some of the diffraction peaks occurs when going from QC to AQC naturally leads to considering dual cells and introducing the concept of coherence length for the probing radiation.

## 2. Definitions.

Our method starts from the periodic  $n$ -grid, where  $n$  is the dimension of superspace conventionally used to describe the  $d$ -dimensional QC. For icosahedral symmetry ( $n=6, d=3$ ) the  $n$ -grid is the hexagrid of Kramer and Neri [12], for pentagonal symmetry ( $n=5, d=2$ ) it is the pentagrid of de Bruijn [13]. For octagonal symmetry ( $n=4, d=2$ ) it is a tetragrid. Let the  $n$  unit vectors used in  $\mathbb{R}^d$  to describe the QC be  $e_i, i = 1, \dots, n$ ; they are obtained by projection of vectors  $a_i, i = 1, \dots, n$  in  $\mathbb{R}^n$ . A combination of  $d$  different vectors  $e_i$  defines a tile (a rhombohedron for  $d=3$ , a rhomb for  $d=2$ ). There are  $\binom{n}{d}$  such combinations possible which we will label  $c$ . In each of the point group symmetries considered here there are two shapes of tiles. We also use the label  $c$  as an index to design a set  $S_c$  of  $d$  different unit vectors, a specific tile  $T_c$  which has as edges the  $d$  unit vectors of  $S_c$  or its dual tile  $D_c$ . In the concept tile we are including not only the shape but also its orientation. We call  $\mathcal{F}_c$  the union of the  $d$  families of planes from the  $n$ -grid which are orthogonal to the vectors of  $S_c$  and at integer distances of the origin;  $\mathcal{F}_c$  cuts real space into tiles  $D_c$ . If  $d=2$  then the tiles  $D_c$  and  $T_c$  are congruent but with a different orientation. In the three-dimensional case of icosahedral symmetry the dual tile of a prolate rhombohedron is a rhombohedron with edge angle  $\alpha = 2\pi/5$ . The dual tile of an oblate rhombohedron is a rhombohedron with  $\alpha = \pi/5$ . In the case of pentagonal and icosahedral symmetry the two shapes of tiles  $D_c$  are equally represented among the  $\binom{n}{d}$  combinations  $c$ . We will mainly focus on the case of icosahedral symmetry since this is the most complicated one; However the arguments developed can be generalised to other cases.

A point  $P$  of the QP tiling derived from the periodic hexagrid can be specified by its position vector  $OP = 1/2 \sum_{i=1}^6 K_i e_i$  with  $(K_1, K_2, \dots, K_6)$  a cell index. The intertwining construction shows that this point is (up to a scaling factor  $1/(\tau+2)$ ) the centre of mass of ten points  $P_c$  where  $P_c$  is a point of the system  $\mathcal{F}_c$  and the labels  $c$  correspond to the ten different thin (or conversely fat) tiles. Here  $\tau$  is the golden number. In each system  $\mathcal{F}_c$ , with  $c = (i, j, k)$  the point  $P_c$  is the intersection of the planes at distances  $K_i, K_j, K_k$  from the origin and measured along  $e_i, e_j, e_k$  respectively. Due to the cell index criterion these ten points are lying close to each other. The families  $\mathcal{F}_c$  make up ten interpenetrating congruent sublattices involving thus only one type of unit cell. With  $e_1 = (0, \tau, -1), e_2 = (0, \tau, 1), e_3 = (\tau, 1, 0), e_4 = (1, 0, -\tau), e_5 = (-1, 0, -\tau), e_6 = (-\tau, 1, 0)$  the "fat" combinations are  $(1, 2, 3), (1, 3, 4), (1, 4, 5), (1, 5, 6), (1, 6, 2), (3, 4, 6), (3, 5, 6), (2, 4, 6), (2, 4, 5),$  and  $(2, 3, 5)$ . They correspond to the triplets of the vertices of the triangles in the diagram of figure 1. In the Appendix we derive some additional results on the intertwining construction.

## 3. Domain structure for the AQC. First approach.

Let us now turn to the problem of the domain structure of the AQC. A purely mathematical approach consists in partitioning  $\mathbb{R}^d$  into arbitrary domains (crystallites)  $\delta$  and labeling each of

them with one of the indices  $c$ . We postulate the domain  $\delta_c$  to have as lattice planes the restriction of  $\mathcal{F}_c$  to  $\delta_c$  if  $c$  is a "thin" label and the restriction of  $\mathcal{F}_C$  if  $c$  is a "fat" label or *vice versa*; here  $C$  is the complementary set of  $c$ . By mere construction the crystallites are then coherent in phase and if we do not bias our labeling we will obtain a QC diffraction pattern. Herewith we mean vaguely that all types of orientations  $c$  should occupy more or less the same total volume so that the diffraction patterns do not display unphysical intensities. This is a boundary condition that might be due to the special origin of the AQC. In the case of external pressure this may be different. Of course in this construction one may take a periodic subset of  $\mathcal{F}_c$  to correspond to the lattice planes, which only means that we are taking a larger size of unit cell than  $D_c$ , e.g. it could be a large approximant. This way the microcrystal can also keep reminiscences of the planes of  $\mathcal{F}_C$ . We will not go further into this detail. The planes corresponding to the restriction of  $\mathcal{F}_c$  to  $\delta_c$  do not have to be the set  $\{100\}$ : other indices  $(hkl)$  are allowed as well. We will see further that this freedom results in different possibilities for microcrystal geometries.

Two fundamental questions remain to be answered: (1) How can we give a physically meaningful and precise criterion on how not to "bias"; (2) What is happening at the domain boundaries. In fact we are expecting here some defects or frustration. To answer these questions the following observation is instrumental. As can be seen e.g. for the case of pentagonal symmetry from figure 2 of reference [14] or our figure 2 for the case of octagonal symmetry - there are some regions in direct space where the  $n$ -grid has a high plane density, i.e.  $n$  planes are almost intersecting in a same point, which we will call a node. This is a consequence of theorems for Diophantine equations in number theory [15]. For any arbitrarily small number  $\varepsilon > 0$  we can find a set of nodes such that all intersection points of a node are within a distance  $\rho = \varepsilon$ . These nodes follow a QP tiling pattern. In fact in the case of 3D icosahedral symmetry the nodes represent places in space where the two periods 1 and  $\tau$  in a certain direction get into phase (they are beating in the sense of the acoustic analogue). According to equations (A3), (A4) and (A5) the nodes are for  $\varepsilon = \tau^{-3p}$  nothing else than the  $\tau^{3p}$  inflated pattern of the original QP tiling derived from the hexagrid ( $p$  is an integer).

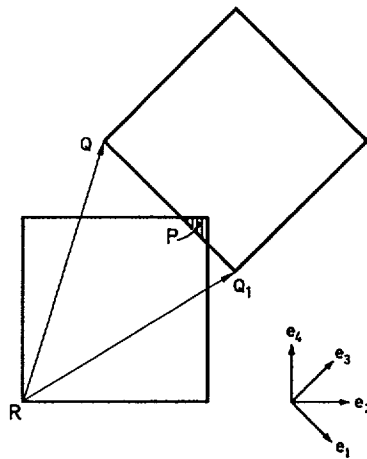


Fig. 3. — The shaded area is a cell defined by the tiles based on R and Q. The point  $Q_1$  is closer to R than Q, still it does not define a cell with R. A point  $\mathcal{P}$  inside the cell can be indexed within each family of lines by drawing a line perpendicular to each  $e_i$ :  $\mathcal{P}$  will have non integer indices  $(K_1 + \varepsilon_1, K_2 + \varepsilon_2, K_3 + \varepsilon_3, K_4 + \varepsilon_4)$ .

We call the tiles of the pattern generated by the nodes the node tiles. A plane connecting  $d$  nodes is to a very high precision  $\varepsilon$  a lattice plane for all the lattice plane systems  $\mathcal{F}_c$ . (The choice of  $(hkl\dots)$  determines which nodes have to be connected in this construction. Normally one should take planes of the type  $(1\pm 10000)$ ). This means that if we take the node tiles as domains to partition  $\mathbb{R}^d$  then we will be able to minimise the strain at the domain boundaries relative to the stacking of lattice planes in the direction perpendicular to these boundaries. This is only a partial minimisation, and therefore Figure 2 produced in the present paper shows boundaries that are not perfect. If the unit cells are large approximants these effects may be less dramatic than they look in the figure. If one draws the partial unit cells at the boundaries in full then they will overlap. This does not produce frustration if the decorations of the overlapping cells match in the region in common [7]. In any case the energy associated with the stacking of the lattice planes is a relevant physical quantity. As discussed further it is not obvious that one can do better. Of course we are not obliged to include all the nodes into the domain geometry. A realistic case will probably look like a Moiré pattern as shown in figure 3 in the work of Levine and Steinhardt[14]. The interferences seen in this Figure are due to the same effects in Diophantine approximations as in the nodes. If we restrict ourselves for the diffraction patterns to the discussion of the positions of the peaks and the requirement that the point group symmetry be respected in the intensities then only the mean geometry comes into play for it and one special choice should be sufficient to reproduce these features of the diffraction patterns. For the discussion we can thus restrict ourselves to the simple sample case where the node tile pattern is a QP tiling which is an inflation of the original QP tiling. It is important to stress that in this first approach we are trying to tile the domains which are of the Amman type ( $\cos \alpha = \pm 1/\sqrt{5}$ ) with unit cells which have the shape of the dual tiles ( $\alpha = 36^\circ/72^\circ$ ). The inflated QP Amman tiling represents exactly the domain structure where the frustration between these two incommensurate distances (cell and domain size) is minimised. In 2D the direct and dual tiles are congruent, however the problem of frustration exists here also since direct and dual tiles show a different orientation.

#### 4. Diffraction patterns.

Both the QP tiling and the diffraction pattern are described by  $\sum_i K_i \mathbf{e}_i$ . This way a diffraction spot can also be constructed as the centre of gravity of ten points belonging to ten sublattices. The ten points are replaced by their centre of gravity due to interference. There is no cell index criterion . the set of diffraction spots is dense. But one can expect that the spots which are meeting this criterion will be more intense since the interfering points are lying close to each other. Apparently this can be made more or less rigorous, at least in the following examples. In fact in the case of icosahedral symmetry the vectors  $\mathbf{a}_i$  are defined by  $\mathbf{a}_i^\parallel = \mathbf{e}_i$  and  $\mathbf{a}_1^\perp = (0, 1, \tau)$ ,  $\mathbf{a}_2^\perp = (0, 1, -\tau)$ ,  $\mathbf{a}_3^\perp = (1, -\tau, 0)$ ,  $\mathbf{a}_4^\perp = (-\tau, 0, -1)$ ,  $\mathbf{a}_5^\perp = (\tau, 0, -1)$ ,  $\mathbf{a}_6^\perp = (-1, -\tau, 0)$ . It follows that  $\mathbf{Q} = \sum_{i=1}^6 K_i \mathbf{a}_i$  projects onto  $\mathbf{Q}_\parallel = \mathbf{OP}$  and that the coordinates of  $\mathbf{Q}_\perp$  are given by the left hand sides of equations (A3) (abstraction made of the sign). It is well known that if  $\mathbf{Q}_\perp$  is small then the diffraction spot at  $\mathbf{Q}_\parallel$  is intense. In the case of pentagonal 2D symmetry the vectors  $\mathbf{a}_k$  are given by :

$$\mathbf{a}_{k+1} = (\cos 2k\pi/5, \sin 2k\pi/5, \cos 6k\pi/5, \sin 6k\pi/5, 1/\sqrt{2}) \quad (1)$$

where  $k = 0, 1, \dots, 4$ . This leads to intensity conditions  $\sum_{i=1}^5 K_i \rightarrow 0$  and  $K_1 \mathbf{e}_1 + K_2 \mathbf{e}_4 + K_3 \mathbf{e}_2 + K_4 \mathbf{e}_5 + K_5 \mathbf{e}_3 \rightarrow 0$ . By eliminating  $\mathbf{e}_1$  using the first condition after some algebra the second condition reduces to  $(K_2 \mathbf{b}_5 - K_5 \mathbf{b}_2) - \tau (K_4 \mathbf{b}_3 - K_3 \mathbf{b}_4) \rightarrow 0$  expressing the fact that the points  $P_{(25)}$  and  $P_{(34)}$  are lying close to each other. The  $\mathbf{b}_i$  are here vectors homothetic to the vectors

$e_i$  but rotated over  $\pi/2$  in the counterclockwise sense. Once the sublattices used in the intertwinning construction are chosen the splitting of the diffraction spots becomes unambiguous : the vectors  $e_i$  (or  $e_i - e_1$ , with  $i \neq 1$  in the pentagonal case) are linearly independent if the  $K_i$  ( $K_i - K_1$  in the pentagonal case) are restricted to  $\mathbb{Z}$ . Remark that this shows that in the case of microdomains a splitting of the diffraction peaks produces a peak broadening proportional to  $Q_{\perp}$ . The proportionality factor can depend on the size of the domains through the form factors.

## 5. Other schemes for domain geometries.

(a) In the phase transition of AlFeCu some spots now split. It is tempting to interpret these multiple spots as contributions from different domains constructed according to the procedure outlined above. This can however not be correct. The subspots are spanned by vectors  $e_i \wedge e_j$  while the sets  $\mathcal{F}_c$  still will provide spots that are spanned by  $e_i$ . The first approach described above corresponds thus rather to the merohedric case where the diffraction spots do not split. If the domains are coherent then the diffraction pattern will depend on the coherence length of the probing radiation. If the domains are smaller than the coherence length they can still interfere and no change in the diffraction pattern occurs except changes in the form factors. If the domains are larger then the spots will split (except in the merohedric case) and the set of spots becomes non dense. Subspots spanned by  $e_i \wedge e_j$  are produced by domains for which the Amman tiles describe the orientations of the twinning planes and the unit cell is one type of Amman tile. The twin plane construction above is still useful since it provides a partitioning of direct space where the transition from the  $n$ -grid to an Amman tile based lattice costs the least of (stacking) energy. This can be understood as follows. Both Amman tiles have the same shape of rhombus as faces. A "fat" Amman domain can be filled with fat tiles by stacking equidistant planes parallel to its faces. The same stacking can be used to fill a "thin" Amman domain if it has the appropriate size: take e.g.  $(e_1, e_2, e_3)$  as a combination to define the directions of the edges of a fat domain, and  $(e_1, e_2, e_4)$  to define a thin domain. The heights  $h_3$  of  $e_3$  and  $h_4$  of  $e_4$  above the plane defined by the vectors  $(e_1, e_2)$  are in the ratio  $[e_1 e_2 e_3] : [e_1 e_2 e_4] = \tau$ . These heights are both measured along one of the 15 directions of the edges of the 5 cubes inscribed in the dodecahedron, e.g. along  $e_3 - e_6$  in our example. This means that if we take a thin domain height  $H_4 h_4$  such that  $\exists H_3 \cdot |H_4 h_4 - H_3 h_3| = h_4 |H_4 - H_3 \tau| < \varepsilon$  according to a beat of the type expressed by equations (A3) then the thin Amman domain of height  $H_4$  will contain to a high precision  $H_3$  planes stacked as in a fat domain. In the intertwinning construction a node is also a point of the QP pattern and the nodes are thus the points that are particularly stable in such a construction. Hence our construction of domain boundaries minimises the frustration in the stacking of the planes at those boundaries in the directions perpendicular to them. However it does not minimise the strain in the directions inside a boundary : the planes parallel to  $(e_2, e_3)$  and to  $(e_3, e_1)$  are cutting the face parallel to  $(e_1, e_2)$  into rhombs, but the faces parallel to  $(e_2, e_4)$  or  $(e_4, e_1)$  are cut into parallelograms with edges in the ratio  $H_3 \cdot H_4 \simeq \tau : 1$ . Even situations  $\tau : \tau$  are possible. The 2D analogon of this situation is illustrated in figure 2. It is far from clear if a global coherence between the domains can be obtained which minimises also faults in the directions inside the boundaries. This depends on the relative orientations of the lattices inside adjacent domains. This is thus a problem of decorating the domains with lattices such that the decorations match at the common boundaries, i.e. it is related to the existence of matching rules. For the case of icosahedral symmetry no simple matching rules exist [14]. Even in the case of the 2D Penrose tiling the decoration of Penrose node tiles with periodic lattices would require that the parallel edges of the tiles would have the same decoration, which is exactly the contrary of the actual matching rules. (This remark may be an argument in favour of the random tiling model). Therefore further

optimisation of the domain structure requires a much deeper study. The smaller the value of  $\varepsilon$ , the larger the node tiles are : the domain size increases according to a sequence  $N + \tau M$  with critical values at  $\varepsilon_p = |\tau N - M| \sim \tau^{-3p}$ . This mismatch value  $\varepsilon_p$  adds to the (stacking part of the ) energy  $U$  of the system in the form of strain at the domain boundaries. Only special relative orientations of the lattices inside the domains allow to make also the error in the directions inside the common boundaries small. The AQC can however minimise the strain further by remaining QP (in one dimension less) inside the domain boundary. Such a possibility has been described in reference [16]. The situation we propose at the boundaries is thus not more exotic than in other existing cases. (It also illustrates well the physical idea behind the intertwinning construction). It remains a problem to find a displacive transformation describing the actual transition from the QC structure to the structure based on an Amman unit cell : as mentioned in the introduction the periodic lattices obtained from the multigrid cannot be considered as average lattices from which the QC could be obtained by modulation. For the geometry in direct space our proposition for the domain geometry agrees well with the result of a superb paper by Audier and Guyot [17]. The difference is that in our case this structure has been derived starting from a general approach.

(b) One cannot exclude that the transition from the QC to the microcrystal would be not immediate and could show an intermediate regime with a modulated structure. The unit cells need not to be a type of Amman or dual tile. For instance there could be a microstructure of cubic lattices with five different orientations corresponding to those of the five cubes that can be inscribed in a regular dodecahedron. Note that in each Amman domain three cubes are immediately evident. E.g. in the case of a domain with edges in the directions  $(e_1, e_2, e_3)$  one set of cubic directions is given by  $e_1 \pm e_2$  inside a face  $(e_1, e_2)$  and  $e_3 - e_6$  in the direction of the height  $H_3$  as explained in the previous paragraph. Within each of these five cubic reference systems the QC is closely related to the structure one obtains by intertwinning two cubic grids with periods 1 and  $\sqrt{5}$ . In fact equations (A1-A3) are written in one of these five systems. The expressions  $N + M\tau$  such that  $M - N\tau = \varepsilon$  can be rewritten as  $1/2(A + B\sqrt{5})$  such that  $A - B\sqrt{5} = -2\varepsilon/\tau$  by putting  $A = M + 2N$ ,  $B = M$  (The equations (A1-A3) do not represent an intertwinning of two periods 1 and  $\tau$ ). However the cell index criterium connected with the hexagrid cannot be reformulated as an independent criterium for a grid obtained from  $e_x, e_y, e_z, \sqrt{5}e_x, \sqrt{5}e_y, \sqrt{5}e_z$ . Two of the cubic lattices always have a threefold axis in common. The boundary planes represent a beating of the periods 1 and  $\sqrt{5}$  in the main directions of the cubic lattices. A transition could be characterised by a change of the periods to a commensurate ratio according to  $\sqrt{5} \rightarrow (F_{p+1} + F_{p-1})/F_p$ . The corresponding splitting of diffraction peaks could be calculated from equations (A1) and (A3). This way we have described three different scenarios for microcrystal geometries.

(c) If we want that all lattice orientations within the AQC have the same weight (in order to respect the point group symmetry for the diffraction intensities) we have to establish a rule that gives the lattice orientation inside each domain. The easiest way to achieve this is to define a bijection between the orientation of the domain tile and the lattice orientation (We know that in the inflated tiling all orientations of the same shape of tile have the same weight). We have called this labeling. We can now sketch a rule for labeling the domains. A boundary between two domains is a plane orthogonal to one of the vectors  $v_k$ . It also represents a place in space where the two periods 1 and  $\tau$  occurring in this direction get into phase. In figure 1 we can see that a self-consistent procedure consists in labeling the domains at both sides of a boundary with the combinations given by the vertices of two adjacent triangles in the Schlegel diagram of the icosahedron and corresponding to Amman tiles with the same orientations as the node tiles. The choice of a lattice orientation inside a domain corresponds to a decoration for the node tiling. The choice which indices  $(hkl)$  we will attribute to the planes of  $\mathcal{F}_c$  corresponds to a choice of microcrystal geometry and a unit cell as explained above. Of course this procedure should be considered as an existence proof leading to an acceptable average structure rather than an image

of the actual physical situation which might be much more random and will also depend on the specific value of the structure factor.

## 6. Conclusion.

We have shown how the intertwining construction is very appropriate to study problems of microcrystals with average QC structures. Different microcrystal geometries are possible, the ultimate choice depending on the experimental data. The present findings stress the importance of the question raised by Duneau and Oguey [10] which has not received a fully satisfactory answer yet: how do the atoms move from a QP lattice to a crystalline lattice in a transition between related phases. It seems urgent to find a modification of the intertwining construction that conserves the density of points. However the observation that in the phase transitions the QC always breaks up to a microcrystal rather than becoming a monocrystal may be tantamount to the experimental manifestation of the fact that there is no such thing as a single average lattice for a QC (in contradistinction to the situation in some other incommensurate structures). To check the model proposed here further it would be interesting to explore the domain geometry in the low temperature AQC phase of AlFeCu to see if it really exhibits a Moiré pattern corresponding to a Fibonacci 15-grid. Small angle scattering should be able to show the existence of such a grid. Some parts of our model are reminiscent of the work of Stephens and Goldman [18] but in our case the structure is much more ordered. Especially important is that our model predicts intensities and peak widths which can depend on  $Q_{\perp}$  and describes the splitting of diffraction peaks.

## Acknowledgements.

The author is indebted to Prof. G. Heger and Drs. P.A. Bancel and P. Launois for helpful discussions.

## Appendix

From the coordinates of  $e_i$  one obtains for P :

$$\mathbf{OP} = 1/2((K_3 - K_6)\tau + (K_4 - K_5); (K_1 + K_2)\tau + (K_3 + K_6); -(K_4 + K_5)\tau + (K_2 - K_1)) \quad (\text{A1})$$

It should be mentioned that the Euclidean distance function is here not very well suited to express the closeness criterion according to which the points  $P_c$  have to merge into a common centre of gravity : this can be seen in the intertwining construction for eightfold symmetry illustrated in figure 3. There exist pairs of points  $(R, Q)$  and  $(R, Q_1)$  such that  $R$  and  $Q$  merge and  $R$  and  $Q_1$  do not merge while  $|RQ_1| < |RQ|$ . This way the cell index criterion cannot be related to a single Euclidean distance. There exists however a different distance function  $\rho$  such that the cell index criterion can be expressed by means of one specified distance. This distance is defined by :

$$\rho(P_1, P_2) = \sup_{i=1}^6 | \mathbf{P}_1 \mathbf{P}_2 \cdot \mathbf{e}_i | = \| \mathbf{P}_1 \mathbf{P}_2 \|_{\rho} \quad (\text{A2})$$

Of course a similar remark applies to this distance function; it allows however a simple description of the cell index criterion. If  $(K_1, K_2, K_3, K_4, K_5, K_6)$  is a cell index then  $\mathcal{D} = \bigcap_c D_c(P_c) \neq \emptyset$ ,

with  $D_c(\mathcal{P}_c)$  the tile in between the planes  $K_i, K_i + 1, K_j, K_j + 1, K_k, K_k + 1, (K_i, K_j, K_k) = c$ . This means that there exists a point  $\mathcal{P} \in \mathcal{D}$  which we could index by the non integer numbers  $K_i + \varepsilon_i$ . By expressing that  $\mathcal{P} \in D_{(123)} \cap D_{(126)}, \mathcal{P} \in D_{(145)} \cap D_{(245)}, \mathcal{P} \in D_{(345)} \cap D_{(456)}$  we obtain equations translating the fact that the pairs of points  $P_{(345)}/P_{(456)}, P_{(123)}/P_{(126)}$ , and  $P_{(145)}/P_{(126)}$  are lying "close" to each other and find some of the conditions for a point of the QP tiling :

$$(\exists \varepsilon = (\varepsilon_1, \varepsilon_2, \varepsilon_3, \varepsilon_4, \varepsilon_5, \varepsilon_6) \in \mathbb{R}^6)(\|\varepsilon\| < 1) :$$

$$(K_4 - K_5)\tau - (K_3 - K_6) = -(\varepsilon_4 - \varepsilon_5)\tau + (\varepsilon_3 - \varepsilon_6) \tag{A3a}$$

$$(K_3 + K_6)\tau - (K_1 + K_2) = -(\varepsilon_3 + \varepsilon_6)\tau + (\varepsilon_1 + \varepsilon_2) \tag{A3b}$$

$$(K_1 - K_2)\tau - (K_4 + K_5) = -(\varepsilon_1 - \varepsilon_2)\tau + (\varepsilon_4 + \varepsilon_5) \tag{A3c}$$

These equations follow from  $OP_{(ijk)} = (\sum_{\text{cycl}} K_i e_j \wedge e_k) / [e_i e_j e_k]$  and table I. ( The sum is over cyclic permutations of  $(i, j, k)$ . The norm used is the supremum norm in  $\mathbb{R}^6$ ).

Table I. — Values of  $(k, l)$  such that  $e_i \wedge e_j = e_k + e_l$  with the convention  $e_{-k} = -e_k$ .

$j$	2	3	4	5	6
$i$					
1	(3, -6)	(4, -2)	(5, -3)	(6, -4)	(2, -5)
2		(1, 5)	(5, 6)	(-3, -4)	(-4, -1)
3			(1, 6)	(2, 6)	(-4, -5)
4				(1, 2)	(2, 3)
5					(1, 3)

Consider the 30 vectors  $v_k$  of the form  $(\pm e_i \pm e_j) / |\pm e_i \pm e_j|$ . For each pair of vectors  $\pm v_k$  the family  $\mathcal{G}_k$  consists of planes orthogonal to  $v_k$  and at distances from the origin  $O$  given by  $(N\tau + M)/2$  with  $(N, M) \in \mathbb{Z}^2$  such that  $1/2 | M\tau - N | < 1/2(\tau + 1)$ . We call the union of the 15 families  $\mathcal{G}_k$  a Fibonacci grid. The vectors  $v_k$  are pointing to the centres of the edges of an icosahedron. The planes have the directions of the faces of a triacontahedron or of the faces of the Amman tiles with their different orientations. Equation (A3) with equation (A1) shows that  $P$  belongs to the families  $\mathcal{G}_k$  defined by the vectors  $v_k$  which are parallel to the axes  $Ox, Oy$ , and  $Oz$ . The choice of the coordinate system is thus such that the planes  $Oxy, Oyz$ , and  $Ozx$  correspond to the faces of one of the five cubes that can be inscribed inside the dodecahedron defined by the unit vectors  $e_i$ . By symmetry it follows that  $P$  belongs to the 15 families of the Fibonacci grid. The number  $\tau$  is an inflation for the 15-grid and for the QP tiling but not for the hexagrid. This follows from :

$$N\tau - M = \varepsilon \Rightarrow M\tau - (N + M) = -\varepsilon/\tau \tag{A4}$$

and

$$\tau(M\tau + N) = (N + M)\tau + M \tag{A5}$$

Define  $\bar{K}_1, \bar{K}_2, \bar{K}_3, \bar{K}_4, \bar{K}_5, \bar{K}_6$  by :

$$\bar{K}_3 - \bar{K}_6 = 3(K_3 - K_6) + 2(K_4 - K_5) \quad (\text{A6a})$$

$$\bar{K}_4 - \bar{K}_5 = 2(K_3 - K_6) + (K_4 - K_5) \quad (\text{A6b})$$

$$\bar{K}_1 + \bar{K}_2 = 3(K_1 + K_2) + 2(K_3 + K_6) \quad (\text{A6c})$$

$$\bar{K}_3 + \bar{K}_6 = 2(K_1 + K_2) + (K_3 + K_6) \quad (\text{A6d})$$

$$\bar{K}_4 + \bar{K}_5 = 3(K_4 + K_5) + 2(K_1 - K_2) \quad (\text{A6e})$$

$$\bar{K}_1 - \bar{K}_2 = 2(K_4 + K_5) + (K_1 - K_2) \quad (\text{A6f})$$

The  $\tau^3$  inflation maps  $\text{OP} = \sum_{i=1}^6 K_i \mathbf{e}_i$  onto  $\tau^3(\text{OP}) = \sum_{i=1}^6 \bar{K}_i \mathbf{e}_i$ . According to equation (A4) the errors  $\varepsilon$  in equation (A3) are thereby reduced by a factor  $\tau^3$ . The fact that  $\tau$  or  $\tau^2$  do not work as inflations for the QP tiling comes from the requirement that the  $\bar{K}_i$  must be integers.

#### References

- [1] For reviews on quasicrystals see : P.J. Steinhardt and S. Ostlund eds., *The Physics of Quasicrystals* (World Scientific Publishing Co, Singapore, 1987);  
Ch. Janot and J.M. Dubois eds., *Quasicrystalline Materials* (World Scientific Publishing Co, Singapore, 1988);  
HENLEY C.L., *Comm. Cond. Mat. Phys.* **13** (1987) 59;  
JANOT Ch. and DUBOIS J.M., *J.Phys.* **F18** (1988) 2303.
- [2] KALUGIN P.A., KITAEV A.Yu. and LEVITOV L.C., *JETP Lett* **41** (1985) 145; *J. Phys. Lett. France* **46** (1985) L601;  
ELSER V., *Phys. Rev. Lett.* **54** (1985) 1730; *Phys. Rev.* **B32** (1985) 4982;  
DUNEAU M. and KATZ A., *Phys. Rev. Lett.* **54** (1985) 2477;  
KATZ A. and DUNEAU M., *J. Phys. France* **47** (1986) 181.
- [3] MOSSERI R., OQUEY C. and DUNEAU M., *Quasicrystalline Materials* (World Scientific Publishing Co., Singapore, 1987) p.224.
- [4] PAULING L., *Nature* **317** (1985) 512; *Phys. Rev. Lett.* **58** (1987) 365. The cubic lattice planes  $(F_{n+1}, \pm F_n, 0)$ ,  $(0, F_{n+1}, \pm F_n)$ ,  $(\pm F_n, 0, F_{n+1})$  where  $F_n$  and  $F_{n+1}$  are taken from the Fibonacci sequence define pentagonododecahedra that approximate the shape of the regular dodecahedron to any desired precision. The case  $F_{n+1} = 2$  is occasionally found in text books (see e.g. SIROITINE Y. and CHASKOLSKAJA M., *Fondements de la physique des cristaux* (Mir ed., Moscow, 1984)).
- [5] JIANG W.J., HEI Z.K., GUO Y.X. and KUO K.H., *Philos. Mag.* **A52** (1985) L53;  
WANG N., CHEN H., KUO K.H., *Phys. Rev. Lett.* **59** (1987) 1010.
- [6] BENDERSKY L.A., CAHN J.W., and GRATIAS D., *Philos. Mag.* **B60** (1989) 837;  
DENOYER F., HEGER G., LAMBERT M., AUDIER M. and GUYOT P., *J. Phys. France* **51** (1990) 651;

- AUDIER M. and GUYOT P., AAR Conf. ICTP, Trieste (July 1989) to be published; 3rd International Meeting on Quasicrystals, incommensurate structure in condensed matter, 27 May - 2 June 1989, Vista Hermosa, Mexico, to be published;
- BANCEL P.A., *Phys. Rev. Lett.* **63** (1989) 2741.
- [7] CODDENS G., *Solid State Comm.* **65** (1988) 637; *Int. J. Mod. Phys. B4* (1990) 347; Colloque Quasicristaux, Orsay, 29-30 March 1990, p. 7.
- [8] SPAL R.D., *Phys. Rev. Lett.* **56** (1986) 1823.
- [9] KALUGIN P.A. and LEVITOV L.C., *Int. J. Mod. Phys. B3* (1989) 877.
- [10] DUNEAU M. and OGUEY C., *J. Phys. France* **51** (1990) 5.
- [11] GODRÈCHE C. and OGUEY C., *J. Phys. France* **51** (1990) 21.
- [12] KRAMER P. and NERI N., *Acta Cryst.* **A40** (1984) 580.
- [13] DE BRUIJN N.G., *Kon. Nederl. Akad. Wetensch. Proc. Ser. A84* (1981) 39.
- [14] BAK P., *Phys. Rev.* **B32** (1985) 5764;  
see also Fig. 3 of LEVINE D. and STEINHARDT P., *Phys. Rev.* **B34** (1986) 594, which shows a Moiré pattern that could be a domain pattern.
- [15] see e.g. DESCOMBES R., *Eléments de théorie des nombres* (Presses Universitaires de France, Paris 1986) theorem 2.2.1 on p. 52.
- [16] RIVIER N. and LAWRENCE J.A., *Quasicrystalline Materials*, Ch. Janot and J.M. Dubois eds. (World Scientific Publishing Co, Singapore, 1988) p. 255.
- [17] AUDIER M. and GUYOT P., *Acta Metall.* **36** (1988) 1321.
- [18] STEPHENS P.W. and GOLDMAN A.I., *Phys. Rev. Lett.* **56** (1986) 1168;  
HENDRICKX S. and TELLER E., *J. Chem. Phys.* **10** (1942) 147.

Cet article a été imprimé avec le Macro Package "Editions de Physique Avril 1990".