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Michel Duneau. Modulated quasicrystals : a mechanism induced by 6D shears. Journal de Physique I, 1992, 2 (9), pp.1731-1740. <10.1051/jp1:1992241>. <jpa-00246655>

HAL Id: jpa-00246655

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Submitted on 4 Feb 2008

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HAL Authorization

Classification

Physics Abstracts

61.50J — 61.50K — 61.70N — 64.70K

Modulated quasicrystals : a mechanism induced by 6D shears

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(Received 17 March 1992, accepted 4 June 1992)

Résumé. — Des quasicristaux modulés ont été observés récemment comme états intermédiaires possibles dans une transformation structurale entre une phase icosaédrique et une phase rhomboédrique périodique du système AlFeCu. Nous proposons un mécanisme basé sur des cisaillements « fractionnaires » à 6D produisant des structures quasipériodiques modulées. Ces cisaillements fractionnaires sont bornés, localement linéaires et transforment le réseau cubique à 6D en un réseau pour lequel l'espace physique est rationnel. En particulier, les nœuds 6D associés à une structure quasipériodique subissent de faibles déplacements qui engendrent les structures modulées après projection. Ces structures ont des plans de défauts caractéristiques parallèles aux faces de la maille rhomboédrique primitive et leurs transformées de Fourier montrent des satellites déplacés le long des axes 5, comme observé expérimentalement.

Abstract. — Modulated quasicrystals have been recently observed as possible intermediate states in structural transformations between an icosahedral phase and a periodic rhombohedral one in the AlFeCu system. We propose a mechanism based on « fractional » shears in 6D which yields modulated quasiperiodic structures. These fractional shears are bounded and locally linear mappings which transform the 6D cubic lattice into lattices for which the 3D physical space is rational. In particular, the 6D nodes associated to quasiperiodic structures undergo small displacements which give rise to modulated structures after projection. These structures show characteristic planar defects parallel to the facets of the primitive rhombohedral cell and their Fourier transforms show satellite peaks shifted in the 5-fold directions as experimentally observed.

1. Introduction.

In [1], a reversible transformation between an icosahedral phase of composition $\text{Al}_{63.5}\text{Fe}_{12.5}\text{Cu}_{24}$ and a microcrystalline rhombohedral phase is reported on the basis of TEM observations. An intermediate modulated icosahedral phase is observed during thermal treatments and the structure of the satellite spots is described [1, 2].

In this paper, we propose a mechanism for displacive transformations between a perfect icosahedral phase and a modulated icosahedral phase, with characteristic planar defects and

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satellite spectrum, which could afterward be related to a rhombohedral microcrystalline phase by means of local diffusion.

The mechanism is based on the existence of simple *bounded* transformations which map the 6D face-centered cubic lattice L onto lattices for which the 3D physical space is rational. More precisely, we define different 6D linear mappings, as product of 3 shear transformations with lattice directions, which transform L into a lattice L' containing the physical space E_{\parallel} as a 3D lattice subspace. The corresponding intersection lattice is rhombohedral with primitive axes parallel to 2-fold axes and angle 36° . Then, different *bounded* transformations between L and L' are obtained by replacing each linear shear by the corresponding « fractional » shear as defined in [4, 5]. These bounded transformations can then be applied to the set of nodes of L selected by a cut algorithm, whatever the acceptance domains or atomic surfaces. This results in a set of nodes of the lattice L' , still close to E_{\parallel} . This set differs from any set selected through a definite cut, as for an approximant structure, by two kinds of specific defects. The projection of the nodes finally gives a modulated icosahedral structure which shows a characteristic network of domains bounded by planar defects parallel to the facets of the primitive rhombohedral cell. The Fourier transform of this structure can be computed analytically. The fractional shears produce particular modulations which give rise to well defined satellites peaks for the projected structure involving Δq along 5-fold directions.

2. Shear transformations and fractional shears.

A shear is a linear transformation which reads $S = I + |s\rangle \langle \sigma|$ so that $S(x) = x + \langle \sigma | x \rangle s$. We shall assume that s is an irreducible vector of some lattice L (the indices of s with respect to a basis of L are coprime) and that the covector σ , which specifies a 5D invariant plane, satisfies $\langle \sigma | x \rangle = 0$, a condition equivalent to $\det(S) = 1$. Then s is also an irreducible lattice vector of the transformed lattice $S(L)$ so that L and $S(L)$ have at least a 1D intersection. This mapping is unbounded in the sense that $S(x) - x$ diverges when x runs over L . The corresponding « fractional » shear Σ is defined [4, 5] by $\Sigma(x) = x + \text{Frac} [\langle \sigma | x \rangle] s$. In this paper, $\text{Frac} [t]$ denotes the fractional part of t , between $-1/2$ and $1/2$, so that $t = \text{Round} [t] + \text{Frac} [t]$ and $\text{Round} [t]$ denotes the nearest integer. The fractional shear satisfies $\Sigma(x) = S(x) - \text{Round} [\langle \sigma | x \rangle] s$. Since s belongs to L and $S(L)$, we see that the fractional shear Σ also maps L onto $S(L)$. The displacement field $\Sigma(x) - x = \text{Frac} [\langle \sigma | x \rangle] s$ is parallel to the lattice vector s and is bounded by $\|s\|/2$ so that Σ is a *bounded* one to one mapping between L and $S(L)$. Actually Σ maps any point x of L onto the nearest point of $S(L)$ lying on the common lattice line parallel to s . It is proven in [5] that such fractional shears are the most simple bounded one to one mappings between lattices which involve a 1D displacement field. The fractional shear Σ is locally linear and its discontinuities are localized on the planes $\langle \sigma | x \rangle - 1/2 \in \mathbb{Z}$ where $\text{Round} [\langle \sigma | x \rangle]$ undergoes discontinuities. The modulation $\text{Frac} [\langle \sigma | x \rangle] s$ is periodic although generally incommensurate with the lattice L . In Fourier space, such a term will generate satellite peaks with $\Delta q = n\sigma$ (n integer).

A linear shear preserves the density but any two lattices L and L' of the same density cannot, in general, be related by a single shear transformation. On the contrary, $n + 1$ shears are generically required to map a n -dimensional lattice L on a lattice L' [4]. In any case, if a product of shear transformations maps a lattice L onto a lattice L' then a bounded one to one mapping from L onto L' is readily obtained by replacing each linear shear by its bounded fractional version.

3. Rhombohedral approximants and related shear transformations.

The icosahedral quasicrystal of composition close to $\text{Al}_{63.5}\text{Fe}_{12.5}\text{Cu}_{24}$ was first reported in [6] and extensively studied by other groups (see for instance [7, 8, 9]). This icosahedral phase can be indexed by the reciprocal lattice of the 6D face-centered cubic lattice $L = F(2a)$ which is of index 2 in the cubic lattice $P(a) = aZ^6$ of parameter $a \approx 6.31 \text{ \AA}$. A reversible transformation, induced by thermal treatments, between this phase and a microcrystalline rhombohedral phase with overall icosahedral symmetry was reported in [1]. An intermediate modulated icosahedral phase was also observed in TEM [1, 2] and X-ray diffraction [3].

In view of a quasicrystal to crystal displacive transformation, we look for shear transformations which map the lattice L onto a lattice L' for which the physical space E_{\parallel} is rational and contains a rhombohedral lattice. The rational 3D space related to the rhombohedral approximant observed in [1] is spanned by the following lattice vectors (up to the labelling of the axes) of norm $\approx 32.17 \text{ \AA}$:

$$\begin{aligned} A_1 &= 3[12] + 2[\bar{4}6] = (3, 3, 0, \bar{2}, 0, 2) \\ A_2 &= 3[26] + 2[13] = (2, 3, 2, 0, 0, 3) \\ A_3 &= 3[16] + 2[25] = (3, 2, 0, 0, 2, 3). \end{aligned} \quad (3.1)$$

The projections of these vectors in E_{\parallel} span a rhombohedral lattice of parameter $\approx 32.16 \text{ \AA}$ with angle 36° which is closely related to an Ammann prolate rhombohedra [1] (see Fig. 1).

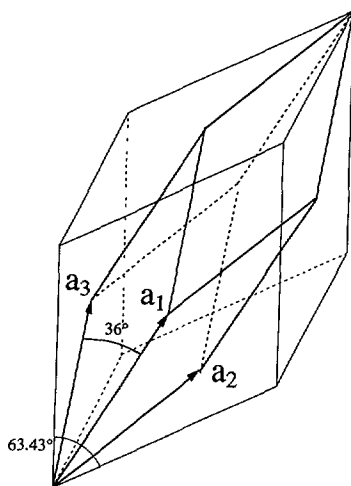


Fig. 1. — The primitive cell of the rhombohedral approximant lattice compared to the Ammann prolate rhombohedra (arbitrary scale). The basis vectors are parallel to 2-fold axes and the angle is 36° .

Each lattice vector A_i belongs to a 2D lattice plane of L , a 2-fold axis of the icosahedral group, which cut the physical space along a line of slope $\tau^{-1} = (\sqrt{5} - 1)/2$. We choose the following shear vectors in order to preserve the 3-fold symmetry :

$$\begin{aligned} s_1 &= m[12] + n[\bar{4}6] = (m, m, 0, -n, 0, n) \\ s_2 &= m[26] + n[13] = (n, m, n, 0, 0, m) \\ s_3 &= m[16] + n[25] = (m, n, 0, 0, n, m) \end{aligned} \quad (3.2)$$

where m and n are integers to be specified.

In accordance with the observed satellite structure, we look for covectors σ_1 , σ_2 and σ_3 which belong to 5-fold axes. They must satisfy the conditions $\langle \sigma_i | s_i \rangle = 0$ for all $i = 1, 2, 3$. The most simple solution is given by :

$$\begin{aligned}\sigma_1 &= x[\bar{4}] + y[12\bar{3}\bar{5}\bar{6}] = (y, y, -y, -x, -y, -y) \\ \sigma_2 &= x[3] + y[\bar{1}24\bar{5}6] = (-y, y, x, y, -y, y) \\ \sigma_3 &= x[5] + y[1\bar{2}\bar{3}46] = (y, -y, -y, y, x, y)\end{aligned}\quad (3.3)$$

where x and y are real parameters to be specified as functions of m and n .

The orthogonality condition reads $\langle \sigma_1 | s_1 \rangle = \langle \sigma_2 | s_2 \rangle = \langle \sigma_3 | s_3 \rangle = nx + (2m - n)y = 0$, which gives a first equation for x and y . Furthermore we see that $\langle \sigma_i | s_j \rangle = 0$ for all $i \neq j$ so that the three shears $S_i = I + |s_i\rangle \langle \sigma_i|$ are uncoupled and commute. The corresponding product \hat{S} is given by :

$$\hat{S}(x) = \left[\prod_{i=1}^3 S_i \right] (x) = x + \sum_{i=1}^3 \langle \sigma_i | x \rangle s_i. \quad (3.4)$$

The amplitudes x and y are finally specified by the condition that $S_i(A_i)$ is in E_{\parallel} , for $i = 1, 2, 3$, so that the 3D space spanned by A_1 , A_2 and A_3 is mapped onto E_{\parallel} . In other words, the transformed lattice $\hat{S}(L)$ contains E_{\parallel} as a 3D rational subspace spanned by $a_1 = \hat{S}(A_1)$, $a_2 = \hat{S}(A_2)$ and $a_3 = \hat{S}(A_3)$. Since

$$\begin{aligned}S_1(A_1) &= A_1 + \langle \sigma_1 | A_1 \rangle s_1 = A_1 + 2(x + 2y) s_1 \\ &= [3 + 2(x + 2y)m](1, 1, 0, 0, 0, 0) + 2[1 + (x + 2y)n](0, 0, 0, -1, 0, 1)\end{aligned}$$

this second condition simply gives : $3 + 2(x + 2y)m = 2\tau[1 + (x + 2y)n]$ from which follows the following equations for x and y :

$$x = \frac{(2m - n)(2\tau - 3)}{2(2m - 3n)(m - n\tau)} \quad y = \frac{-n(2\tau - 3)}{2(2m - 3n)(m - n\tau)}. \quad (3.5)$$

The 3D lattice $\hat{S}(L) \cap E_{\parallel}$ is a rhombohedral lattice and its primitive parameter a_R is a function of m and n given by :

$$a_R = |\hat{S}(A_i)| = \sqrt{2} a \sqrt{[3 + 2(x + 2y)m]^2 + 4[1 + (x + 2y)n]^2}. \quad (3.6)$$

The angle is 36° since the generators are parallel to 2-fold axis $A_{1\parallel}$, $A_{2\parallel}$ and $A_{3\parallel}$.

Since each shear S_i of the product (3.4) has a bounded « fractional » version Σ_i , as explained in section 2, the corresponding product $\hat{\Sigma}$ of fractional shears gives a bounded transformation between L and the lattice L' which reads :

$$\hat{\Sigma}(x) = \left[\prod_{i=1}^3 \Sigma_i \right] (x) = x + \sum_{i=1}^3 \text{Frac} [\langle \sigma_i | x \rangle] s_i. \quad (3.7)$$

The « modulation » $\hat{\Sigma}(x) - x$ is clearly bounded by $\Sigma \|s_i\|/2$. In particular, the nodes of L which are close to E_{\parallel} , such as those selected by a cut algorithm, are mapped onto nodes of L' which remain close to the physical space. However, since the modulation has a perpendicular component ($s_{i\perp}$ does not vanish), the property that x_{\perp} belongs to some fixed

acceptance domain need not be conserved for $\hat{\Sigma}(x)$. Figure 2 below is a 2-dimensional illustration of this feature for a single fractional shear transformation in the plane.

After projection into E_{\parallel} we get :

$$\hat{\Sigma}(x)_{\parallel} = x_{\parallel} + \sum_{i=1}^3 \text{Frac} [\langle \sigma_i | x \rangle] s_{i\parallel} \tag{3.8}$$

so that the modulation $\hat{\Sigma}(x)_{\parallel} - x_{\parallel}$ is bounded by $\Sigma \|s_{i\parallel}\|/2$.

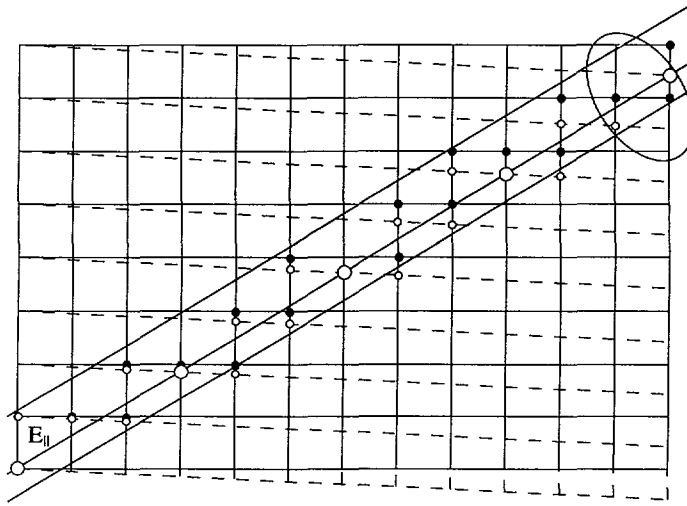


Fig. 2. — The initial quasiperiodic set is represented by black circles on the square lattice L. The shear S has direction (0, 1) and transforms L into the dashed lattice L' for which the line E_{\parallel} is rational. The intersection lattice (large white circles) is spanned by the vector (3, 2) of L'. The fractional shear Σ transforms the initial set into the set of white circles. At the upper right a discontinuity of Σ is shown. Notice that the white circles don't correspond to a definite cut of L' but to a union of different unit cells of the approximant.

The discontinuities of $\hat{\Sigma}$ are localized on 3 families of 5D planes defined by the conditions $\langle \sigma_i | x \rangle - 1/2 \in \mathbb{Z}$. These hyperplanes cut E_{\parallel} on 3 periodic families of 2D planes, given by the equations $\langle \sigma_{i\parallel} | x_{\parallel} \rangle - 1/2 \in \mathbb{Z}$, on which the discontinuities of the modulation are localized (provided the 6D nodes are close to E_{\parallel}). We observe that $\sigma_{i\parallel}$ is orthogonal to the lattice plane spanned by a_j and a_k , where $i \neq j \neq k$ so that these planes are parallel to the facets of the primitive rhombohedral cell, with an average distance $d = |\sigma_{i\parallel}|^{-1}$. They built a periodic tiling of E_{\parallel} with domains of linear size $\approx d$ within which the modulation is affine (notice that d is typically incommensurate with the quasiperiodic structure). More precisely, if x and y are nodes which project in the same domain then $\hat{\Sigma}(x)_{\parallel} - \hat{\Sigma}(y)_{\parallel}$ reduces to the term $\hat{S}(x - y)_{\parallel}$ which is linear with respect to $x - y$. Moreover, since the shear parameters x and y are small (see Tab. I below) $\hat{S}(x - y)$ is close to $x - y$. On the contrary, if x and y are separated by a plane normal to σ_i , so that $\text{Round} [\langle \sigma_i | x \rangle] - \text{Round} [\langle \sigma_i | y \rangle] = \pm 1$, the relative position will be of the form $\hat{S}(x - y)_{\parallel} \pm s_{i\parallel}$; such discontinuities $\pm s_{i\parallel}$ can be considered as flip-flops induced by the fractional shear on the nodes close to the planes $\langle \sigma_i | x \rangle - 1/2 \in \mathbb{Z}$.

The example of a modulated octagonal tiling is given in figure 3. We observe that the domains between linear defects contain *different* unit cells of the same approximant structure (these cells are pointed out by the nodes which belong to the square lattice $L' \cap E_1$). The modulated structure is therefore different from a microcrystalline structure where all domains would show a unique approximant unit cell. The occurrence of different approximant tilings for a given approximant orientation is a general consequence of the standard cut algorithm. In the rational case, a global shift of the lattice with respect to the acceptance domains will generally induce finitely many structures with the same Bravais lattice but with different non equivalent unit cells. Usually, none of them will have the full symmetry expected from the approximant orientation: the symmetry of the acceptance domain implies a degenerate situation where nodes fall on the boundaries of the cut (see [10] for a discussion of the octagonal case).

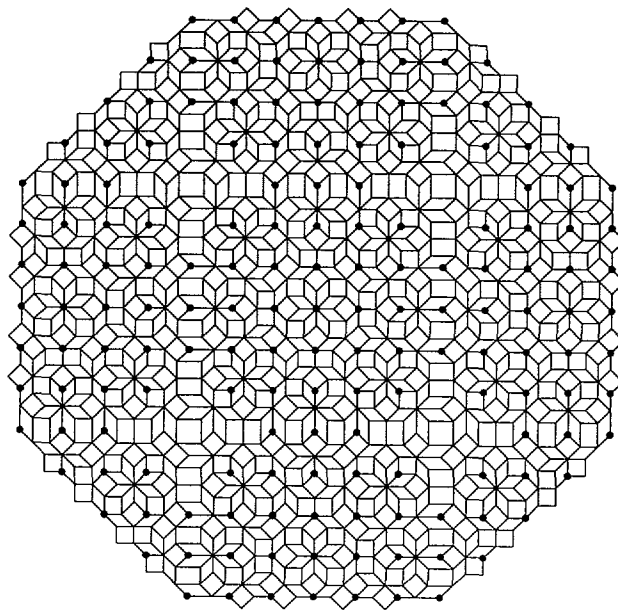


Fig. 3. — In the framework of the octagonal tiling two shears are required to map $L = \mathbb{Z}^4$ onto a lattice L' such that $L' \cap E_1$ is a square lattice. The projection of the L -nodes deformed by the fractional shears gives the above modulated structure. The discontinuities of the transformation gives rise to defect lines. The nodes belonging to $L' \cap E_1$ are indicated by black circles.

Table I gives the simplest solutions corresponding to the shortest shear vectors s in a 2-fold plane; the sequence corresponds to the first approximants of the orthogonal direction $(1, -\tau)$.

4. Fourier spectrum of the modulated quasiperiodic structure.

In this section we compute the spectrum of a standard quasiperiodic structure modified by fractional shears in the high dimensional space as described in the previous section.

Assume we are given p fractional shears $\Sigma_1, \Sigma_2, \dots, \Sigma_p$, in the N -dimensional space, associated to the pairs $(s_1, \sigma_1), (s_2, \sigma_2), \dots, (s_p, \sigma_p)$ where s_1, s_2, \dots, s_p belong to the

Table I. — *Different shear parameters of possible QC to rhombohedral transformations. The shear vectors are of the type (m, m, 0, -n, 0, n) and project in E_{||} with lengths s_{||}. The rhombohedral approximant has a primitive lattice parameter a_R (with the 6D parameter 6.3146 Å). The discontinuities of the fractional shears are localized on 5D planes orthogonal to covectors of the form (y, y, -y, -x, -y, -y); they give rise to planar defects on pseudo 5-fold planes, parallel to primitive facets, with an average distance d in physical space.*

m	n	x	y	a _R	d	s
0	1	$\frac{3\tau - 5}{6}$	$\frac{3\tau - 5}{6}$	31.49 Å	113.49 Å	4.69 Å
1	-1	$\frac{3(\tau - 8/5)}{2}$	$\frac{\tau - 8/5}{2}$	32.44 Å	189.14 Å	2.90 Å
1	-2	2(13/8 - τ)	13/8 - τ	32.08 Å	302.63 Å	1.79 Å
2	-3	$\frac{7(\tau - 21/13)}{2}$	$\frac{3(\tau - 21/13)}{2}$	32.22 Å	491.78 Å	1.11 Å

reference lattice L. Furthermore, we assume that σ_i · s_j = 0 for all i, j so that the transformations are uncoupled and commute (this is possible if p ≤ N/2, and satisfied by the shears defined in the previous section). The action of the product $\hat{\Sigma} = \Sigma_1 \cdot \Sigma_2 \cdot \dots \cdot \Sigma_p$ is simply given by :

$$\hat{\Sigma}(x) = x + \sum_{i=1}^p \phi(\sigma_i \cdot x) s_i \tag{4.1}$$

where φ(σ_i · x) = Frac [⟨σ_i, x⟩].

Let X denote the set of points of L selected by a cut of characteristic function χ in the perpendicular space E_⊥. Then the N-dimensional Fourier transform S₀ of X is given by :

$$S_0(k) = \sum_{x \in X} e^{2i\pi k \cdot x} = \sum_{q \in L^*} \tilde{\chi}(k_{\perp} - q_{\perp}) \delta(k_{\parallel} - q_{\parallel}) \tag{4.2}$$

where q runs over the reciprocal lattice L* of L, $\tilde{\chi}$ denotes the Fourier transform of χ and k = (k_{||}, k_⊥) is the orthogonal decomposition with respect to E_{||} and E_⊥ (see [11] for general references). The Fourier transform of the projected structure X_{||} of X is given by the restriction of S₀ to the physical reciprocal space (k_⊥ = 0) :

$$S_0(k_{\parallel}) = \sum_{q \in L^*} \tilde{\chi}(-q_{\perp}) \delta(k_{\parallel} - q_{\parallel}) . \tag{4.3}$$

The Fourier transform S of the set $\hat{\Sigma}(X)$ is formally given by

$$S(k) = \sum_{x \in X} e^{2i\pi k \cdot \left[x + \sum_{i=1}^p \phi(\sigma_i \cdot x) s_i \right]} = \sum_{x \in X} e^{2i\pi k \cdot x} \mathfrak{F}(x, k) \tag{4.4}$$

with
$$\mathfrak{F}(x, k) = e^{2i\pi k \cdot \left[\sum_{i=1}^p \phi(\sigma_i \cdot x) s_i \right]} \tag{4.5}$$

Now, for any k , the function $\mathfrak{F}(x, k)$ is periodic with respect to the variables (σ_i, x) since ϕ (the Frac function) is periodic with period 1. Therefore we have a Fourier expansion of the form :

$$\mathfrak{F}(x, k) = \sum_{n \in \mathbb{Z}^p} a_n(k) e^{2i\pi\sigma^{(n)} \cdot x} \quad (4.6)$$

where $n = (n_1, n_2, \dots, n_p)$ is in \mathbb{Z}^p and $\sigma^{(n)} = \sum n_i \sigma_i$.

The Fourier transform now writes :

$$\begin{aligned} \mathfrak{S}(k) &= \sum_{n \in \mathbb{Z}^p} a_n(k) \sum_{x \in X} e^{2i\pi(k + \sigma^{(n)}) \cdot x} = \sum_{n \in \mathbb{Z}^p} a_n(k) \mathfrak{S}_0(k + \sigma^{(n)}) \\ &= \sum_{q \in L^*} \sum_{n \in \mathbb{Z}^p} a_n(k) \tilde{\chi}(k_{\perp} + \sigma_{\perp}^{(n)} - q_{\perp}) \delta(k_{\parallel} + \sigma_{\parallel}^{(n)} - q_{\parallel}) \end{aligned} \quad (4.7)$$

where $\sigma_{\parallel}^{(n)} = \sum n_i \sigma_{i\parallel}$ and $\sigma_{\perp}^{(n)} = \sum n_i \sigma_{i\perp}$ are the parallel and perpendicular projections of $\sigma^{(n)}$.

The Fourier transform $\mathfrak{S}(k_{\parallel})$ of the projected structure is again obtained for $k_{\perp} = 0$:

$$\mathfrak{S}(k_{\parallel}) = \sum_{q \in L^*} \sum_{n \in \mathbb{Z}^p} a_n(q_{\parallel} - \sigma_{\parallel}^{(n)}) \tilde{\chi}(\sigma_{\perp}^{(n)} - q_{\perp}) \delta(k_{\parallel} + \sigma_{\parallel}^{(n)} - q_{\parallel}). \quad (4.8)$$

The Dirac peaks are located at points $k_{\parallel} = q_{\parallel} - \sigma_{\parallel}^{(n)}$ where q_{\parallel} belongs to the projection of the reciprocal lattice L^* . The terms $\sigma_{\parallel}^{(n)}$ are responsible for the satellite shifts which are indexed by n in \mathbb{Z}^p . The amplitude of the Fourier transform at k_{\parallel} is simply the product of the structure factor $\tilde{\chi}(\sigma_{\perp}^{(n)} - q_{\perp})$ by the Fourier coefficient $a_n(q_{\parallel} - \sigma_{\parallel}^{(n)})$.

The Fourier coefficients $a_n(k)$ can be easily computed by integration of $\mathfrak{F}(x, k)$ with respect to the variables (σ_i, x) in the p -dimensional unit cube C defined by $-1/2 < (\sigma_i, x) < 1/2$. In this case the Frac functions reduce to $\phi(\sigma_i \cdot x) = \sigma_i \cdot x$ all over C so that :

$$\begin{aligned} a_n(k) &= \int_C e^{-2i\pi\sigma^{(n)} \cdot x} \mathfrak{F}(x, k) \prod_{i=1}^p d(\sigma_i, x) \\ &= \int_C e^{2i\pi \sum_{i=1}^p [(k \cdot s_i) - n_i] (\sigma_i, x)} \prod_{i=1}^p d(\sigma_i, x) \\ &= \prod_{i=1}^p \frac{\sin[\pi(k \cdot s_i - n_i)]}{\pi(k \cdot s_i - n_i)} \end{aligned} \quad (4.9)$$

As explained above the Fourier coefficient a_n must be evaluated at points $k_{\parallel} = q_{\parallel} - \sigma_{\parallel}^{(n)}$, where q belongs to L^* and where $\sigma_{\parallel}^{(n)} = \sum n_i \sigma_{i\parallel}$. Now, we observe that $(q_{\parallel} - \sigma_{\parallel}^{(n)}) \cdot s_i = (q - \sigma^{(n)}) \cdot s_i - (q_{\perp} - \sigma_{\perp}^{(n)}) \cdot s_i$; on the other hand $q \cdot s_i$ is an integer and $\sigma^{(n)} \cdot s_i$ vanishes ($\sigma_j \cdot s_i = 0$ for all i, j), so that we get :

$$|a_n(q_{\parallel} - \sigma_{\parallel}^{(n)})| = \prod_{i=1}^p \left| \frac{\sin[\pi(q_{\perp} - \sigma_{\perp}^{(n)}) \cdot s_{i\perp}]}{\pi((q_{\parallel} - \sigma_{\parallel}^{(n)}) \cdot s_{i\parallel} - n_i)} \right|. \quad (4.10)$$

In the cases of the modulations of icosahedral structures presented in table I, we can check that the terms $\sigma_{\perp}^{(n)} \cdot s_{i\perp}$ are very small (of order 10^{-3} to 10^{-2}). If the structure factor $\tilde{\chi}$ varies slowly at such a scale, the relative amplitudes of the satellites for a given peak

q_{\parallel} will be principally governed by the Fourier coefficient a_n . We also observe that symmetric satellites will usually have different intensities since a_n is not symmetric with respect to the integers n_i . Moreover, high order satellites could exceed the first ones according to the denominator $(q_{\parallel} - \sigma_{\parallel}^{(n)}) \cdot s_{i\parallel} - n_i$ of (4.10) being close to 0. Finally, since noticeable reflections correspond to usually small q_{\perp} , the numerator will show few oscillations so that the behaviour of a_n will be more clearly organized when plotted in perpendicular space (such a feature has been observed for the TEM diffractions of the modulated icosahedral AlFeCu phase [12]).

5. Conclusion.

The modulated icosahedral phase seems to occur as an intermediate state during a structural transformation between an icosahedral phase and a microcrystalline rhombohedral phase. The mechanism proposed in this paper allows to build modulated icosahedral structures by means of displacive transformations. In this respect, the modulated state seems to be much closer to the icosahedral phase than to any microcrystalline phase : the second step of the structural transformation seems to require diffusion processes, even limited to the extent of the domains of the modulated state. Such a transformation is much more dependent on the precise structure of the icosahedral phase since, for instance, the possibility of simple collective displacements requires a strong closeness property of the atomic surfaces [13].

On the other hand, the mechanism presented here can also be used to build modulated crystalline structures starting from perfect approximant ones. In that case the modulated states would be much closer to the crystalline phase than to any icosahedral phase in the sense that the second step of a structural transformation would certainly also require diffusion processes.

Acknowledgments.

The author is grateful to P. Bancel and D. Gratias for fruitful discussions.

References

- [1] AUDIER M., BRÉCHET Y., DE BOISSIEU M. and GUYOT P., *Philos. Mag. B* **63** (1991) 1375-1393.
- [2] MENGUY N. and AUDIER M., Stability and instability in the different phases in the rhombohedral-icosahedral transition of AlFeCu alloys. *Methods of Structural Analysis of Modulated Structures and Quasicrystals*, J. M. Perez-Mato *et al.* Eds. (Lekeitio, Spain ; World Scientific, 1992).
- [3] BANCEL P., private communication.
- [4] DUNEAU M. and OGUEY C., *J. Phys. A* **24** (1991) 461-475.
- [5] DUNEAU M., *J. Phys. I France* **2** (1992) 871-886.
- [6] TSAI A. P., INOUE A. and MASUMOTO T., *J. Mat. Sci. Lett.* **6** (1987) 1403 and *Jpn J. Appl. Phys.* **26** (1987) L1505.
- [7] EBALARD S. and SPAEPEN F., *J. Mater. Res.* **4** (1989) 39.
- [8] a) DEVAUD-RZEPSKI J., QUIVY A., CALVAYRAC Y., CORNIER-QUINQUANDON M. and GRATIAS D., *Philos. Mag. B* **60** (1989) 855. b) CALVAYRAC Y., QUIVY A., BESSIÈRE M., LEFEBVRE S., CORNIER-QUINQUANDON M. and GRATIAS D., *J. Phys. France* **51** (1990) 417-431.
- [9] AUDIER M. and GUYOT P. : a) *Phil. Mag. Lett.* **58** (1989) 17. b) Proceedings of the Third International Conference on Quasicrystals, Mexico, J. Yacaman *et al.* Eds. (Singapore : World Scientific, 1989) p. 288.

- [10] DUNEAU M., *J. Phys. I France* **1** (1991) 1591-1601.
- [11] P. J. Steinhardt and S. Ostlund Eds., *The Physics of Quasicrystals* (Singapore, World Scientific) (1987); Ch. Jannot and J. M. Dubois Eds., *Quasicrystalline Materials* (Singapore, World Scientific) (1988);
HENLEY C. L., *Comm. Cond. Mat. Phys.* **13** (1987) 59 ;
JANNOT Ch. and DUBOIS J. M., *J. Phys. France* **18** (1988) 2303.
- [12] LEVITOV L. C., unpublished work.
- [13] CORNIER-QUINQUANDON M., QUIVY A., LEFEBVRE S., ELKAIM E., HEGER G., KATZ A. and GRATIAS D., *Phys. Rev. B* **44** (1991) 2071-2084.