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Indexing approximants of icosahedral quasicrystals

M. Quiquandon,[a] A. Katz,[b] F. Puyraimond[c] and D. Gratias[c]*

[a]*CECM/CNRS, 15 rue Georges Urbain, F-94407 Vitry CEDEX, France,* [b]*CPT, Ecole Polytechnique, F-91128 Palaiseau, France, and* [c]*LEM-CNRS/ONERA, BP 72, F-92322 Châtillon, France. E-mail: gratias@onera.fr*

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Abstract

It is well known that the crystallography of approximants is directly related to that of the parent quasicrystal, once its unit-cell vectors are identified as parallel projections of certain N-dimensional lattice nodes $\mathbf{A}^i$. Derived here are explicit simple relations for calculating the shear matrices $\boldsymbol{\varepsilon}$ and the related crystallographic properties of the corresponding approximants, including diffraction indexing and the determination of the lattice in perpendicular space. Applied to low-dimensional approximants, the derivation shows that the systematic 'accidental' extinction rules observed in the pentagonal phases are generic extinctions that are due to the geometrical properties of the projected 1D lattice and are independent of the actual model of the quasicrystal.

1. Introduction

Significant effort has been devoted over the last few years to improving our understanding of the geometric connections between approximants and their parent quasicrystals for deciphering the hidden symmetries of complex periodic intermetallic phases using a unique framework merging both crystalline and quasicrystalline concepts (see, for instance, Duneau, 1988; Duneau *et al.*, 1989; Jarić & Qiu, 1990; Janssen, 1991; Ishii, 1989, 1990, 1992; Yamamoto & Ishihara, 1988; Yamamoto, 1990; Duneau & Audier, 1994; Gratias *et al.*, 1995). The present paper is a contribution to this problem, devoted more specifically to the technical question of relating the standard crystallographic indexing to that developed in the N-dimensional scheme used to describe quasicrystal structures. We shall see here that the apparent redundancy of introducing $N > 3$ indices in a framework that basically requires only three is a natural way of keeping track of the geometric properties of the parent quasicrystal in the approximant.

Although the method we shall discuss here is easily transposable to other systems, we restrict our attention to the case of icosahedral symmetry with an embedding of the physical space in a 6D space. The 6D lattice Λ projects in a uniformly dense manner on both physical (parallel), $\mathbf{E}_\parallel$, and perpendicular, $\mathbf{E}_\perp$, 3D spaces.

We generate periodic approximants of a given quasicrystal using the perpendicular shear technique as first proposed by Jarić & Qiu (1990) and subsequently developed by several authors (Janssen, 1991; Ishii, 1989, 1990, 1992; Yamamoto & Ishihara, 1988; Yamamoto, 1990; Duneau, 1988; Duneau *et al.*, 1989; Duneau & Audier, 1994; Gratias *et al.*, 1995). This simple and efficient technique has proved to be very accurate in practice, for example in the crystallographic characterization of the approximant phases of the icosahedral quasicrystal. Such phases have been identified, for example, in the Al–Mn–Si (Elser & Henley, 1985; Guyot & Audier, 1985) and Al–Cu–Fe (Audier & Guyot, 1990*a*,*b*; Calvayrac *et al.*, 1990; Bancel, 1993; Menguy *et al.*, 1993; Quiquandon *et al.*, 1996) ternary systems, and Al–Si–Cu–Fe (Quivy *et al.*, 1996), Al–Si–Cu–Ru (Sugiyama, Kaji *et al.*, 1998*a*,*b*; Sugiyama, Takeshi *et al.*, 1998), Al–Si–Pd–Mn (Hiraga *et al.*, 1997, 1998; Sugiyama, Kaji *et al.*, 1998*a*,*b*; Sugiyama, Takeshi *et al.*, 1998) quaternary systems.

The paper proceeds in three steps.

The first step is to establish the basic relations between the standard 3D indexing (H, K, L) of the reflections of the approximant crystal with the 6D indexing of icosahedral quasicrystals. We propose a simple way of computing these 3D indices starting from the sole knowledge of three 6D lattice vectors, the parallel projections of which define the primitive unit cell of the approximant. This knowledge is also sufficient to compute the unit cells of the other 3D lattices generated in both direct and reciprocal spaces parallel and perpendicular.

The second step consists in examining the 3D lattice generated in perpendicular space (both direct and reciprocal) for explicitly completing the 6D description of the approximant in the hyperspace.

Finally, in the third step, we illustrate the technique in the simple cases of approximants of low dimensionality – typically, the pentagonal phase that is periodic in one direction only – and briefly discuss their peculiar properties of exhibiting diffraction patterns with 'systematic'

extinctions. We will show that these extinctions are due to geometric properties of the projection in the reciprocal space of the sheared (reciprocal) 6D lattice and do not directly depend on the actual locations and specific shapes of the atomic surfaces.

2. The shear method

The perpendicular shear method consists in applying a linear shear along the perpendicular space $\mathbf{E}_\perp$ on the 6D lattice Λ, in order to bring a rational 3D hyperplane of Λ parallel to the physical space $\mathbf{E}_\parallel$ (see Fig. 1). Once this is achieved, the 3D lattice of the periodic approximant is the image in $\mathbf{E}_\parallel$ of the 3D lattice of the chosen rational hyperplane. A detailed description of this technique can be found in Gratias *et al.* (1995) and Quiquandon *et al.* (1996).

For the present purpose, it is enough to mention that, for the case of icosahedral phases, the perpendicular shear $\boldsymbol{\varepsilon}$ is defined by a 3×3 matrix with real entries for 3D periodic approximants. Following Jarić & Qiu (1990), any node λ of the 6D lattice Λ projecting as $\lambda = \{x_\parallel, x_\perp\}$ along $\mathbf{E}_\parallel$ and $\mathbf{E}_\perp$, respectively, projects, after shear, according to

$$\begin{aligned} x'_\parallel &= x_\parallel \\ x'_\perp &= x_\perp - \boldsymbol{\varepsilon} x_\parallel, \end{aligned} \tag{1}$$

which, in reciprocal space,† translates into

$$\begin{aligned} q'_\parallel &= q_\parallel + \tilde{\boldsymbol{\varepsilon}} q_\perp \\ q'_\perp &= q_\perp, \end{aligned} \tag{2}$$

where $\tilde{\boldsymbol{\varepsilon}}$ designates the transposed matrix of $\boldsymbol{\varepsilon}$ and $\{q_\parallel, q_\perp\}$ are the coordinates of a wavevector of the initial reciprocal 6D lattice Λ^* and $\{q'_\parallel, q'_\perp\}$ those of the transformed wavevector after the action of the shear.

Parallel and perpendicular components of a 6D vector of Λ are described here in the notation of Cahn *et al.* (1986), hereafter called CSG notation. They are obtained by applying to Λ the 6×3 matrices $\boldsymbol{M}_\parallel$ for the parallel components and $\boldsymbol{M}_\perp$ for the perpendicular ones, defined by

$$\boldsymbol{M}_\parallel = \alpha \boldsymbol{m}_\parallel = \alpha \begin{pmatrix} 1 & \tau & 0 & -1 & \tau & 0 \\ \tau & 0 & 1 & \tau & 0 & -1 \\ 0 & 1 & \tau & 0 & -1 & \tau \end{pmatrix}$$

and

$$\boldsymbol{M}_\perp = \alpha \boldsymbol{m}_\perp = \alpha \begin{pmatrix} -\tau & 1 & 0 & \tau & 1 & 0 \\ 1 & 0 & -\tau & 1 & 0 & \tau \\ 0 & -\tau & 1 & 0 & \tau & 1 \end{pmatrix},$$

where $\tau = (5^{1/2} + 1)/2$ is the golden mean and $\alpha = 1/[2(2 + \tau)]^{1/2}$.

We take the 6D lattice parameter of Λ as the unit length. Hence, any vector λ of Λ has components on the CSG orthonormal bases of $\mathbf{E}_\parallel$ and $\mathbf{E}_\perp$ of the form $\alpha(n + n'\tau)$ in parallel space and $\alpha(n' - n\tau)$ in perpendicular space with n and n' integers. We designate by *reduced coordinates* these coordinates relieved from the scaling factor α.

Some of the properties of the CSG matrices which will be useful in the paper are the following:

$$\boldsymbol{M}_\parallel \tilde{\boldsymbol{M}}_\parallel = \mathbf{1}_{\mathbf{E}_\parallel}, \quad \boldsymbol{M}_\perp \tilde{\boldsymbol{M}}_\perp = \mathbf{1}_{\mathbf{E}_\perp},$$
$$\boldsymbol{M}_\parallel \tilde{\boldsymbol{M}}_\perp = \boldsymbol{M}_\perp \tilde{\boldsymbol{M}}_\parallel = 0,$$

where $\tilde{\boldsymbol{M}}_\mathbf{E}$ designates the transposed matrix of $\boldsymbol{M}_\mathbf{E}$ and $\mathbf{1}_\mathbf{E}$ designates the identity matrix on space $\mathbf{E}$. They are connected to the usual projectors $\boldsymbol{\pi}_\parallel$ and $\boldsymbol{\pi}_\perp$ in parallel and perpendicular spaces by

$$\tilde{\boldsymbol{M}}_\parallel \boldsymbol{M}_\parallel = \boldsymbol{\pi}_\parallel, \quad \tilde{\boldsymbol{M}}_\perp \boldsymbol{M}_\perp = \boldsymbol{\pi}_\perp$$

so that

$$\tilde{\boldsymbol{M}}_\parallel \boldsymbol{M}_\parallel + \tilde{\boldsymbol{M}}_\perp \boldsymbol{M}_\perp = \mathbf{1}_{6\mathrm{D}}.$$

We generate a 3D periodic approximant to the icosahedral phase by choosing three vectors $\mathbf{A}^1$, $\mathbf{A}^2$ and $\mathbf{A}^3$ of the hyperlattice Λ, the parallel projections of which are linearly independent in $\mathbf{E}_\parallel$, which define the *primitive*

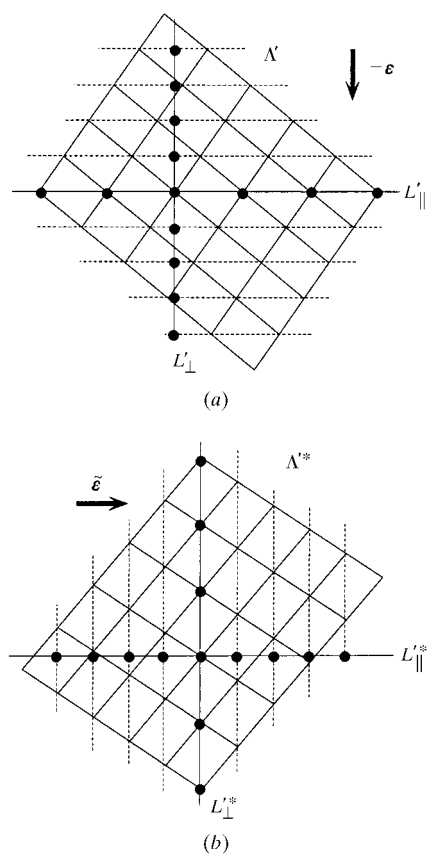

Fig. 1. Geometry of the perpendicular shear technique (*a*) in direct space; (*b*) in reciprocal space.

† We write reciprocal vectors in columns to conform with practical use.

unit cell of the (periodic) approximant. Aligning the 3D plane spanned by these three vectors parallel to $\mathbf{E}_\parallel$ consists in computing the shear matrix $\boldsymbol{\varepsilon}$ such that the perpendicular components of the transform of the vectors $\mathbf{A}^i$ vanish.

For that purpose, we build the two 3×3 matrices $\boldsymbol{A}_\parallel$ and $\boldsymbol{A}_\perp$ defined by

$$\boldsymbol{A}_\parallel = \alpha \boldsymbol{a}_\parallel = \alpha \begin{pmatrix} a^1_{\parallel,x} & a^2_{\parallel,x} & a^3_{\parallel,x} \\ a^1_{\parallel,y} & a^2_{\parallel,y} & a^3_{\parallel,y} \\ a^1_{\parallel,z} & a^2_{\parallel,z} & a^3_{\parallel,z} \end{pmatrix}$$

$$\boldsymbol{A}_\perp = \alpha \boldsymbol{a}_\perp = \alpha \begin{pmatrix} a^1_{\perp,x} & a^2_{\perp,x} & a^3_{\perp,x} \\ a^1_{\perp,y} & a^2_{\perp,y} & a^3_{\perp,y} \\ a^1_{\perp,z} & a^2_{\perp,z} & a^3_{\perp,z} \end{pmatrix},$$

where the matrix elements – written in lower case – are the reduced coordinates of the projection of the vectors $\mathbf{A}^i$ onto, respectively, the parallel and perpendicular spaces. We can proceed backwards and compute the vectors $\mathbf{A}^i$ as a 3×6 matrix, say $\boldsymbol{\mathcal{A}}$, with integer entries, the columns of which are respectively $\mathbf{A}^1$, $\mathbf{A}^2$ and $\mathbf{A}^3$ starting from $\boldsymbol{a}_\parallel$ and $\boldsymbol{a}_\perp$ through the relation

$$\boldsymbol{\mathcal{A}} = \alpha^2 \big(\widetilde{\boldsymbol{m}}_\parallel \boldsymbol{a}_\parallel + \widetilde{\boldsymbol{m}}_\perp \boldsymbol{a}_\perp \big).$$

With these notations, the searched shear matrix $\boldsymbol{\varepsilon}$ is given by

$$\boldsymbol{a}_\perp - \boldsymbol{\varepsilon} \boldsymbol{a}_\parallel = \mathbf{0},$$

which leads to

$$\boldsymbol{\varepsilon} = \boldsymbol{a}_\perp \boldsymbol{a}_\parallel^{-1}. \tag{3}$$

The procedure applies in the same way for lower-dimensional, 2D or 1D, periodic approximants by choosing, respectively, two or one vector(s) $\mathbf{A}$ and solving (3) with, respectively, 2×2 matrices or simple numbers, as will be shown in the third part of this paper.

We observe that the shear matrices (3) associated with periodic approximants have entries of the form $(n + m\tau)/d$, where n, m and d are integers (and $d > 0$). In other words, the matrix is built on the field of fractions $\mathbb{Q}(\tau)$ of the standard $\mathbb{Z}$-module $\mathbb{Z}(\tau)$. This property makes exact computations possible. Indeed, each entry being a triplet of integers (n, m, d), we can code the arithmetic operations on these elements as combinations of additions and multiplications of integers: if (n, m, d) and (n', m', d') are two elements of $\mathbb{Q}(\tau)$, then

$$\begin{aligned} (n, m, d) \pm (n', m', d') &= (d'n \pm dn', d'm \pm dm', dd') \\ (n, m, d) * (n', m', d') &= (nn' + mm', nm' + n'm + mm', dd') \\ (n, m, d)/(n', m', d') &= (d'(nn' + nm' - mm'), d'(mn' - m'n), \\ &\qquad d(n'^2 + n'm') - m'^2) \\ (n, m, d)^{-1} &= (dn + dm, -dm, n^2 + nm - m^2), \end{aligned}$$

so there is no need to introduce in the computation a floating-point approximation of τ.

Expression (3) is very convenient for computing the shear matrices associated with a series of 'convergent' approximants within a given symmetry class. Let $\boldsymbol{\varepsilon}_0$ be the shear matrix associated with a certain starting set of $\mathbf{A}$'s. We can apply to that set the 'quasidilatation' $\boldsymbol{\tau}$ matrix defined by

$$\boldsymbol{\tau} = \tau \mathbf{1}_{\mathbf{E}_\parallel} + (1 - \tau) \mathbf{1}_{\mathbf{E}_\perp}.$$

This quasidilatation inflates the parallel components of the initial vectors $\mathbf{A}^i$ by the factor τ and deflates the perpendicular components by $1 - \tau$. Because of (3), the shear matrix is therefore transformed into

$$\boldsymbol{\varepsilon}^{(1)} = -\tau^{-2} \boldsymbol{\varepsilon}_0.$$

After n applications of this transformation, we obtain

$$\boldsymbol{\varepsilon}^{(n)} = (-1)^n \tau^{-2n} \boldsymbol{\varepsilon}_0.$$

More generally, if we apply to a set of initial vectors $\mathbf{A}^i$ a transformation of the form $p \times \mathbf{1} + q \times \boldsymbol{\tau}$, where p and q are integers, we obtain $\boldsymbol{\varepsilon}$ with the form

$$\boldsymbol{\varepsilon}^{(p,q)} = \frac{p + q(1 - \tau)}{p + q\tau} \boldsymbol{\varepsilon}_0,$$

since the parallel components are multiplied by $p + q\tau$ and the perpendicular components by $p + q(1 - \tau)$.

In most practical cases, approximants of high symmetry of the icosahedral phases can be defined by their 'order' p/q with coprime integers p and q that

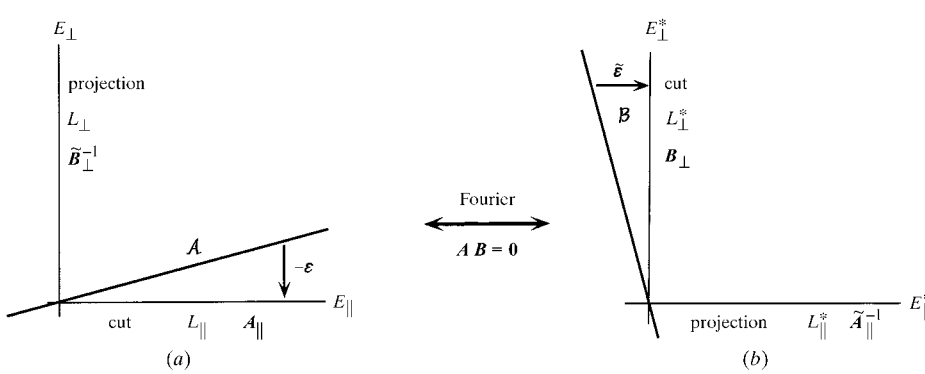

Fig. 2. 3D lattices induced by the shear ε: (*a*) in direct space; (*b*) in reciprocal space.

entirely parametrize the vectors $\mathbf{A}^i$. We make the following choice:

(i) for pentagonal symmetry (1D periodic), $A = (2p - q, q, q, q, q, \bar{q})$;

(ii) for cubic symmetry, $\mathbf{A}^1 = (q, p, 0, \bar{q}, p, 0)$, $\mathbf{A}^2 = (0, q, p, 0, \bar{q}, p)$ and $\mathbf{A}^3 = (p, 0, q, p, 0, \bar{q})$;

(iii) for rhombohedral symmetry, $\mathbf{A}^1 = (p, p, q, 0, q, 0)$, $\mathbf{A}^2 = (q, p, p, 0, 0, q)$ and $\mathbf{A}^3 = (p, q, p, q, 0, 0)$;

This choice† is closest to approximate algebraic numbers by rational fractions: here, the approximant order p/q is the fraction we use to approximate τ in the whole geometric framework. For example, the trace of $\mathbf{E}_\parallel$ in the fivefold 2D plane spanned by the 6D vectors $(1, 0, 0, 0, 0, 0)$ and $(0, 1, 1, 1, 1, \bar{1})$ is a line oriented along the vector $(2\tau - 1, 1, 1, 1, 1, \bar{1})$. Hence, approximating τ by p/q leads to the direction $(2p - q, q, q, q, q, \bar{q})$.

3. Lattices in parallel space

We now turn to the specific geometric aspects of the transformed 6D lattice Λ' for perpendicular shears generating 3D periodic approximants. This 6D lattice has the following properties (see Fig. 2):

(i) its intersection by a 3D space parallel to $\mathbf{E}_\parallel$ going through a node is a 3D lattice $L_\parallel$; the components of its unit-cell vectors are given by the columns of the matrix $\boldsymbol{A}_\parallel$;

(ii) in reciprocal space, the projection of Λ'^* onto $\mathbf{E}_\parallel^*$ is a 3D lattice $L_\parallel^*$ that is the reciprocal lattice of $L_\parallel$; the components of its unit-cell vectors are given by the columns of the matrix $\widetilde{\boldsymbol{A}}_\parallel^{-1}$.

These 3D lattices are periodic *subsets* of the original $\mathbb{Z}$-modules defined by the uniformly dense projections of Λ and Λ^* in $\mathbf{E}_\parallel$ and $\mathbf{E}_\parallel^*$. This is a general property of all 3D periodic approximants obtained by a perpendicular shear of the 6D lattice Λ. In that scheme, the Wyckoff positions W_j of the jth atom of the structure of an approximant deriving from a quasicrystal is given by

$$W_j = \boldsymbol{a}_\parallel^{-1} \boldsymbol{m}_\parallel \lambda_j,$$

where λ_j is the initial location of the corresponding atomic surfaces in the 6D space. If this location is a rational vector of Λ in the quasicrystal, the coordinates of the corresponding Wyckoff position, in the approximant, take the form $(n + m\tau)/d$ with n, m, d integers: the atoms of the approximant are at positions which are a subset of the original $\mathbb{Z}$-module of the atomic positions of the quasicrystal.‡ This property can be used backwards to identify a possible hyperspace embedding of a complex intermetallic phase: if all the atomic positions of the phase are located on nodes of a $\mathbb{Z}$-module, then it is easy to construct a hyperspace of dimension equal to the rank of the $\mathbb{Z}$-module in which the structure appears as a rational cut of a hypothetical quasicrystal.

It is now straightforward to find the 3D indices H, K, L of the reciprocal lattice $L_\parallel^*$ of the 6D reflection $q' = (q'_\parallel, q'_\perp)$ of a given periodic approximant, imaged after shear of a 6D reflection q of the parent quasicrystal. These indices are defined by

$$q'_\parallel = \widetilde{\boldsymbol{A}}_\parallel^{-1} \begin{pmatrix} H \\ K \\ L \end{pmatrix}.$$

Using relations (2) and (3), we obtain

$$\begin{aligned} q'_\parallel &= \alpha(\boldsymbol{m}_\parallel + \widetilde{\boldsymbol{a}}_\parallel^{-1} \widetilde{\boldsymbol{a}}_\perp \boldsymbol{m}_\perp) q \\ &= \widetilde{\boldsymbol{A}}_\parallel^{-1} \alpha^2 (\widetilde{\boldsymbol{a}}_\parallel \boldsymbol{m}_\parallel + \widetilde{\boldsymbol{a}}_\perp \boldsymbol{m}_\perp) q \\ &= \widetilde{\boldsymbol{A}}_\parallel^{-1} (\boldsymbol{A}_\parallel q_\parallel + \boldsymbol{A}_\perp q_\perp) \end{aligned}$$

and therefore

$$\begin{pmatrix} H \\ K \\ L \end{pmatrix} = \alpha^2 \left(\widetilde{\boldsymbol{a}}_\parallel \begin{pmatrix} h + h'\tau \\ k + k'\tau \\ l + l'\tau \end{pmatrix} + \widetilde{\boldsymbol{a}}_\perp \begin{pmatrix} h' - h\tau \\ k' - k\tau \\ l' - l\tau \end{pmatrix} \right). \quad (4)$$

It is easily checked that, despite the presence of α^2, this expression leads to integer values for H, K and L. Let us apply this expression to the unit vectors of Λ, for example the node $(1, 0, 0, 0, 0, 0)$, which, in CSG notation, is written as $(1/0, 0/1, 0/0)$. Let us designate by $(u/u', v/v', w/w')$ the parallel components of $\mathbf{A}^1$. The calculation of H leads to:

$$\begin{aligned} h &= \alpha^2 [u + u'\tau + (v + v'\tau)\tau - \tau(u' - u\tau) + (v' - v\tau)] \\ &= [(\tau + 2)(u + v')]/2(\tau + 2) \\ &= n, \end{aligned}$$

since $u + v'$ is an even integer for a primitive lattice. The same property holds for the other components of the vector A^1 and for the other basis vectors of Λ; it holds therefore for any vector of Λ.

Explicit expressions of the indices (H, K, L) are listed in Table 1 for the main crystallographic systems derived from icosahedral symmetry as a function of the approximant order p/q. As an example, in the case of the rhombohedral approximant of order p/q, using the unit-cell vectors $\mathbf{A}^i$ listed in Table 1, we obtain the following matrices $\boldsymbol{a}$:

$$\boldsymbol{a}_\parallel = (q + p\tau) \begin{pmatrix} \tau & 1 & \tau - 1 \\ 1 & \tau - 1 & \tau \\ \tau - 1 & \tau & 1 \end{pmatrix}$$

and

† We extract these choices from Gratias & Cahn (1986) for the cubic (see also Yamamoto, 1992) and Calvayrac *et al.* (1990) for the rhombohedral systems.

‡ This, of course, neglects the position relaxations that are expected when passing from quasicrystal to crystal.

Table 1. *Indexing for the crystallographic systems corresponding to the approximant phases identified in the alloy Al–Cu–Fe and characterized by the ratio of integers p/q*

Symmetry	p/q ratio	Unit cell	Indexing
Pentagonal			
$\bar{5}m$	p/q	$\mathbf{A} = (2p-q, q, q, q, q, \bar{q})$	From a 6D node $(r, s, s, s, s, \bar{s}) \in$ fivefold axis $L = (2p-q)r + 5qs$
Cubic			
$m\bar{3}$	p/q	$\mathbf{A}^1 = (q, p, 0, -q, p, 0)$ $\mathbf{A}^2 = (0, q, p, 0, -q, p)$ $\mathbf{A}^3 = (p, 0, q, p, 0, -q)$	$H = q(n1-n4) + p(n2+n5)$ $= qh + ph'$ $K = q(n2-n5) + p(n3+n6)$ $= qk + pk'$ $L = q(n3-n6) + p(n1+n4)$ $= ql + pl'$
Orthorhombic			
mmm	p_α/q_α p_β/q_β p_γ/q_γ	$\mathbf{A}^1 = (q_\alpha, p_\alpha, 0, -q_\alpha, p_\alpha, 0)$ $\mathbf{A}^2 = (0, q_\beta, p_\beta, 0, -q_\beta, p_\beta)$ $\mathbf{A}^3 = (p_\gamma, 0, q_\gamma, p_\gamma, 0, -q_\gamma)$	$H = q_\alpha(n1-n4) + p_\alpha(n2+n5)$ $= q_\alpha h + p_\alpha h'$ $K = q_\beta(n2-n5) + p_\beta(n3+n6)$ $= q_\beta k + p_\beta k'$ $L = q_\gamma(n3-n6) + p_\gamma(n1+n4)$ $= q_\gamma l + p_\gamma l'$
Trigonal			
$\bar{3}m$	p/q	$\mathbf{A}^1 = (p, p, q, 0, q, 0)$ $\mathbf{A}^2 = (q, p, p, 0, 0, q)$ $\mathbf{A}^3 = (p, q, p, q, 0, 0)$	$H = q(n3+n5) + p(n1+n2)$ $= (1/2)[q(h'+k-l+l') + p(h+h'+k'+l)]$ $K = q(n1+n6) + p(n2+n3)$ $= (1/2)[q(h-k+k'+l') + p(h'+k+l+l')]$ $L = q(n2+n4) + p(n1+n3)$ $= (1/2)[q(h'-h+k'+l) + p(h+k+k'+l')]$

$$\boldsymbol{a}_\perp = (p - q\tau)\begin{pmatrix} 1-\tau & 1 & -\tau \\ 1 & -\tau & 1-\tau \\ -\tau & 1-\tau & 1 \end{pmatrix},$$

which, by applying equation (4), lead, after a few algebraic manipulations, to the indexing presented in Table 1 (trigonal case).

4. Lattices in perpendicular space

The initial uniformly dense projection of Λ onto $\mathbf{E}_\perp$ collapses, after shear (3), into a discrete set of points that defines a lattice $L_\perp$. Following the same procedure as in the preceding section, we compute the shear matrix $\boldsymbol{\varepsilon}$ in defining three vectors of the *reciprocal* 6D space, say $\mathbf{B}^i$, the parallel projection of which vanishes after shear (see Fig. 2). Introducing the 3×3 matrices $\boldsymbol{B}_\parallel$ and $\boldsymbol{B}_\perp$, the columns of which are the components on the CSG basis of respectively the parallel and perpendicular projections of the $\mathbf{B}^i$ in reciprocal space:

$$\boldsymbol{B}_\parallel = \alpha \boldsymbol{b}_\parallel = \alpha \begin{pmatrix} b^1_{\parallel,x} & b^2_{\parallel,x} & b^3_{\parallel,x} \\ b^1_{\parallel,y} & b^2_{\parallel,y} & b^3_{\parallel,y} \\ b^1_{\parallel,z} & b^2_{\parallel,z} & b^3_{\parallel,z} \end{pmatrix}$$

$$\boldsymbol{B}_\perp = \alpha \boldsymbol{b}_\perp = \alpha \begin{pmatrix} b^1_{\perp,x} & b^2_{\perp,x} & b^3_{\perp,x} \\ b^1_{\perp,y} & b^2_{\perp,y} & b^3_{\perp,y} \\ b^1_{\perp,z} & b^2_{\perp,z} & b^3_{\perp,z} \end{pmatrix},$$

we obtain through (2)

$$\boldsymbol{b}_\parallel + \tilde{\boldsymbol{\varepsilon}} \boldsymbol{b}_\perp = \mathbf{0},$$

i.e.

$$\boldsymbol{\varepsilon} = -\tilde{\boldsymbol{b}}_\perp^{-1} \tilde{\boldsymbol{b}}_\parallel. \tag{5}$$

Equating the two expressions (3) and (5) for $\boldsymbol{\varepsilon}$, we obtain

$$\tilde{\boldsymbol{a}}_\parallel \boldsymbol{b}_\parallel + \tilde{\boldsymbol{a}}_\perp \boldsymbol{b}_\perp = \mathbf{0},$$

which can be written as

$$\tilde{\boldsymbol{\mathcal{A}}} \boldsymbol{\mathcal{B}} = \mathbf{0},$$

where $\boldsymbol{\mathcal{A}}$ and $\boldsymbol{\mathcal{B}}$ are 3×6 matrices, the columns of which are the vectors $\mathbf{A}^i$ and $\mathbf{B}^i$, respectively.

Thus, the vectors $\mathbf{B}^i$ are vectors of the *reciprocal* 3D sublattice orthogonal to the *direct* 3D sublattice generated by the vectors $\mathbf{A}^i$: identifying the vectors $\mathbf{B}^i$ simply consists in constructing the elementary basic vectors of the reciprocal 3D sublattice orthogonal to $\boldsymbol{\mathcal{A}}$. Examples of such $\mathbf{B}^i$ vectors are:†

(i) for cubic symmetry, $\mathbf{B}^1 = (\bar{p}, q, 0, p, q, 0)$, $\mathbf{B}^2 = (q, 0, \bar{p}, q, 0, p)$ and $\mathbf{B}^3 = (0, \bar{p}, q, 0, p, q)$;

† The present vectors are of course not primitive for all values of p and q, in which case the reduced basis is obtained by simple linear combinations.

(ii) for rhombohedral symmetry, $\mathbf{B}^1 = (\bar{q}, 0, 0, p, p, q)$, $\mathbf{B}^2 = (0, \bar{q}, 0, q, p, p)$ and $\mathbf{B}^3 = (0, 0, \bar{q}, p, q, p)$.

We obtain therefore the following properties:

(i) in reciprocal space, the intersection of Λ'^* with a 3D space parallel to $\mathbf{E}_\perp^*$ passing through a reciprocal node is a 3D lattice $L_\perp^*$ whose unit-cell vectors have components given by the columns of the matrix $\boldsymbol{B}_\perp$;

(ii) in direct space, the projection of Λ' onto $\mathbf{E}_\perp$ is a 3D lattice $L_\perp$ (the reciprocal lattice of which is $L_\perp^*$); the components of its unit-cell vectors are given by the columns of the matrix $\widetilde{\boldsymbol{B}}_\perp^{-1}$.

It is straightforward to find the labelling of any node of Λ projected onto $\mathbf{E}_\perp$ after shear as we did for indexing the reciprocal lattice of the approximant. Let (U, V, W) be the indices in $\mathbf{E}_\perp$ of a node $\lambda' = (x'_\parallel, x'_\perp)$, imaged after shear of the 6D node λ. Its indices are defined by

$$x'_\perp = \widetilde{\boldsymbol{B}}_\perp^{-1} \begin{pmatrix} U \\ V \\ W \end{pmatrix}.$$

Using relations (1) and (3), we obtain

$$\begin{aligned} x'_\perp &= \alpha(\boldsymbol{m}_\perp + \widetilde{\boldsymbol{b}}_\perp^{-1}\widetilde{\boldsymbol{b}}_\parallel \boldsymbol{m}_\parallel)\lambda \\ &= \widetilde{\boldsymbol{B}}_\perp^{-1}\alpha^2(\widetilde{\boldsymbol{b}}_\perp \boldsymbol{m}_\perp + \widetilde{\boldsymbol{b}}_\parallel \boldsymbol{m}_\parallel)\lambda \\ &= \widetilde{\boldsymbol{B}}_\perp^{-1}(\widetilde{\boldsymbol{b}}_\perp x_\perp + \widetilde{\boldsymbol{b}}_\parallel x_\parallel) \end{aligned}$$

and therefore

$$\begin{pmatrix} U \\ V \\ W \end{pmatrix} = \alpha^2 \left(\widetilde{\boldsymbol{b}}_\perp \begin{pmatrix} u' - u\tau \\ v' - v\tau \\ w' - w\tau \end{pmatrix} + \widetilde{\boldsymbol{b}}_\parallel \begin{pmatrix} u + u'\tau \\ v + v'\tau \\ w + w'\tau \end{pmatrix} \right). \quad (6)$$

As in the preceding case, it is easily checked that the indices (U, V, W) are indeed integers.

Let us, for example, calculate the unit cell of the 3D lattice $L_\perp$ for the simple case of the cubic approximant of order p/q. Using the vectors $\mathbf{B}^i$ given above, we obtain after simple calculations

$$\widetilde{\boldsymbol{b}}_\parallel = 2(q\tau - p)\mathbf{1}; \quad \widetilde{\boldsymbol{b}}_\perp = 2(q + p\tau)\mathbf{1}$$

and

$$\widetilde{\boldsymbol{B}}_\perp^{-1} = \alpha \frac{2 + \tau}{q + p\tau}\mathbf{1}.$$

This lattice $L_\perp$ is therefore a 3D cubic lattice with unit vectors aligned along three mutually perpendicular twofold axes of length $\alpha(2 + \tau)/(q + p\tau)$. For example, the cubic 1/1 approximant generates a 3D cubic lattice in $\mathbf{E}_\perp$ with lattice parameter $\alpha(2 + \tau)/(1 + \tau) = \alpha(3 - \tau)$. A generic node of Λ, $\lambda = (u/u', v/v', w/w')$ in CSG notation, projects after shear on a node of $L_\perp$ with indices $U = qu' - pu$, $V = qv' - pv$, $W = qw' - pw$ obtained by equation (6). As expected, when p/q approaches τ, the reduced coordinates of the perpendicular projections tend towards those of the icosahedral case. For example, considering the x coordinate, we find from expression (6)

$$x_\perp = \frac{2 + \tau}{q + p\tau}(qu' - pu) = \frac{2 + \tau}{1 + (p/q)\tau}\left(u' - \frac{p}{q}u\right),$$

which, indeed, tends towards $u' - u\tau$ when p/q tends towards τ.

5. Low-dimensional periodic approximants

The present technique is particularly simple to use in the case of low-dimensional periodic approximants, especially for 1D periodic approximants for which all former matrices reduce to numbers.

To exemplify this point, let us consider the case of the pentagonal phases as observed in the Al–Cu–Fe ternary system by Bancel (1993), Menguy *et al.* (1993), Quiquandon *et al.* (1996) and Duneau & Audier (1998). Here, the shear applies in a fivefold 2D plane spanned by the vectors $(1, 0, 0, 0, 0, 0)$ and $(0, 1, 1, 1, 1, 1)$. The traces of $\mathbf{E}_\parallel$ and $\mathbf{E}_\perp$ are lines defined by the vectors

$$\begin{aligned} \mathbf{e}_\parallel &= (2\tau - 1, 1, 1, 1, 1, \bar{1}) \\ \mathbf{e}_\perp &= (2\tau - 1, \bar{1}, \bar{1}, \bar{1}, \bar{1}, 1). \end{aligned} \quad (7)$$

Once normalized, these vectors define the 6×1 projection matrices $\boldsymbol{M}_\parallel$ and $\boldsymbol{M}_\perp$:

$$\begin{aligned} \boldsymbol{M}_\parallel &= \alpha_5(2\tau - 1, 1, 1, 1, 1, \bar{1}) \\ \boldsymbol{M}_\perp &= \alpha_5(2\tau - 1, \bar{1}, \bar{1}, \bar{1}, \bar{1}, 1), \end{aligned}$$

where, here, $\alpha_5 = 1/[(2\tau - 1)2^{1/2}]$.

The pentagonal approximant of order p/q is obtained by aligning along $\mathbf{E}_\parallel$ the 6D vector $\mathbf{A} = (2p - q, q, q, q, q, \bar{q})$, which projects along $\mathbf{e}_\parallel$ and $\mathbf{e}_\perp$ according to $\boldsymbol{A}_\parallel = \boldsymbol{M}_\parallel \boldsymbol{A}$ and $\boldsymbol{A}_\perp = \boldsymbol{M}_\perp \boldsymbol{A}$; they reduce to the numbers

$$\begin{aligned} \boldsymbol{A}_\parallel &= \alpha_5[(2\tau - 1)(2p - q) + 5q] = 2^{1/2}(p - q + q\tau) \\ \boldsymbol{A}_\perp &= \alpha_5[(2\tau - 1)(2p - q) - 5q] = 2^{1/2}(p - q\tau), \end{aligned}$$

leading to:

$$\boldsymbol{\varepsilon}^{(p,q)} = \frac{p - q\tau}{p - q + q\tau}$$

and a 1D lattice parameter of

$$A_5 = A_\parallel = 2^{1/2}(p - q + q\tau).$$

A reflection in the fivefold 2D plane $Q = (r, s, s, s, s, \bar{s})$ projects according to

$$\begin{aligned} q_\parallel &= \alpha_5[(2\tau - 1)r + 5s] \\ q_\perp &= \alpha_5[(2\tau - 1)r - 5s], \end{aligned}$$

thus leading to the L index:

$$L = 2^{1/2}\alpha_5\{(p - q + q\tau)[(2\tau - 1)r + 5s] + (p - q\tau)[(2\tau - 1)r - 5s]\} = (2p - q)r + 5qs.$$

The coordinates r and s being integer numbers, the index L varies by integer steps of the greatest common divisor of $2p - q$ and $5q$. Hence, if $2p - q = 5k$ and k and q are coprime, a 'systematic' extinction occurs where only $L = 5n$ reflections are present.

Such extinctions have been observed in $P1$ ($p/q = 4/3$, with a period equal to 52.31 Å) and $P2$ ($p/q = 7/4$, with a period equal to 84.49 Å) pentagonal phases of the (Al–Cu–Fe) system: all reflections in a row parallel to the periodic fivefold axis are indexed in multiples of five.

These results agree with those recently published by Duneau & Audier (1998), who developed a sophisticated mathematical description of the specific crystallography of those two phases. Besides its simplicity, our present derivation shows that these properties are a direct consequence of the geometry of the projected lattices and are independent on the exact location and shape of the atomic surfaces of the parent quasicrystal.

They have a great influence on the HREM images because the major interference effects in the image formation are due to the fifth harmonic of the basic frequency along the fivefold direction and contribute to form a pseudoperiod five times smaller than the actual one. This is indeed what is observed on high-resolution images where the major fringes along the periodic axis show an apparent periodicity of $A_5/5$ [see, for instance, the images presented in Menguy *et al.* (1993)].

Another way of explaining this pseudoperiod consists in projecting the entire 6D lattice onto the fivefold 2D plane as shown in Figs. 3 and 4. The projected nodes of Λ define a 2D lattice (see Figs. 3*b* and 4*b*) with a unit vector along the $(0, 1, 1, 1, 1, \bar{1})$ direction five times shorter than the one of the cut lattice and the traces of the cuts for both approximants are rational directions with a period five times smaller than the initial one. This shows that any row in Fourier space along the fivefold direction (which is equivalent to calculating the Fourier transform of the projected structure along the fivefold axis) must exhibit a pseudoperiod five times shorter than the real one; this leads to an $L = 5n$ systematic extinction rule.

These drawings are also convenient for qualitatively explaining what the fringes profile obtained on HREM images taken with a systematic row along the fivefold axis should look like.

In the pentagonal phase $P1$, we observe that the pseudoperiod can be decomposed in the stacking of four different layers of alternately strong (corresponding to the large atomic surfaces located at the nodes) and weak scatterers (corresponding to the small atomic surfaces located at the body centers). We expect the image to exhibit two strong fringes per pseudoperiod with perhaps weak secondary fringes in between.

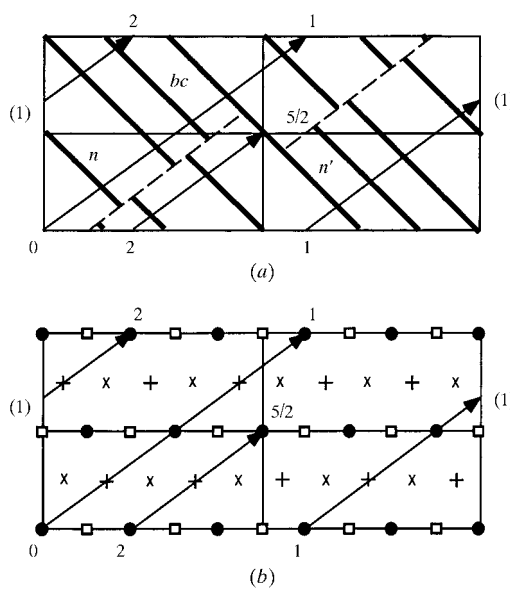

Fig. 3. Representation of the pentagonal phase $P1$ ($p/q = 4/3$) onto the fivefold plane: (*a*) the thick lines represent the traces of the atomic surfaces of the structural model as proposed by Katz & Gratias (1995); the arrows represent the trace of the cut space passing through the origin; (*b*) the complete structure projected onto the fivefold plane: the symbol • corresponds to the projection of the n sites (center of a large triacontahedron), □ to the projection of the n' sites (center of a large truncated triacontahedron), + to the projection of the bc sites (center of a small triacontahedron) and × to the projection of bc' sites (empty).

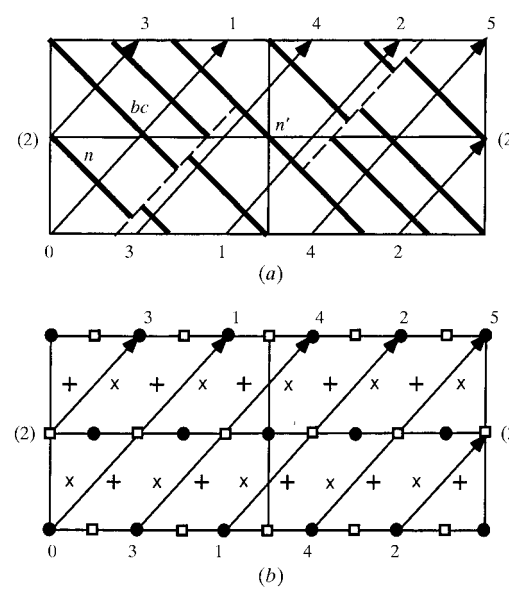

Fig. 4. Representation of the pentagonal phase $P2$ ($p/q = 7/4$) onto the fivefold plane: (*a*) the cut of the pentagonal structure; (*b*) the projected structure with the same notations as Fig. 3.

In the pentagonal phase $P2$, the situation is more complicated: the pseudoperiod decomposes into ten different layers in the sequence of four consecutive weakly scattering, one strong, then four weak again and finally one strong. The fringe profile in one pseudoperiod should show clearly two equispaced strong fringes separated by four weak ones which may well not be resolved.

These qualitative features, specific to the pentagonal phases in the Al–Cu–Fe system, are indeed observed on the HREM images (Menguy *et al.*, 1993).

In the same spirit, we can calculate what extinction conditions should fulfil 1D periodic approximants† built from shears along a twofold and a threefold axis.

The starting vectors in those cases are, respectively,

$$\mathbf{A}_2 = (q, p, 0, \bar{q}, p, 0)$$

and

$$\mathbf{A}_3 = (p+q, p+q, p+q, p-q, p-q, p-q),$$

leading to the shear matrices:

$$\boldsymbol{\varepsilon}_{2f} = (p - q\tau)/(q + p\tau)$$
$$\boldsymbol{\varepsilon}_{3f} = (p - q\tau)/(q + p\tau)$$

with 1D lattice parameters

$$A_2 = 2^{1/2}(q + p\tau)/(2 + \tau)^{1/2}$$

and

$$A_3 = 6^{1/2}(q + p\tau)/(2 + \tau)^{1/2}.$$

Indexing along the 1D periodic directions gives

$$\begin{cases} L_{2f} = 2(qr + ps) \\ \qquad\qquad \text{for } (r, s, 0, \bar{r}, s, 0) \text{ type reflections} \\ L_{3f} = 3[(p+q)r + (p-q)s] \\ \qquad\qquad \text{for } (r, r, r, s, s, s) \text{ type reflections.} \end{cases}$$

Here, too, a nontrivial common divisor is found, 2 for the twofold case and 3 for the threefold one, showing that systematic extinctions should always occur, independent of the value of the approximant order p/q.

6. Conclusions

We have shown that periodic approximants of quasicrystals described by the perpendicular shear technique can easily be indexed from the standard indexing scheme of the parent quasicrystal. This makes explicit the geometrical relationship between the structures and allows one to define the 'unit-cell' volumes in direct and reciprocal spaces sufficient for calculating an approximant structure from a quasicrystal model. It gives also one way, among others, of quantifying the notion of approximant with respect to the parent phase from the crystallographic point of view. Because the transformation is entirely parametrized by the shear matrix, which, in turn, has entries depending only on a set of rational fractions p/q, we have a simple criterion of quantitatively evaluating the 'closeness' of the approximant with respect to the parent phase; in the case of the icosahedral phases, the closer the value of the fraction is to the number τ, the closer is the approximant structure to the quasicrystal. This would avoid calling 'approximants' those crystalline phases exhibiting some local icosahedral clusters but that are eventually quite far away from any quasiperiodic stacking in their atomic long-range order.

† No such phases have, so far, been experimentally identified.

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