

GROUP 1: NEW APPROACHES TO CRYSTALS AND ANALYSIS OF THEIR STRUCTURES

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Towards a More Comprehensive Crystallography

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Abstract

Despite the intrinsic difference from the point of view of structure of various types of crystals, such as commensurate, incommensurately modulated, intergrowth crystals and quasicrystals, a common approach to their symmetry seems to be possible which eventually will lead to a more comprehensive crystallography. The unifying elements become apparent through treatments which, at first, seem to be contradictory with the geometry of the crystal structures involved. Examples are the description of aperiodic crystals in terms of lattice-periodic structures (going beyond three-dimensionality), the investigation of scaling symmetry in quasicrystals by means of a \mathbb{Z} -module of translations generating a dense set of translationally equivalent atomic positions (going beyond discreteness) and finally the characterization of Euclidean properties of normal crystals through non-Euclidean symmetries (going beyond Euclidean metric). These changing approaches do not modify, however, the fundamental nature of crystals to be three-dimensional, discrete and Euclidean. They only allow implicit symmetry groups like the superspace groups (unifying the crystallography of incommensurate and commensurate crystals) and the multimetric space groups (unifying the possible symmetries of quasicrystals and normal crystals) to be made explicit. Aspects of crystal diffraction, morphology and crystal structure are presented from this unifying point of view, without intending to cover the whole crystallography.

1. Introduction

During the last 20 years the experimental evidence of the existence in nature of new types of crystal structures, such as incommensurately modulated, intergrowth crystals and quasicrystals, profoundly modified the views about the very basic concepts of what is a crystal and which are the crystallographic symmetries of an ideal crystal structure.

Crystals need not be lattice-periodic, so that one currently speaks of *aperiodic crystals*, examples of which are the incommensurate crystals. It is clear that the association of the two words *aperiodic* and *crystal* implies implicit mutual restrictions and extensions. In a number of cases these relations have been made explicit and precise through the characterization of the corresponding symmetry groups. This development started from the modulated crystals, leading to $(3 + d)$ -dimensional *superspace groups*, basically connected with the existence of a set of main reflections of an underlying basic (or average) structure with lattice periodicity (Janner & Janssen, 1977, 1979; de Wolff, 1974, 1977; de Wolff, Janssen & Janner, 1981). The same approach could also be applied to intergrowth (or composite) crystal structures, despite the fact that in this case more than one set of mutually incommensurate main reflections occurs (Janner & Janssen, 1980*a,b*; Kato, 1990; Kato & Onoda, 1991; Petricek *et al.*, 1991; Van Smaalen, 1991; Yamamoto, 1992, 1993). Quasicrystals were first recognized as having long-range order and a diffraction pattern with a noncrystallographic point-group symmetry (Cahn, Gratias, Shechtman & Blech, 1984). Amazingly enough, the same superspace approach could be applied to their structure analysis, even without a lattice of main reflections, because a lattice is incompatible with a noncrystallographic point group (Bak, 1985; Janssen, 1986). A new type has to be added, consisting of what we may call *algorithmic* crystals because they are obtained by molecular beam epitaxy of crystal planes stacked according to a given sequential algorithm, such as that of Fibonacci or Thue–Morse (Axel & Terauchi, 1991; Karkut, Triscone, Ariosa & Fischer, 1986; Merlin, Bajema, Clarke, Juang & Bhattacharya, 1985; Terauchi *et al.*, 1990). More can be expected in the future. This all underlines the open character of crystallography at present.

The originally defined superspace groups, fully classified in the $(3 + 1)$ -dimensional case (Janssen, Janner, Looijenga-Vos & de Wolff, 1992; de Wolff, Janssen & Janner, 1981), are inadequate to describe

the symmetry of all these new types of crystals. Nevertheless, a common approach seems to be possible which eventually will lead to a more comprehensive crystallography. The stage of conceptual simplicity has not yet been reached. What is presented here is a personal view only, as reflected in the literature quoted. Extensive lists of references can be found in some books (DiVincenzo & Steinhardt, 1991; Janot, 1992; Jarić, 1988; Senechal, 1994; Steinhardt & Ostlund, 1987) and review papers (Cummins, 1990; Janssen & Janner, 1987; Janssen, Janner, Loijenga-Vos & de Wolff, 1992; Le Tu Quoc Thang, Piunikhin & Sadov, 1993; Steurer, 1990; Van Smaalen, 1992, 1995).

The unifying fundamental element presented here is a finitely generated group of symmetry translations leading to a \mathbb{Z} -module structure of both the Bragg peaks and of the translationally equivalent atomic positions.* This simplifying assumption (counter examples can be given where the \mathbb{Z} -module structure only occurs either in the reciprocal space or in the direct space) is general enough to include lattice periodicity as a special case and to allow a common symmetry approach for commensurate and incommensurate crystals (Kato & Onoda, 1991; Perez-Mato, 1991; Petricek *et al.*, 1993; Van Smaalen, 1987; Yamamoto, 1981). In the incommensurate case, the important new feature is the possibility of generating a dense set of symmetry-equivalent points. Discreteness is imposed by physical constraints only. For the atomic positions it is, like finiteness, more connected to growth conditions and atomic sizes than to crystallographic symmetry. For the Bragg peaks it is allowed because of a decreasing intensity distribution.

The \mathbb{Z} -module structure of the translations opens not only the possibility of noncrystallographic point groups, but also of rotations of infinite order (Le Tu Quoc Thang, Piunikhin & Sadov, 1993) and of scaling symmetries [involving non-Euclidean transformations (Janner, 1991a, 1992)]. Lifting the \mathbb{Z} -module to a lattice (of higher dimension in the incommensurate case) allows an n -dimensional crystallographic formulation which is, however, richer than the Euclidean crystallography in n -dimensions, even in the case of commensurate crystals (Janner, 1991b,c).

Aspects of crystal diffraction, morphology and crystal structure will be presented from this unifying point of view. For the diffraction, only the cases for which a finite indexing of the Bragg peaks is possible are considered. Algorithmic crystals, like the Thue-Morse one (Axel & Terauchi, 1991), are excluded

* The elements of a \mathbb{Z} -module of rank m can be expressed as integral linear combinations of m vectors defining the basis of the \mathbb{Z} -module. These basis vectors need not be linearly independent over \mathbb{R} as it is required if the \mathbb{Z} -module is an m -dimensional lattice.

here. Concerning morphology, dendritic and fractal crystal growth forms are also disregarded, and only forms limited by flat facets are considered. The discussion of crystal structures is based on the distinction between a *real* and an *ideal* structure. In both cases, imperfections or defects are excluded. By ideal we mean a geometric object defining an atomic arrangement in terms of a set of symmetry-equivalent positions (point-like), whereas a real crystal structure consists of existing atoms. An ideal crystal with space-group symmetry is always infinite, whereas a real crystal is always finite. In the incommensurate case an ideal crystal structure is lattice-periodic in a higher-dimensional space (which need not be Euclidean), it possibly forms a dense set of points in a lower-dimensional space, whereas a real crystal consists of atoms (and not of points), is three-dimensional, Euclidean, discrete and finite. The symmetry of a real crystal is considered to be that of the corresponding ideal structure. Which ideal structure one has to adopt depends on the crystal properties considered.

The aim of the present paper is to illustrate how this distinction between real and ideal (according to Aristotle one would distinguish between actual and potential) allows three-dimensional crystallography to be extended to higher dimensions, to go beyond discreteness and even go beyond Euclidean metric (Janner, 1992).

The approach adopted in the following is always the same. It consists of identifying hidden symmetries in crystals in a way which, at first, seems to be contradictory with the geometrical structure of the given crystal, like the description of aperiodic crystals in terms of lattice-periodic structures, the investigation of scaling symmetry in discrete atomic systems (which cannot be self-similar) and finally the characterization of non-Euclidean symmetries in crystals (which do have an Euclidean structure).

All these developments did not follow from a wish to be original, but reflect mathematical structures which allow a description of crystals occurring in nature in the attempt to deduce physical properties from algebraic structures expressed in terms of suitable symmetry groups.

2. Beyond three dimensions towards higher-dimensional crystallography

The basic assumption is that of Bragg peaks at positions expressible as integral linear combination of n vectors $\mathbf{a}_1^*, \dots, \mathbf{a}_n^*$ spanning the three-dimensional space

$$\mathbf{k} = (k_1, \dots, k_n)_* = \sum_{i=1}^n k_i \mathbf{a}_i^*. \quad (2.1)$$

One also assumes that the reflections having an intensity above a critical value can be labeled by

integral numbers (indices). This implies that they form a discrete set, so that for $n > 3$ a sufficient rapid decrease of intensity for high-indices reflections is required. All such vectors are elements of a \mathbb{Z} -module M^* with basis $\{\mathbf{a}_1^*, \dots, \mathbf{a}_n^*\}$, rank n and dimension 3. The Fourier vectors of the crystal belong to M^* which is, therefore, also called a Fourier module. For $n = 3$, this \mathbb{Z} -module generates the reciprocal lattice. If $n > 3$ and the basis (\mathbf{a}^*) is independent over the rationals, the crystal is incommensurate and thus aperiodic. The rationally dependent case also occurs when the formalism is extended to crystals forming a superstructure. For simplicity, however, if nothing else is stated explicitly, we will here assume rational independence for the basis (\mathbf{a}^*) .

The following steps characterize the superspace approach, which allows to recover lattice periodicity for an n -dimensional ideal crystal structure representing the three-dimensional real crystal.

(1) The three-dimensional (reciprocal) space V^* is embedded as subspace in an n -dimensional one

$$V_s^* = V^* \oplus V_I^*. \quad (2.2)$$

The orthogonal complement V_I^* of V^* in V_s^* is the internal space. [After the discovery of quasicrystals the two components of V_s^* are often designated as *parallel* and *perpendicular* components, respectively: $V^* = V_{\parallel}^*$, $V_I^* = V_{\perp}^*$ (Katz & Duneau, 1986)].

(2) The module basis (\mathbf{a}^*) is lifted to a basis $(\mathbf{a}_s^*) = \{\mathbf{a}_{s1}^*, \dots, \mathbf{a}_{sn}^*\}$ of a reciprocal lattice Σ^* in V_s^* , having the basis (\mathbf{a}^*) as orthogonal projection. Thus

$$\pi \Sigma^* = M^* \quad \text{for} \quad \pi V_s^* = V^*. \quad (2.3)$$

For a rationally independent basis the projection is one-to-one and the elements \mathbf{k}_s of Σ^* have the same components with respect to (\mathbf{a}_s^*) as the elements $\pi \mathbf{k}_s = \mathbf{k}$ of M^* with respect to (\mathbf{a}^*)

$$\mathbf{k} = (k_1, \dots, k_n)_* \longleftrightarrow \mathbf{k}_s = \sum_{i=1}^n k_i \mathbf{a}_{si}^*. \quad (2.4)$$

In the same way, to a three-dimensional linear transformation A of V^* , represented on (\mathbf{a}^*) by an $n \times n$ matrix (A_{ik}) , is associated an n -dimensional linear transformation A_s of V_s^* represented on (\mathbf{a}_s^*) by the same matrix

$$(A_{ik}) = A(\mathbf{a}^*) = A_s(\mathbf{a}_s^*). \quad (2.5)$$

(3) The Fourier components $\hat{\rho}(\mathbf{k})$ of the crystal density $\rho(\mathbf{r})$ are then (uniquely in the incommensurate case) identified with Fourier components at the corresponding reciprocal lattice points of an n -dimensional density ρ_s , which becomes lattice-periodic

$$\hat{\rho}_s(\mathbf{k}_s) \stackrel{\text{def}}{=} \hat{\rho}(\mathbf{k}) \quad \text{for} \quad \pi \mathbf{k}_s = \mathbf{k}. \quad (2.6)$$

(4) Symmetry related are rotationally equivalent Bragg peaks (around the origin) of the same intensity. As the indexed ones form a discrete set, the Laue point group K_L , symmetry of the diffraction pattern, is finite and so also the corresponding group of integral matrices $K_L(\mathbf{a}^*) \subset Gl(n, \mathbb{Z})$. This group represents faithfully a crystallographic point group in n dimensions because it is always possible to define for the basis (\mathbf{a}_s^*) an Euclidean metric tensor left invariant by $K_L(\mathbf{a}^*) = K_L(\mathbf{a}_s^*)$. In this way a same $n \times n$ integral matrix (R_{ik}) represents a three-dimensional and an n -dimensional rotational symmetry.

The Euclidean reciprocal spaces V_s^* , V^* and V_I^* can now be identified with the corresponding direct ones. The density ρ_s describes an Euclidean lattice-periodic structure in n dimensions with space-group symmetry G_s , which is the ideal crystal obeying the rules of n -dimensional crystallography. The relation between ρ_s and ρ is simply an intersection

$$\rho_s(\mathbf{r}_s) \cap V = \rho(\mathbf{r}). \quad (2.7)$$

The Fourier transformation being one-to-one, ρ_s contains the same amount of structural information as ρ

$$\rho(\mathbf{r}) \xleftrightarrow{\text{FT}_3} \hat{\rho}(k_1, \dots, k_n) \equiv \hat{\rho}_s(k_1, \dots, k_n) \xleftrightarrow{\text{FT}_n} \rho_s(\mathbf{r}_s), \quad (2.8)$$

where FT_n denotes the n -dimensional Fourier transform. Therefore, in the incommensurate case an extinction in n dimensions implies an extinction in the diffraction pattern of the real crystal.

This scheme contains as a special case the three-dimensional crystallography and can be applied to most of the known incommensurate crystal structures.

2.1. Systematic extinctions

Concrete examples of observed extinctions (Tables 1, 2 and 3) show that the same type of systematic extinctions are observed in nature, quite independent of the dimension of the ideal crystal.

2.2. Crystal growth forms

Crystal growth forms are interesting objects for crystallographic symmetry considerations because basic structural features become macroscopically visible. The point symmetry of the form reveals, first of all, the geometric crystal class of the point group. The lattice translational symmetry is reflected in the law of rational indices which is based on faces parallel to lattice planes and on the Bravais-Friedel law of the morphological importance of low-indices faces (Friedel, 1911). According to Bravais the indices of a crystal face can be expressed by the components of the Fourier wavevector perpendicular to the given face (Bravais, 1850). One then obtains from the morphology the ratio of the lattice-cell

Table 1. Examples of centering extinctions

CuAu II
 (3 + 1)-dimensional orthorhombic, density modulated (Yamamoto, 1982a)
 Reflections: $ha^* + kb^* + lc^* + mq$; $q = \beta b^* = (0\beta 0)_a$; $M^* = \{a^*, b^*, c^*, q\}$
 Reflection conditions: $hk\ell m$: $h + k = 2n$, $h + l + m = 2n$, $k + l + m = 2n$
 Lattice: (3 + 1)-dimensional orthorhombic centered
 Centering translations: $(\frac{1}{2}100)$, $(\frac{1}{2}010)$, $(0\frac{1}{2}10)$
 Superspace group: $Fmmm(1\beta 0)$

Fe_{1-x}O (wustite)
 (3 + 3)-dimensional cubic modulated, nonstoichiometric (Yamamoto, 1982b)
 Reflections: $ha^* + kb^* + lc^* + m_1q_1 + m_2q_2 + m_3q_3$; $q_1 = \alpha a^* = (\alpha 00)_a$, $q_2 = \alpha b^* = (0\alpha 0)_a$, $q_3 = \alpha c^* = (00\alpha)_a$; $M^* = \{a^*, b^*, c^*, q_1, q_2, q_3\}$
 Reflection conditions: $hk\ell m_1m_2m_3$; $h + k = 2n$, $k + l = 2n$, $h + l = 2n$
 Lattice: (3 + 3)-dimensional cubic $F(1,2,3)$ centered
 Centering translations: $(\frac{1}{2}\frac{1}{2}000)$, $(0\frac{1}{2}\frac{1}{2}00)$, $(\frac{1}{2}0\frac{1}{2}00)$
 Superspace group: $Fm\bar{3}m(\alpha 00)$

Cu₉BiS₆
 (3 + 3)-dimensional cubic modulated (Tomeoka & Ohmasa, 1982)
 Reflections: $ha^* + kb^* + lc^* + m_1q_1 + m_2q_2 + m_3q_3$; $q_1 = (\alpha\alpha\alpha)_a$, $q_2 = (\alpha\bar{\alpha}\alpha)_a$, $q_3 = (\alpha\alpha\bar{\alpha})_a$; $M^* = \{a^*, b^*, c^*, q_1, q_2, q_3\}$
 Reflection conditions: $hk\ell m_1m_2m_3$; $h + k = 2n$, $k + l = 2n$, $h + l = 2n$, $m_1 + m_2 = 2n$, $m_2 + m_3 = 2n$, $m_1 + m_3 = 2n$
 Lattice: (3 + 3)-dimensional cubic $F(1,2,3)F(4,5,6)$ centered
 Centering translations: $(\frac{1}{2}\frac{1}{2}000)$, $(0\frac{1}{2}\frac{1}{2}00)$, $(\frac{1}{2}0\frac{1}{2}00)$, $(000\frac{1}{2}0)$, $(0000\frac{1}{2})$, $(000\frac{1}{2}0)$
 Superspace group: $Fm\bar{3}m(\alpha\alpha\alpha)$

Table 2. Examples of nonsymmorphic group extinctions

γ Na₂CO₃
 (3 + 1)-dimensional monoclinic modulated (van Aalst, den Hollander, Peterse & de Wolff, 1976; Brouns, Visser & de Wolff, 1969)
 Reflections: $ha^* + kb^* + lc^* + mq$; $q = (\alpha 0 \gamma)_a$; $M^* = \{a^*, b^*, c^*, q\}$
 Reflection conditions: $hk\ell m$: $h + k = 2n$; $0\ell lm$: $m = 2n$
 Lattice: (3 + 1)-dimensional monoclinic C-centered
 Centering translations: $(\frac{1}{2}100)$
 Glide: $\{m_1|000\frac{1}{2}\}$
 Superspace group: $C2/m(\alpha 0 \gamma)(0s)$

SC(NH₃)₂ (thiourea)
 (3 + 1)-dimensional orthorhombic modulated (Moudden, Denoyer, Benoit & Fitzgerald, 1978; Moudden, Denoyer & Lambert, 1978; Yamamoto, 1980)
 Reflections: $ha^* + kb^* + lc^* + mq$; $q = (00\gamma)_a$; $M^* = \{a^*, b^*, c^*, q\}$
 Reflection conditions: $0k\ell m$: $k = 2n$; $h0\ell m$: $h + l + m = 2n$
 Lattice: (3 + 1)-dimensional orthorhombic primitive
 Space glide: $\{m_1|0\frac{1}{2}00\}$; superspace glide: $\{m_1|0\frac{1}{2}\frac{1}{2}\}$
 Superspace group: $Pbnm(00\gamma)(0s0)$

ThBr₄
 (3 + 1)-dimensional tetragonal modulated (Bernard *et al.*, 1983)
 Reflections: $ha^* + kb^* + lc^* + mq$; $q = (00\gamma)_a$; $M^* = \{a^*, b^*, c^*, q\}$
 Reflection conditions: $hk\ell m$: $h + k + l = 2n$ (I-centering); $hk00$: $h(k) = 2n$; $hh\ell m$: $2h + l = 4n$; $0k\ell m$: $m = 2n$
 Lattice: (3 + 1)-dimensional I-centered tetragonal
 Centering translations: $(\frac{1}{2}\frac{1}{2}\frac{1}{2}0)$
 Glides: $\{m_1|0\frac{1}{2}0\}$; $\{m_2|0\frac{1}{2}0\}$; $\{m_3|000\frac{1}{2}\}$
 Superspace group: $I4_1/amd(00\gamma)(s0s0)$

parameters. These same laws allow to deduce lattice centering as well. Finally, nonsymmorphic symmetry elements (glides and screw axes) also modify the morphological importance of a given set of indices, as expressed by the Donnay–Harker extension of Bravais–Friedel’s law (Donnay & Harker, 1937). A

Table 3. Example of a quasicrystal

Al₇₈Mn₂₂ Decagonal phase
 5-modules M and M^* of rank 5 and dimension 3 (Steurer, 1991)
 Fourier module $M^* = \{a_1^*, \dots, a_5^*\}$ with components expressed in an orthonormal basis of V : $a_k^* = a^*[\cos(2\pi k/5), \sin(2\pi k/5), 0]$ for $k = 1, \dots, 4$ and $a_5^* = c^*(0, 0, 1)$
 Translational module $M = \{a_1, \dots, a_5\}$ with $a_k = z a^*[\cos(2\pi k/5) - 1, \sin(2\pi k/5), 0]$ for $k = 1, \dots, 4$ and $a_5 = c^*(0, 0, 1)$, where $a^* a = c^* c = 1$
 Reciprocal lattice Σ^* in $V_s = V \oplus V_s$, with V_s a two-dimensional internal space, direct and reciprocal space being identified:
 $a_{k,s}^* = [a_k^*, a_{k,s}^*]$, where $a_{k,s}^* = a^*[\cos(4\pi k/5), \sin(4\pi k/5)]$ for $k = 1, \dots, 4$ and $a_{5,s}^* = 0$
 Translational lattice Σ : $a_{k,s} = [a_k, a_{k,s}]$ with $a_k = z a^*[\cos(4\pi k/5) - 1, \sin(4\pi k/5)]$ for $k = 1, \dots, 4$ and $a_{5,s} = 0$
 The lattice bases (a_k^*) and (a_k) are reciprocal in the Euclidean five-dimensional superspace
 Superspace group: $P10_c/mmc$
 Occupied positions in Al₇₈Mn₂₂ (given is the multiplicity, the Wyckoff letter, the site symmetry and the coordinates):
 2 (a') $10m\bar{2}$ 00000, 0000 $\frac{1}{2}$
 2 (b') $10m\bar{2}$ $\frac{z}{2}\frac{z}{2}\frac{z}{2}\frac{z}{2}0$, $\frac{z}{2}\frac{z}{2}\frac{z}{2}\frac{z}{2}\frac{1}{2}$
 4 (c') $5m$ $\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}$, $\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}$, $\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}$, $\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}$, $\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}$, $\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}\frac{z}{4}$
 Atom 1 and atom 2 are at c' with $z = 0.1858$ and $z = 0.027$, respectively
 Atom 3 is at a' and atom 4 at b' , all with their own pentagonal acceptance region $\Omega_i(t)$

full set of crystal growth forms contains in this way the same geometrical information as given in a diffraction pattern by its point symmetry and by the conditions for reflection. The main difference between a diffraction pattern and a crystal growth form is a missing correspondence between diffraction intensity and morphological importance. There are good reasons for this: the Bragg intensity is dominantly determined by the charge at atomic positions, whereas the stability of a crystal face mainly depends on the bonding charges, as made explicit in the theory of Hartmann–Perdok of periodic bond chains [PBC (Hartman & Perdok, 1955)]. Nevertheless, an indirect relation is certainly present because of the structural relevance of both the Fourier wavevectors \mathbf{k} and of the Fourier components $\hat{\rho}(\mathbf{k})$ of the charge distribution.

Concluding, the whole space-group symmetry can be deduced from morphological measurements only: not, of course, the absolute length of the symmetry translations.

The morphology of incommensurate crystals is a challenging subject because there is no lattice symmetry anymore and thus also no lattice planes for the atoms. The superspace approach allows, in this case also, a natural extension of the concepts involved, with facets which are the intersection in space of lattice hyperplanes labeled by the reciprocal lattice vectors obtained from the Fourier module M^* [see Fig. 1 (Janner, 1983)].

One then verifies the validity of the classical law of rational indices and (even if in a less evident way) Bravais–Friedel (Janner, Rasing, Bennema & Van der Linden, 1980), Donnay–Harker and Hartmann–Perdok. For the latter the PBC’s are sets of bonds between the extended ‘world lines’ of the modulated atomic positions in superspace (Kremers, Meekes,

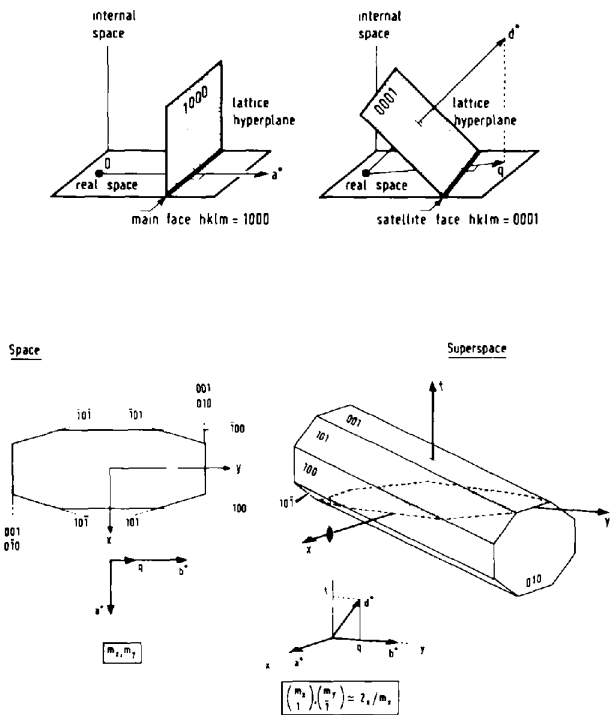


Fig. 1. Superspace lattice planes (here in three dimensions) giving rise by intersection with the physical space (here two-dimensional) to crystal growth forms with (normal) main faces and (additional) satellite faces. The point group of the forms in space and in superspace is consistent with the embedding of the rotational symmetries [Figs. 13 and 15 of Janner (1983)].

Bennema, Balzuweit & Verheijen, 1994). Despite the fact that crystal faces are no more lattice planes, and that their microscopic structure is still fairly unclear, flat faces are thermodynamically stable: deviation from orthogonality with respect to the Fourier wavevector labeling the face gives rise to a (reversible) roughening transition (Dam & Bennema, 1987). It has been possible to determine in this way, in addition to the ratios of the average-lattice parameters, the variation in temperature of the components of the modulation wavevector (Dam & Janner, 1986). Up to an arbitrary unit of length, the generators of the Fourier module M^* are, therefore, morphologically determined. What is known on the morphology of quasicrystals is consistent with the results obtained for modulated crystal structures. (See, for example, Ho, 1991 and references therein; Janssen, Janner & Bennema, 1989.)

Despite the conceptual relevance of these results, incommensurate crystal morphology is still under developed: very little is known so far on the morphology of intergrowth crystals and all the investigations on incommensurately modulated crystals have been carried out in the Netherlands only, mainly in Nijmegen. For this reason it was essential to have at our disposal the very careful measurements on the mineral calaverite by Herbeth Smith (Smith, 1902) and by the team Goldschmidt, Palache & Peacock (1931) at a time when incommensurability was still nothing more than an intuition.

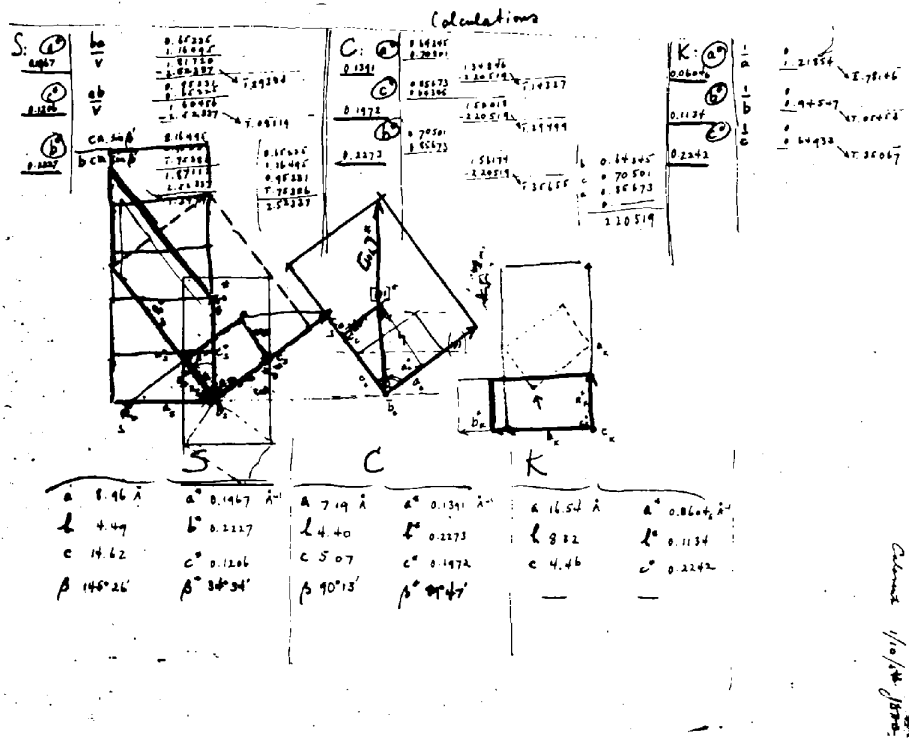


Fig. 2. Work sheet with annotations of J. D. H. Donnay, dating from 1954, in one of his attempts to solve the puzzling morphology of calaverite. It is presented here in honor of the man who played an essential rôle in the process of understanding the morphology of incommensurate crystals. It is one of the historical documents he gave to the author.

J. D. H. Donnay suggested to reconsider these measurements in the frame of incommensurate crystals. At that time (1984) he was still not satisfied with the interpretation he gave in 1935 in a preliminary communication [see Fig. 2 (Donnay, 1935)]. Calaverite could eventually be shown to be incommensurate (Tendeloo, Gregoriades & Amelinckx, 1983), a result confirmed by a crystal structure analysis (Schutte & de Boer, 1988). By means of a superspace approach applied to the measurements of Goldschmidt, Palache & Peacock (1931), full agreement could be reached between theory and experiment [see Fig. 3 (Dam, Janner & Donnay, 1985; Janner & Dam, 1989)]. A Wulff-plot approach, giving a thermodynamical explanation of the superspace extension of the

law of rational indices, has been developed by Van Smaalen (Van Smaalen, 1993).

3. Beyond discreteness towards positional self-similarity

Most quasiperiodic tilings are obtained by deflation (and inflation) procedures generating self-similar structures with scaling factors intimately related to the orientational symmetry of the tiling. Replacing each of the tiles by a corresponding atomic arrangement (the so-called decoration) leads to models of quasicrystal structures having a Fourier module M^* of finite rank (larger than the dimension), so that one can apply the superspace approach (Janssen, 1986). The tiling approximation (where the relaxation of the structure due to the different local surrounding of a given atomic position in space is neglected) corresponds to a basic structure description. Therefore, the translational module M obtained from the orthogonal projection into the physical space of the lattice of symmetry translations Σ is dual to M^* , and it has the same rank (Janner, 1991a)

$$M^* \xleftarrow{\pi} \Sigma^* \xleftrightarrow{\text{duality}} \Sigma \xrightarrow{\pi} M. \quad (3.1)$$

This is the basis of the cut-and-projection method (Katz & Duneau, 1986). Translationally equivalent points (by Σ) in superspace give, by projection, translationally equivalent points (by $M = \pi\Sigma$) in space. The corresponding atomic positions differ by vectors \mathbf{a} which are an integral linear combination of a set of vectors $\{\mathbf{a}_1, \dots, \mathbf{a}_n\}$ basis of the translational module M

$$\begin{aligned} \mathbf{a} &= (z_1, \dots, z_n) \\ &= z_1 \mathbf{a}_1 + \dots + z_n \mathbf{a}_n \quad \text{with integers } z_i. \end{aligned} \quad (3.2)$$

By construction, the n -dimensional point-group symmetry (holohedry) of the lattice Σ is represented by integral matrices $R_s(\mathbf{a}_s) \in GL(n, \mathbb{Z})$ which define, when referred to the projected basis (\mathbf{a}) for M , a three-dimensional rotation $R(\mathbf{a})$ leaving the \mathbb{Z} -module M invariant

$$\begin{aligned} R\mathbf{M} &= \mathbf{M} \quad \text{for } R \in O(3), \\ R_s \Sigma &= \Sigma \quad \text{for } R_s \in O(n), \end{aligned} \quad (3.3)$$

with $R_s(\mathbf{a}_s) = R(\pi\mathbf{a}_s) = R(\mathbf{a}) \in GL(n, \mathbb{Z})$. Therefore, symmetry-equivalent atomic positions in space are related by three-dimensional Euclidean transformations forming a group G , which is the projection of the n -dimensional space group G_s of the embedded ideal crystal structure (Janner, 1992). The converse is not true, as only superspace positions having their internal space component within a given acceptance region Ω_I (which is the atomic surface in the description adopted in the reciprocal space embedding discussed in the previous section) project to points of

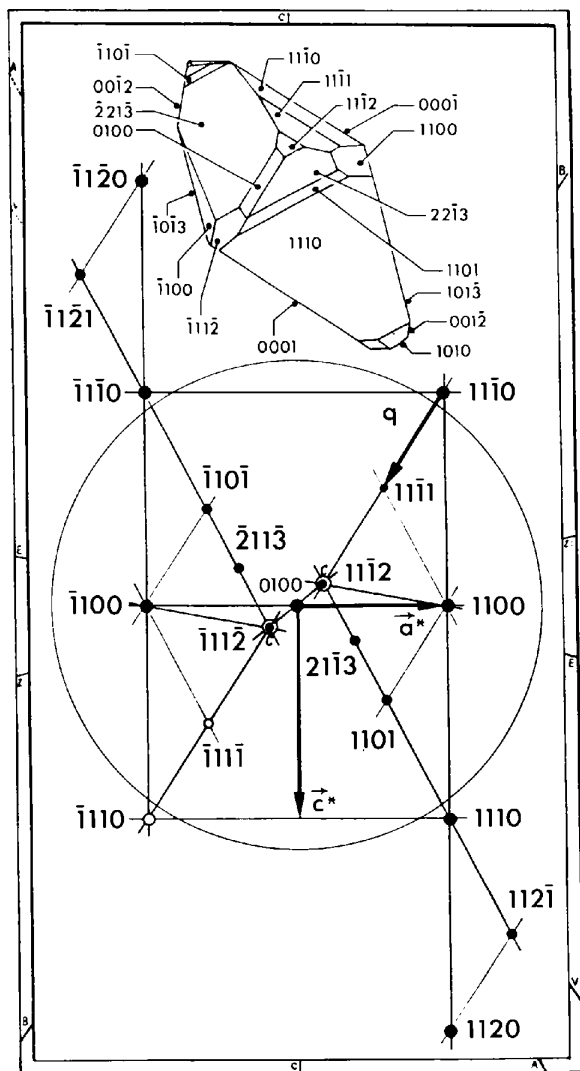


Fig. 3. The original drawing published in the paper by Goldschmidt, Palache & Peacock has been modified in order to show the underlying \mathbb{Z} -module structure of the four-indices labeling of the faces in calaverite crystals [Fig. 2(b) of Janner & Dam (1989)].

the decorated tiling (cut-projection). The reciprocal space and its embedding in the superspace is the most convenient one for quasicrystal structure determination (Janssen, 1986; Mermin, 1991, 1992a,b; Mermin & Lifshitz, 1992; Rabson, Mermin, Rokhsar & Wright, 1991), whereas the direct space and the embedding considered now are suited for discussing the symmetries of a known quasicrystal structure.

Within this last description, atomic positions in space and superspace are represented by points. The acceptance region is the internal component only of a superspace region $\Omega_s = (\Omega, \Omega_i)$ separating a set of symmetry-equivalent occupied positions into *real* ones, projecting to existing atomic positions, and into *virtual* ones where no atom is observed. The space component Ω implies that a real crystal is finite, whereas the internal component Ω_i implies that the structure is a discrete one. The former limits the largest possible interatomic distances, and the latter the shortest possible ones. Both are essential for the real structure, whereas it is on the ideal structure level that symmetry transformations apply. The real structure forms a subset only of the symmetry-equivalent occupied positions, which in space (for n larger than 3) are dense as required for self-similar patterns.

The symmetry one has to consider for self-similarity involves positions only, not the charge distribution in direct space, nor reflections with equal intensity in reciprocal space. A necessary condition for a point-group transformation A to be a symmetry is to leave the set of translationally equivalent positions invariant, *i.e.* the module M , and in order to represent self-similar transformations one also asks that A is a homothety, which is a rotation up to a dilation (or contraction)

$$AM = \lambda M = M \text{ for } A \in H(3),$$

and

$$A(\mathbf{a}) \in Gl(n, \mathbb{Z}), \quad (3.4)$$

where $H(3)$ is a three-dimensional group of homotheties. (The dilations may involve a subspace only.) One recovers the crystallographic discreteness associated with lattice symmetry and finite atomic multiplicity by lifting the \mathbb{Z} -module M to an Euclidean lattice Σ , requiring as in the previous section that space rotations become superspace rotations. The homothety A then becomes an n -dimensional linear symmetry transformation A_s , which cannot be an orthogonal transformation of $O(n)$ (because of infinite order). In the pure scale case, A_s can be cast to be an element of $O(n-d, d)$, but in general it is only expressible as a product of a positive definite and an indefinite metric symmetry transformation. Loosely speaking, the point-group elements of a self-similar quasicrystal are, in general, products of circular and hyperbolic rotations.

The corresponding crystallographic positional symmetry is thus an affine space group obtained from a point group, subgroup of $Gl(n, \mathbb{Z})$, and a lattice group. This leads to a multimetric space group having the space group of the structure as the Euclidean subgroup (Janner, 1991b). The specific conditions implied by these concepts are not essential in the present context. It is relevant that the space group (or the superspace group) is not a complete characterization of the possible symmetries of quasicrystals (Janner, 1991a, 1992).

The crystallography can be extended to include the scaling symmetries of the atomic positions as well. To obtain a feeling of the new features, in the space and in the superspace, consider a linear transformation A acting on the module basis of M

$$A\mathbf{a}_i = \sum_{j=1}^n \mathbf{a}_j A_{ji}(\mathbf{a}). \quad (3.5)$$

With respect to the Euclidean scalar product (here in space) to A , there corresponds a unique adjoint transformation A^\dagger defined by

$$\mathbf{q} \cdot A\mathbf{r} = A^\dagger \mathbf{q} \cdot \mathbf{r} \text{ for any } \mathbf{q}, \mathbf{r} \in V. \quad (3.6)$$

The action of A^\dagger on the dual basis of the module M^* is

$$A^\dagger \mathbf{a}_i^* = \sum_{j=1}^n \mathbf{a}_j^* A_{ji}^\dagger(\mathbf{a}^*). \quad (3.7)$$

The same matrices are obtained when considering the corresponding transformations A_s and A_s^\dagger acting on the dual lattice bases (\mathbf{a}_s) and (\mathbf{a}_s^*) with respect to the Euclidean scalar product in the superspace and for which the duality condition takes the familiar form

$$\mathbf{a}_{si}^* \cdot \mathbf{a}_{sj} = \delta_{ij}. \quad (3.8)$$

From that follows directly

$$A^\dagger(\mathbf{a}^*) = A_s^\dagger(\mathbf{a}_s^*) = \tilde{A}_s(\mathbf{a}_s) = \tilde{A}(\mathbf{a}). \quad (3.9)$$

Note that the step involving an Euclidean superspace is necessary, because duality of the \mathbb{Z} -modules in space is, in general, not expressible as a scalar-product duality between the corresponding bases (\mathbf{a}^*) and (\mathbf{a}) .

In order to leave the module M invariant, $A(\mathbf{a})$ has to be an element of $Gl(n, \mathbb{Z})$. In that case, A^\dagger also leaves M^* invariant. The same can be said for the multimetric point-group symmetries A_s of Σ and A_s^\dagger of Σ^* . Therefore, to a multimetric point group K leaving M (and correspondingly Σ) invariant, there is an adjoint point group K^\dagger leaving M^* (and Σ^* after embedding) which is invariant. In general, these two point groups are different, but in the case when the generators of the point group are either orthogonal transformations (as is usual) or hyperbolic rotations (corresponding to scalings in V), the two point

groups K and K^\dagger are the same. Indeed, for orthogonal transformations R one has

$$R^\dagger = R^{-1}, \tag{3.10}$$

whereas hyperbolic rotations L are self-adjoint

$$L^\dagger = L. \tag{3.11}$$

The different behavior follows from the property of a circular rotation to be antisymmetric and a hyperbolic one to be symmetric when expressed with respect to an orthonormal basis. For general homotheties of the type $A = LR$, however, the groups generated by A and by $A^\dagger = R^{-1}L$ are different.

3.1. Examples of self-similar modules

Decagonal case. (Janner, 1992) The three-dimensional rank 5 modules M and M^* considered in the previous section for the $Al_{78}Mn_{22}$ are invariant with respect to the scaling S by a factor of τ^2 in the quasiperiodic plane {where $\tau = (1 + 5^{1/2})/2$ } giving rise to an hyperbolic rotation S_s around the axis a_s in the superspace. S_s is an element of $O(3,2)$ leaving invariant the indefinite metric tensor $g_m = (1, 1, \bar{1}, \bar{1}, 1)$ of an orthonormal basis of V_s . The symmetry transformations S and S_s are represented by the same integral matrix

$$S(\mathbf{a}) = S_s(\mathbf{a}_s) = \begin{pmatrix} 1 & 0 & -1 & -1 & 0 \\ 1 & 2 & 1 & 0 & 0 \\ 0 & 1 & 2 & 1 & 0 \\ -1 & -1 & 0 & 1 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix} = (\tau^2). \tag{3.12}$$

This transformation permutes the two atomic positions $\frac{11111}{33333}z$, $\frac{44444}{33333}z$, and the other two $\frac{22222}{33333}z$, $\frac{33333}{33333}z$, exactly as the tenfold rotation R (and R_s , respectively)

$$R(\mathbf{a}) = R_s(\mathbf{a}_s) = \begin{pmatrix} 0 & 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & -1 & 0 \\ 1 & 1 & 1 & 1 & 0 \\ -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 \end{pmatrix} = 10. \tag{3.13}$$

Therefore, the quasicrystal structure of $Al_{78}Mn_{22}$ presented above is left invariant by the multimetric space group G_s generated by the lattice translations Σ , together with the elements

$$g_1 = \{R|0,0,0,0,\frac{1}{2}\} = 10_s, \tag{3.14}$$

$$g_2 = \{m_1|0,0,0,0,0\} = m_1, \tag{3.15}$$

$$g_3 = \{m_2|0,0,0,0,\frac{1}{2}\} = c, \tag{3.16}$$

$$g_4 = \{m_z|0,0,0,0,0\} = m_z \tag{3.17}$$

of the five-dimensional space group $P10_s/mmc$ and by the screw hyperbolic rotation

$$g_5 = \{S|0,0,0,0,\frac{1}{2}\}. \tag{3.18}$$

In space, g_5 corresponds to a screw scaling symmetry. In these expressions the point-group elements are matrices and the subscript s can be omitted. The structural interplay between these symmetries becomes evident once one considers the multimetric point group K_0 leaving the decagonal plane (perpendicular to the c axis) invariant

$$K_0 = \{R^2, S^2, m_1, m_2, R, m_2 S, R^5 S, RS\}. \tag{3.19}$$

K_0 is the homothety symmetry group of a self-similar pentagram, involving an infinite series of scaled regular pentagons. From a superspace point of view, pentagram and regular pentagon are external-internal dual (Senechal, 1994), both in the direct and in the reciprocal space. By this we mean that con-

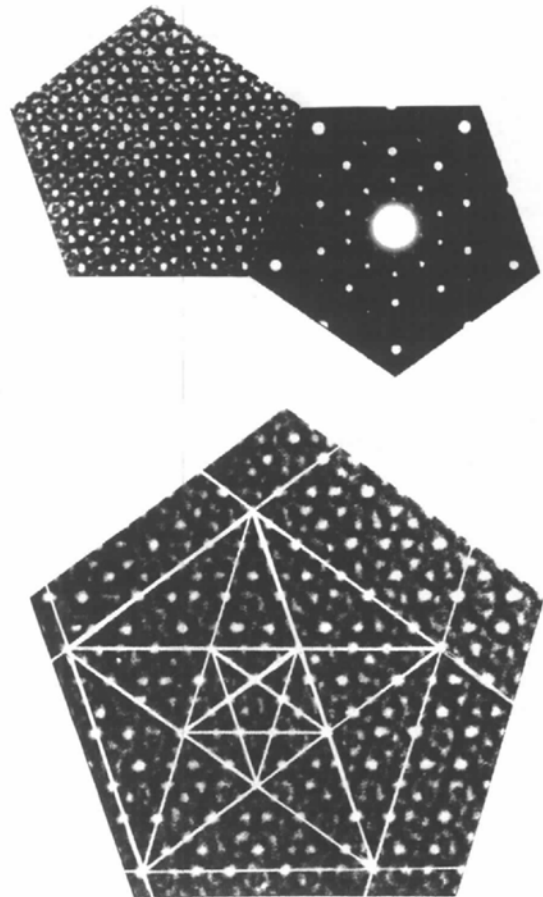


Fig. 4. After having recognized the existence of a pentagrammatic rotation/scale symmetry in the decagonal quasicrystal $Al_{78}Mn_{22}$ (Janner, 1992), the idea came to verify whether that was also the case for the icosahedral $Al_{6}Mn$ phase. The result is shown on a photograph sent with the New Year's greetings for 1986 by the colleagues of the Center for High-Resolution Microscopy of the University of Antwerpen (RUCA).

sidering a pentagon defined by the sequence \mathbf{a}_0^* , \mathbf{a}_1^* , \mathbf{a}_2^* , \mathbf{a}_3^* , \mathbf{a}_4^* , \mathbf{a}_0^* (where $\mathbf{a}_0^* = -\sum_{i=1}^4 \mathbf{a}_i^*$) one obtains a pentagram from the corresponding sequence of internal components \mathbf{a}_{70}^* , \mathbf{a}_{71}^* , ..., \mathbf{a}_{70}^* of the basis (\mathbf{a}_i^*). Conversely, to the pentagram sequence \mathbf{a}_0^* , \mathbf{a}_2^* , \mathbf{a}_4^* , \mathbf{a}_1^* , \mathbf{a}_3^* , \mathbf{a}_0^* corresponds to an internal pentagon. Analogous properties are found in the direct space. A same pentagrammatic structure appears in the diffraction pattern and in a HREM picture of the icosahedral Al_6Mn phase (see Fig. 4).

Octagonal case (Janner, 1991a). In exactly the same way one arrives at an external-internal octagon-octagram duality and to a self-similarity point group of homotheties leaving invariant the translational module M (and the Fourier module M^* , respectively) of an octagonal aperiodic tiling. Despite the fact that the structure of the octagonal CrNiSi alloy obtained by Kuo has not yet been determined, a HREM picture presented in a poster

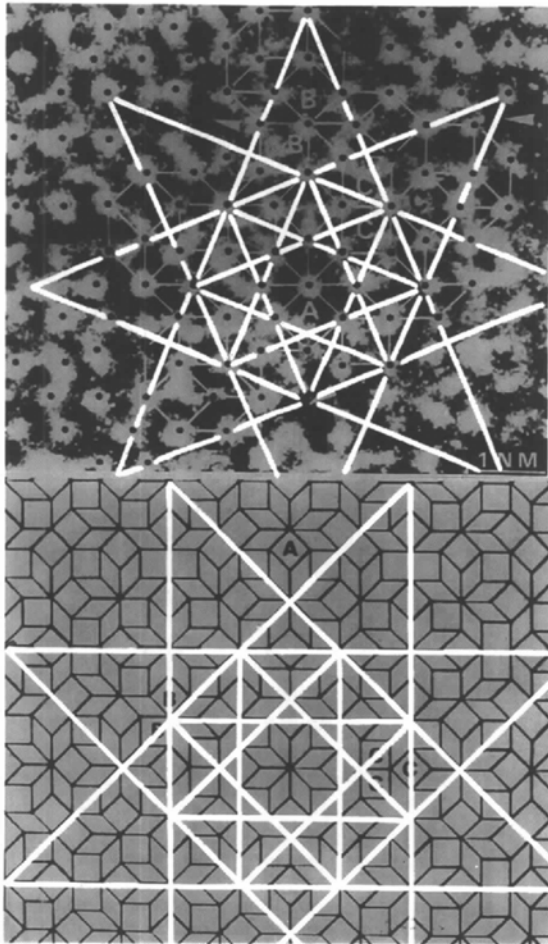


Fig. 5. In the HREM picture of the octagonal CrNiSi alloy presented by K. H. Kuo at the International Crystallography Congress of the IUCr in Perth (Kuo, 1987), the octagrammatic rotation/scale symmetry is made visible.

at the Perth meeting of the IUCr (Kuo, 1987) gives experimental evidence of the octagrammatic symmetry of this quasicrystal (see Fig. 5).

Restricting for simplicity the considerations to the plane and, correspondingly, to a four-dimensional multimetric superspace, one has:

(1) A two-dimensional rank-four-translational module $M = \{\mathbf{a}_1, \dots, \mathbf{a}_4\}$ defined by the vectors $\mathbf{a}_k = a(\cos 2\pi k/8, \sin 2\pi k/8)$, for $k = 1, \dots, 4$.

(2) A translational lattice $\Sigma = \{\mathbf{a}_{s1}, \dots, \mathbf{a}_{s4}\}$ with

$$\mathbf{a}_{sk} = (\mathbf{a}_k, \mathbf{a}_{jk}) \text{ where } \mathbf{a}_{jk} = \mathbf{a}_{3k}. \quad (3.20)$$

(3) The point group of the self-similar M with as generators: a scaling S with scaling factor $\lambda = 2^{1/2} + 1$

$$S(\mathbf{a}) = \begin{pmatrix} 1 & 1 & 0 & \bar{1} \\ 1 & 1 & 1 & 0 \\ 0 & 1 & 1 & 1 \\ \bar{1} & 0 & 1 & 1 \end{pmatrix} = (2^{1/2} + 1), \quad (3.21)$$

an eightfold rotation R and a mirror m

$$R(\mathbf{a}) = \begin{pmatrix} 0 & 0 & 0 & \bar{1} \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \end{pmatrix},$$

$$m(\mathbf{a}) = \begin{pmatrix} 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \\ 0 & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 \end{pmatrix}. \quad (3.22)$$

The multimetric point-group symmetry of the octagram (leaving the octagonal quasicrystal phase invariant) is

$$K_0 = \{R, m, S\} = 8mm(2^{1/2} + 1), \quad (3.23)$$

where $(2^{1/2} + 1)$ symbolizes S by its scaling factor. The atomic positions occupied in the octagonal phase of the CrNiSi alloy are expected to be left invariant by a multimetric space group having at least this point group.

4. Beyond Euclidean metric, towards multimetric crystallography

Taking into account scaling symmetries in quasicrystals together with the rotational ones within a lattice-periodic description of the structure (according to a superspace approach) leads to consider higher-dimensional affine point groups generated by circular rotations (for the rotational symmetries) and hyperbolic rotations (for the scaling symmetries). The compatibility between Euclidean and pseudo-Euclidean transformations is ensured by the condition for both to leave a same lattice of symmetry

translations invariant. The superspace symmetry group of a self-similar quasicrystal (in the sense explained in the previous section) is, therefore, a multimetric space group which combines, in the usual way, point group and lattice symmetry (Janner, 1991b).

The belief in the existence of a more comprehensive crystallography for all crystal structures observed in nature is at the basis of an attempt to understand the possible physical relevance of multimetric space-group symmetry for ordinary crystals (Janner & Nusimovici, 1994). In this exploration one has to keep in mind two basic facts about multimetric symmetry which have already been pointed out in the case of quasicrystals. First, the symmetry applies to atomic positions (and not to charge density). Second, in normal crystals this symmetry is usually broken. Two simple cases will be considered as an illustration, the first is based on the two-dimensional hexagonal lattice, the second deals with the three-dimensional lattice of an ideal hexagonal close-packed structure [for which one has $c/a = (8/3)^{1/2}$]. The ideas will be presented without exploring theoretical details.

4.1. Hexagonal two-dimensional multimetric space groups

The hexagonal lattice A_{hex} is generated by the basis vectors

$$\mathbf{a}_1 = a(1,0), \mathbf{a}_2 = a(-1/2, 3^{1/2}/2) \quad (4.1)$$

with components expressed with respect to an orthonormal basis $\mathbf{e}_1, \mathbf{e}_2$ defining the Euclidean metric tensor $g_e = (1,1)$. As is well known, A_{hex} is invariant with respect to the Euclidean point group $K_0 = 6mm = \{R, m_0\}$, with R a sixfold rotation and m_0 a mirror

$$R(\mathbf{a}) = \begin{pmatrix} 1 & \bar{1} \\ 0 & 1 \end{pmatrix} \text{ and } m_0(\mathbf{a}) = \begin{pmatrix} \bar{1} & 1 \\ 0 & 1 \end{pmatrix}. \quad (4.2)$$

The mirrors of the form

$$m_k = R^k m_0 = m_0 R^{-k} \quad (4.3)$$

generate rotations according to

$$m_{j+k} m_j = R^k. \quad (4.4)$$

Consider now an hyperbolic rotation L with eigenvalues $2 \pm 3^{1/2}$

$$L(\mathbf{e}) = \begin{pmatrix} \cosh\chi & \sinh\chi \\ \sinh\chi & \cosh\chi \end{pmatrix} = \begin{pmatrix} 2 & 3^{1/2} \\ 3^{1/2} & 2 \end{pmatrix}, \quad (4.5)$$

leaving the pseudo-Euclidean metric tensor $g_m = (1, \bar{1})$ invariant, where $\bar{1}$ is -1 . One verifies that L also leaves the lattice A_{hex} invariant. Indeed

$$\begin{aligned} L\mathbf{a}_1 &= a(2, 3^{1/2}) = 3\mathbf{a}_1 + 2\mathbf{a}_2 \\ L\mathbf{a}_2 &= a(1/2, 3^{1/2}/2) = \mathbf{a}_1 + \mathbf{a}_2, \end{aligned} \quad (4.6)$$

Table 4. Multimetric space group $G = p6\hat{4}$

A number of these orbits are noncharacteristic, as one can see by comparison with corresponding sets of equivalent positions for the multimetric space group $p6mm\hat{4}$ (Table 5).

Multiplicity	Wyckoff letter	Site symmetry	Coordinates of equivalent positions
1	(a)	$6\hat{4}$	0,0
2	(b)	$3\hat{4}^2$	$\frac{1}{3}, \frac{1}{3}, \frac{1}{3}$
3	(c)	$2\hat{4}^2$	$\frac{1}{2}, 0, \frac{1}{2}$
6	(d)1	$\hat{4}^2$	$\frac{1}{6}, \frac{1}{6}, \frac{1}{6}; \frac{1}{6}, \frac{1}{6}, \frac{1}{6}; \frac{1}{6}, \frac{1}{6}, \frac{1}{6}; \frac{1}{6}, \frac{1}{6}, \frac{1}{6}; \frac{1}{6}, \frac{1}{6}, \frac{1}{6}; \frac{1}{6}, \frac{1}{6}, \frac{1}{6}$ ($x = \frac{1}{6}, y = \frac{1}{6}$)
6	(d)2	$\hat{4}^6$	$\frac{1}{3}, 0, \frac{1}{3}; \frac{1}{3}, \frac{1}{3}, 0; 0, \frac{1}{3}, \frac{1}{3}$ ($x = \frac{1}{3}, y = 0$)
12	2(d)1	$\hat{4}^4$	$\frac{1}{4}, 0 - \frac{1}{4}, \frac{1}{4}$ ($x_1 = \frac{1}{4}, y_1 = 0; x_2 = \frac{1}{4}, y_2 = \frac{1}{4}$)
...
18	3(d)1	$\hat{4}^6$	$\frac{1}{6}, 0 - \frac{1}{6}, \frac{1}{6} - \frac{1}{6}, \frac{1}{6} -$ ($x_1, y_1; x_2, y_2; x_3, y_3$)
18	3(d)2	$\hat{4}^6$	$\frac{1}{3}, \frac{1}{3} - \frac{1}{3}, \frac{1}{3} -$
...
24	4(d)1	$\hat{4}^3$	$\frac{1}{4}, 0 - \frac{1}{4}, \frac{1}{4} - \frac{1}{4}, \frac{1}{4} -$
24	4(d)2	$\hat{4}^4$	$\frac{1}{2}, \frac{1}{2} - \frac{1}{2}, \frac{1}{2} - \frac{1}{2}, \frac{1}{2} -$
...

so that

$$L(\mathbf{a}) = \begin{pmatrix} 3 & 1 \\ 2 & 1 \end{pmatrix}. \quad (4.7)$$

In the same way as for the circular rotation R , hyperbolic mirrors \hat{m}_k also leaving the lattice invariant

$$\hat{m}_k = L^k \hat{m}_0 \text{ with } \hat{m}_0 = m_0 \quad (4.8)$$

generate hyperbolic rotations L^k

$$\hat{m}_{j+k} \hat{m}_j = L^k. \quad (4.9)$$

Note that $m_0: \mathbf{e}_1 \rightarrow -\mathbf{e}_1, \mathbf{e}_2 \rightarrow \mathbf{e}_2$ leaves both the Euclidean and the pseudo-Euclidean metric invariant. A hyperbolic mirror has the general form

$$\hat{m}(\mathbf{e}) = \pm \begin{pmatrix} \cosh\varphi & \sinh\varphi \\ -\sinh\varphi & -\cosh\varphi \end{pmatrix} \quad (4.10)$$

and reflects a point along a line having, together with the mirror-invariant line, the directions of the isotropic vectors (the light cone) as bisectors.

Summarizing: the hexagonal lattice A_{hex} has as multimetric symmetry the point group

$$K = \{R, m_0, L\} = 6mm\hat{4} = 6mm(2 + 3^{1/2}), \quad (4.11)$$

where the generator L has been symbolized, as in previous publications, by $2\cosh\chi$ (which is here equal to 4), or by its eigenvalue $\lambda = 2 + 3^{1/2}$, the other eigenvalue being the conjugated one $\lambda' = 2 - 3^{1/2}$.

Examples of symmorphous multimetric hexagonal space groups can easily be obtained by adding, for example, a pseudo-Euclidean point-group symmetry to the generators of hexagonal space groups. So

$$\begin{aligned} p6\hat{4} &= \{A_{hex}, R, L\}, p6mm\hat{4} = \{A_{hex}, R, m_0, L\}, p3m\hat{1}\hat{4}^2 \\ &= \{A_{hex}, R^2, m_0, L^2\}, \end{aligned} \quad (4.12)$$

and so on. The general position of a multimetric space group is of infinite multiplicity and, therefore, excluded for atoms in crystals. The low-multiplicity

Table 5. *Multimetrical space group* $G = p6mm\hat{4}$

Multiplicity	Wyckoff letter	Site symmetry	Coordinates of equivalent positions
1	(a)	$6mm\hat{4}$	0,0
2	(b)	$3m\hat{4}^2$	$\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$
3	(c)	$2mm\hat{4}^2$	$\frac{1}{2}, 0, \frac{1}{2}$ $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$
6	(d)	$m\hat{4}^6$	$\frac{1}{2}, 0, \frac{1}{2}$ $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ $\frac{1}{2}, 0, \frac{1}{2}$ $0, \frac{1}{2}, \frac{1}{2}$ ($x = \frac{1}{2}, y = 0$)
6	(e)	$m\hat{4}^2$	$\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ $\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$ ($x = \frac{1}{2}$)
...

cases, however, show surprising compatibilities between Euclidean and multimetrical space groups. As an illustration, in Table 4 a number of sets of equivalent positions is given for the multimetrical space group $p6\hat{4}$ (see also Table 5). Indicated also are the multiplicities, the site symmetries and labels given by letters taken from those of the Euclidean subgroup $p6$. The integer preceding the Wyckoff letter indicates the number of Wyckoff sets obtained when restricting to the Euclidean subgroup. The number after the letter is a sequential index. For the multiplicities 12 and higher, a representative only is given for each set of coordinates belonging to a sixfold rotational orbit. This is also suggested by indicating some of the corresponding x, y parameters involved. (—) indicates the missing coordinates.

Because of the occurrence of fairly large multiplicities, one needs a graphical representation of the various sets of equivalent positions in order to grasp their structural features. Looking at these pictures, one recognizes that the interplay between circular and hyperbolic rotations gives rise to families of

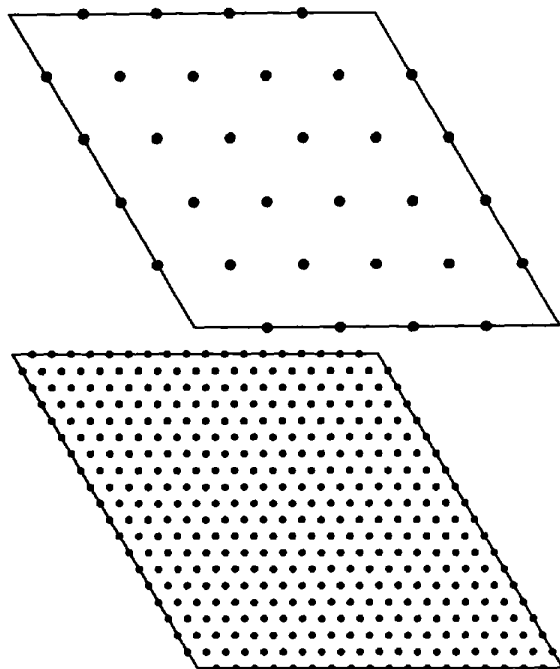


Fig. 6. Lattice-like sets of $p6\hat{4}$ -equivalent positions having site symmetry $\hat{4}^3$ and $\hat{4}^5$, respectively. The missing points belong to an orbit of multiplicity 1 and site symmetry $\hat{4}$ (cf. Fig. 7).

Wyckoff orbits having a group-subgroup relation in the site symmetry and which are relatively scaled with respect to each other.

The simplest family, with points invariant with respect to odd powers of the basic hyperbolic rotation $\hat{4}$, consists of hexagonal lattice points having decreasing lattice parameters: a for $\hat{4}$, $a/5$ for $\hat{4}^3$ and $a/19$ for $\hat{4}^5$ (see Fig. 6). These specific ratios appear to characterize the diffraction pattern of the commensurate modulated phase of niobium ditelluride NbTe_2 (van Landuyt, van Tendeloo & Amelinckx, 1974), an intriguing observation whose meaning has still to be understood (see Fig. 7).

A second family consists of hexagons forming a Kagomé lattice and appears with site symmetry $\hat{4}^2$ (multiplicity 3), $\hat{4}^4$ (multiplicity 12) and $\hat{4}^6$ (multiplicity 48), respectively, scaled by a factor of 2 (see Fig. 8).

A third family can be described as consisting of hexagons linked by regular triangles, with multiplicity 6 for site symmetry $\hat{4}^2$, multiplicity 24 and 96 for site symmetry $\hat{4}^4$, each also scaled by a factor of 2 (see Fig. 9).

Still another family forms honeycomb patterns of various sizes and orientations and more complex atomic arrangements within one single orbit are also observed because of missing points which belong to an orbit of lower multiplicity (see Fig. 10).

Such planar arrangements are easily recognizable to occur (in a more or less deformed version) in many inorganic compounds, in particular those which can be described in terms of a close-packed structure (Wells, 1950). One more reason to look now at the multimetrical symmetry of the lattice of an ideal hexagonal close-packed structure.

4.2. Hexagonal three-dimensional multimetrical space groups

The examples given above have, of course, a straightforward generalization to the hexagonal three-dimensional space groups, as the hyperbolic transformation $\hat{4}$ can be considered to be an hyperbolic rotation around the hexagonal axis. We now investigate the possibility of having additional hyperbolic symmetry rotations around other axes. This implies a metrical relation (not present in the Euclidean case) between the a and the c parameter of an hexagonal lattice. Such a relation is well known to occur for lattices of close-packed structures. Here we will restrict our considerations to the lattice A_{hcp} of an ideal hexagonal close-packed structure, for which the c/a ratio is $(8/3)^{1/2}$.

The basis vectors of the lattice A_{hcp} can be chosen as

$$\mathbf{a}_1 = a(1, 0, 0), \quad \mathbf{a}_2 = a(-1/2, 3^{1/2}/2, 0)$$

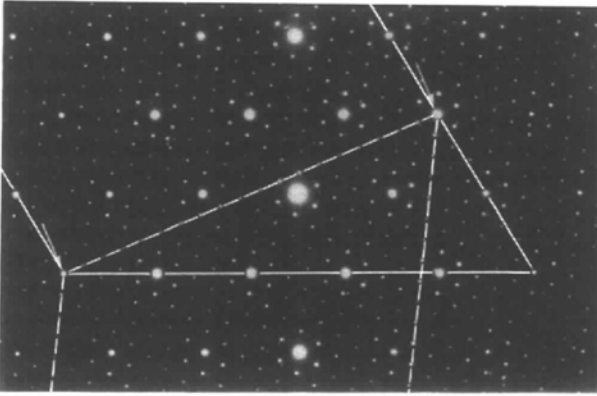


Fig. 7. Diffraction pattern of the commensurately modulated crystal of NbTe₂ (van Landuyt, van Tendeloo & Amelinckx, 1974) with an underlying 5 × 5 and 19 × 19 hexagonal unit cell, to be compared with the Wyckoff sets of equivalent positions of the multimetric space group *p64*, which suggest a kind of multimetric 'star of *k*' structure for the weak satellite reflections.

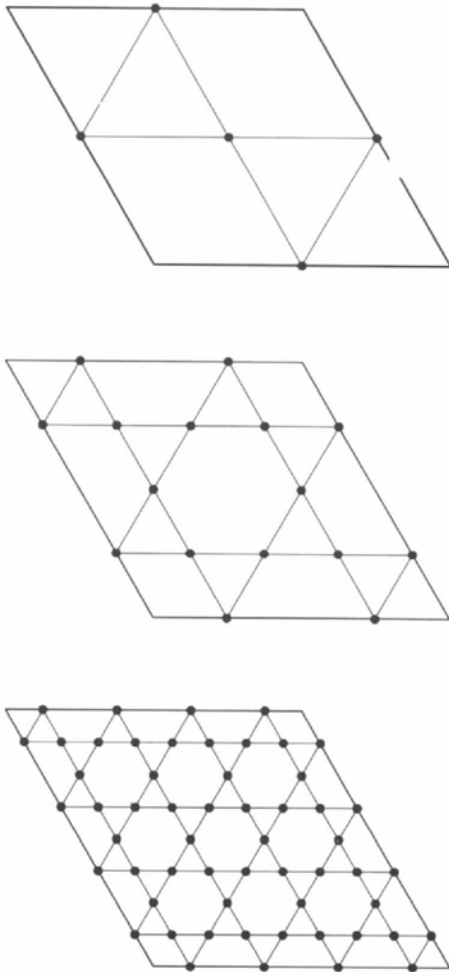


Fig. 8. Kagomé lattices formed by sets of *p64*-equivalent positions. In Table 4 the first two are denoted by (*c*) and by 2(*d*)1, respectively.

and

$$\mathbf{a}_3 = c(0,0,1) = a[0,0,(8/3)^{1/2}]. \quad (4.13)$$

To derive a multimetric point-group symmetry of A_{hcp} (we are at present still unable to obtain the full point symmetry), one considers, in addition to the Euclidean metric tensor g_0 , other pseudo-Euclidean g_μ tensors attached to the same basis

$$\begin{aligned} g_0(\mathbf{e}) &= (1,1,1), \quad g_1(\mathbf{e}) = (\bar{1},1,1), \quad g_2(\mathbf{e}) = (1,\bar{1},1), \\ g_3(\mathbf{e}) &= (1,1,\bar{1}), \end{aligned} \quad (4.14)$$

where $\bar{1}$ is -1 . The notation reflects the choice for the indefinite metric tensor g_j of the direction of the 'pseudo-time' axis, *i.e.* of the unit vector $\boldsymbol{\varepsilon}_0$ of negative squared length: $\boldsymbol{\varepsilon}_0 = \mathbf{e}_j$ for $j \neq 0$.

The scalar products $\mathbf{a}_j \overset{\mu}{\circ} \mathbf{a}_k$ are expressible in terms of the set of metric tensors g_μ given above. As an

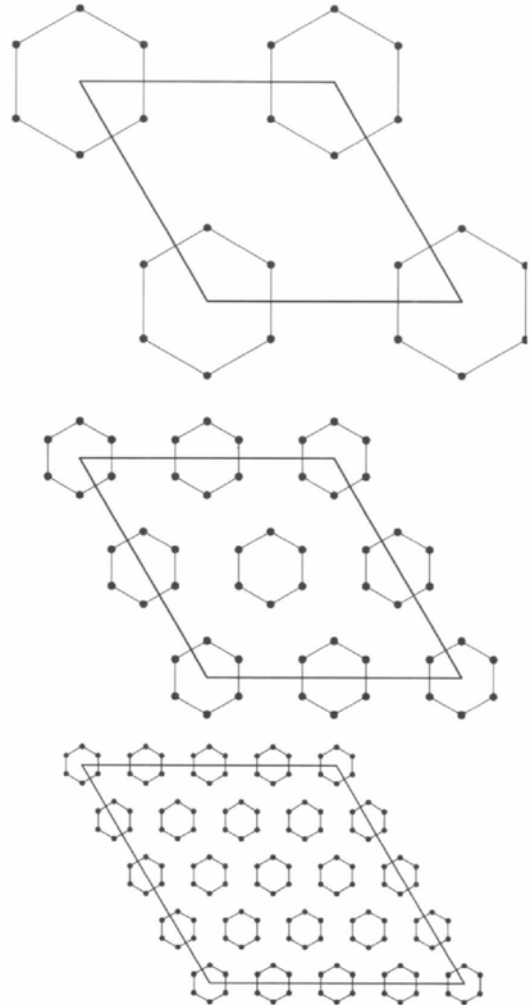


Fig. 9. Families of *p64*-equivalent positions appearing as a lattice of hexagons linked by regular triangles (that link is not shown in the pictures). The first two sets are indicated in Table 4 as Wyckoff positions (*d*)1 and 4(*d*)2, respectively.

illustration we will consider the $g_1(\mathbf{a})$ metric case. One then has $\mathbf{e}_1^2 = -1$, $\mathbf{e}_2^2 = \mathbf{e}_3^2 = 1$ and $\mathbf{e}_i \circ \mathbf{e}_k = 0$ for $i \neq k$. Therefore, one easily derives

$$\begin{aligned} \mathbf{a}_1^2 &= -a^2, \mathbf{a}_2^2 = -1/4a^2 + 3/4a^2 = 1/2a^2, \mathbf{a}_3^2 = 8/3a^2, \\ \mathbf{a}_1 \circ \mathbf{a}_2 &= 1/2a^2, \mathbf{a}_1 \circ \mathbf{a}_3 = 0, \mathbf{a}_2 \circ \mathbf{a}_3 = 0. \end{aligned} \quad (4.15)$$

In this way one finds

$$\begin{aligned} g_0(\mathbf{a}) &= a^2 \begin{pmatrix} 1 & -\frac{1}{2} & 0 \\ -\frac{1}{2} & 1 & 0 \\ 0 & 0 & \frac{8}{3} \end{pmatrix} \\ g_1(\mathbf{a}) &= a^2 \begin{pmatrix} -1 & \frac{1}{2} & 0 \\ \frac{1}{2} & \frac{1}{2} & 0 \\ 0 & 0 & \frac{8}{3} \end{pmatrix} \\ g_2(\mathbf{a}) &= a^2 \begin{pmatrix} 1 & -\frac{1}{2} & 0 \\ -\frac{1}{2} & -\frac{1}{2} & 0 \\ 0 & 0 & \frac{8}{3} \end{pmatrix} \\ g_3(\mathbf{a}) &= a^2 \begin{pmatrix} 1 & -\frac{1}{2} & 0 \\ -\frac{1}{2} & 1 & 0 \\ 0 & 0 & -\frac{8}{3} \end{pmatrix}. \end{aligned} \quad (4.16)$$

We already know that $L_z = \hat{4}$ leaves A_{hcp} and the indefinite metric tensor g_2 invariant (and g_1 also). This same lattice is also invariant by an hyperbolic rotation L_x with $\cosh\varphi = 17$ around the x axis and by another with $\cosh\psi = 49$ around the y axis. Expressing these generators with respect to the lattice basis (\mathbf{a}) one finds

$$\begin{aligned} L_x(\mathbf{a}) &= \begin{pmatrix} 1 & 8 & \overline{16} \\ 0 & 17 & \overline{32} \\ 0 & \overline{9} & 17 \end{pmatrix}, \\ L_y(\mathbf{a}) &= \begin{pmatrix} 49 & \overline{24} & \overline{80} \\ 0 & 1 & 0 \\ \overline{30} & 15 & 49 \end{pmatrix}, \\ L_z(\mathbf{a}) &= \begin{pmatrix} 3 & 1 & 0 \\ 2 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \end{aligned} \quad (4.17)$$

As in the two-dimensional case, all these rotations are generated by hyperbolic reflection symmetries of the lattice A_{hcp} , which involve the mirrors sharing a pseudo-Euclidean metric tensor invariance and the Euclidean one (as discussed in the two-dimensional case). Thus, one arrives at the following multimetri-

cal point group for A_{hcp}

$$\begin{aligned} K &= \{R_z, m_x, m_y, m_z, L_x, L_y, L_z\} \\ &= 6/mmm(34)(98)(4), \end{aligned} \quad (4.18)$$

where now the integral value of $2\cosh\chi$ for the corresponding generator is given in brackets.

At this stage, one can hardly believe that such large-angle hyperbolic transformations do have any structural meaning. A preliminary investigation of (ideal) close-packed structures leads, however, to the conclusion that these additional symmetries are, at least from a geometrical point of view, intimately

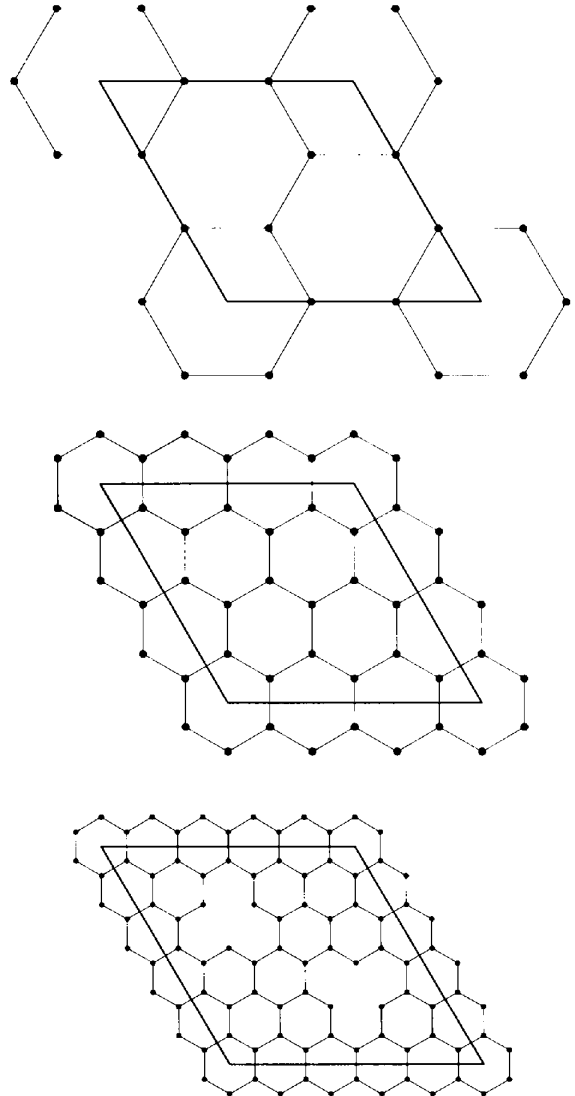


Fig. 10. Families of Wyckoff sets of the multimetrical space group $p6\hat{4}$ forming honeycomb arrangements. The first two correspond to the positions $(d)2$ and $3(d)2$ of Table 4. In the last picture, the missing points belong to another honeycomb orbit with multiplicity 2 [denoted by the Wyckoff letter (b) in the same table].

connected with the structure. In order to present some of these aspects a multimetric space group leaving the Wurtzite structure type invariant will be considered.

4.3. The ideal wurtzite crystal structure: a multimetric analysis

Wurtzite (hexagonal ZnS) has space group $G_0 = P6_3mc$. The observed c/a ratio is 1.6363 (Ulrich & Zachariassen, 1925), whereas that of the ideal closed-packed structure assumed here is $(8/3)^{1/2}$. Occupied are b positions with fractional coordinates $\frac{1}{3}, \frac{2}{3}, z$ and $\frac{2}{3}, \frac{1}{3}, \frac{1}{2} + z$. For the first atom $S = X$ one has $z = 0$, whereas for the second atoms $Zn = Y$ the value is $z = 0.375$.

One then verifies that this structure is left invariant, in addition to the Euclidean generators

$$\begin{aligned} g_1 &= \{R_z | 0, 0, \frac{1}{2}\}, g_2 = \{m_x | 0, 0, \frac{1}{2}\}, \\ g_3 &= \{m_y | 0, 0, 0\}, \end{aligned} \quad (4.19)$$

by the following pseudo-Euclidean ones

$$\begin{aligned} g_4 &= \{L_z | 0, 0, \frac{1}{2}\}, g_5 = \{L_x | 0, 0, \frac{1}{2}\}, \\ g_6 &= \{L_y | 0, 0, 0\}. \end{aligned} \quad (4.20)$$

Indeed, for example

$$L_x Y_1 = \begin{pmatrix} 1 & 8 & \overline{16} \\ 0 & 17 & \overline{32} \\ 0 & \overline{9} & 17 \end{pmatrix} \begin{matrix} \frac{1}{3} \\ \frac{2}{3} \\ 0.375 \end{matrix} = \begin{matrix} \frac{2}{3} + 5 - 6 \\ \frac{1}{3} + 11 - 12 \\ -6 + 6.375 \end{matrix} \sim \begin{matrix} \frac{2}{3} \\ \frac{1}{3} \\ 0.375 \end{matrix}, \quad (4.21)$$

implying $g_5 Y_1 = Y_2$. In the same way one finds that $g_5 Y_2 = Y_1$ and that also X_1 and X_2 are permuted.

Considering now the multimetric space group $G = \{A_{n\phi}, g_1, \dots, g_6\}$, one sees that to obtain a multiplicity-two position, instead of an arbitrary value for the parameter z in the Wyckoff positions (a) and (b), a rational value $z = n/16$ (with given integer n) is required: in that case the site symmetry instead of $3m$ becomes at least $3mL_x^2 L_y, L_z^2$.

This condition is fulfilled not only by the atoms of the close packing ($n = 0$), but also by those at the tetrahedral sites ($n = 6$ and $n = 10$, respectively) and at octahedral sites ($n = 4$). A same situation is found in the case of the other lattices of ideal close-packed structures. Each of these lattices can be obtained by centering of an orthorhombic primitive lattice with parameters $a, b = 2^{1/2}a$ and $c = 3^{1/2}a$. The multimetric extension of $P6_3/mmc$ considered above shares many of the properties of the multimetric extension of the five-dimensional space group $P10_5/mmc$. Actually, the study of the scaling properties of the decagonal $Al_{78}Mn_{22}$ phase was the basis for the multimetric description of the Wurtzite structure (and not the converse).

Considering all that, one becomes motivated to investigate further, trying to understand the still hidden meaning of this new way to look at Euclidean crystal structures. In any case, the additional multimetric symmetries are a way of characterizing Euclidean metric relations not expressible in terms of Euclidean symmetries.

The strength of the present approach is that it leads to the identification of crystallographic symmetries hidden until now which can give rise, in particular, to additional extinctions in the diffraction pattern and it allows the same crystallographic symmetry characterization for crystals and quasicrystals. The weakness is that it may require an idealization of the structure of a given crystal and a discretization of the atomic positions to appropriate rational values.

5. Perspectives

In a somewhat sketchy way and without pretending to have a clear view about future developments, let me indicate some aspects on which I would like to obtain a better insight.

5.1. Metric embedding of modulated structures

For explaining the problem, consider a Fibonacci chain described as a one-dimensional modulated crystal structure. Using the quasicrystal embedding on a square lattice, both \mathbb{Z} -modules M and M^* do have rank 2, whereas using the standard modulated crystal embedding (based on a set of main reflections) M^* has rank 2 but M only rank 1 (and takes into account the properties of the averaged structure). In the first description all atoms of the Fibonacci chain are translationally equivalent, whereas they are not so in the modulated description. Clearly, in order to explore beyond the standard embedding one needs some information on the amplitude of the modulation (information typically obtained from Patterson). Moreover, the form of the modulation wave is also relevant: a squared wave (to be expected near the lock-in transition or in nonstoichiometric crystals) leads more naturally to a metric embedding (for which M and M^* do have the same rank) than when a sinusoidal wave is a good approximation.

5.2. From positional to intensity relations for scaling symmetries

In crystallography the same intensity is required for symmetry-equivalent Bragg reflections. However, this need not to be the case in general. For many symmetries in physics (typically expressed through group representations) it is the ratio between correlated amplitudes which is relevant, not the same value (consider, for example, the Wigner-Eckart theorem). Coming back to Fibonacci again, the same

diffraction intensity can be obtained after deconvolution by the window function, or by averaging to what could represent the main reflections. It seems to me that both ways are available for a better treatment of scaling symmetry in quasicrystals: deconvolution and averaging. The first approach corresponds to deconvolute with respect to a superspace crystal form (as usually done with respect to the shape of a crystal); the second reflects a way of considering scaling symmetry in structures as a process where atoms form clusters which are similar to clusters of those clusters. The approach could then be analogous to that in the theory of 'real-space renormalization', where a new effective Hamiltonian is defined at each stage of a rescaled system. In the crystallographic case this would imply the replacement of the structure factor of a cluster of atoms (with corresponding atomic factors) by a new effective structure factor representing the cluster of one (or more) rescaled pseudo-atom(s).

5.3. Fractal atomic surfaces for quasicrystals

The point of view presented here of an atomic surface as the internal part of a crystal form in superspace, in order to become fully satisfactory needs a better characterization of the possible crystal forms in terms of the symmetries of the (ideal) structure: for example, by an appropriate indexing of the flat pieces of the boundary (like Bravais had done for the crystal growth forms). One already knows that the fractal character of an atomic surface roots in the self-similarity of the underlying quasicrystal structure (Godrèche, Luck, Janner & Janssen, 1993; Luck, Godrèche, Janner & Janssen, 1993; Zobetz, 1992, 1993). However, soon outside the one-dimensional case, the variety and complexity of the possible shapes represent a severe handicap. Possibly a larger experience of the multimetric symmetries of normal crystals could be of great help. As a side remark in this respect as asymptotically an hyperbolic transformation becomes a scaling, if multimetric symmetries are structurally relevant one should be able to see asymptotic consequences in the form of scaling behavior. Indeed, hexagrammatical scaling properties can be recognized in snow crystals. Several examples can be found in the photographic pictures taken by Bentley & Humphreys (1931) as one can see, in particular, on p. 141 (No. 11) and on p. 188 (No. 12) of their book (Bentley & Humphreys, 1931). To find out the precise nature of such structural relations in dendritic crystal forms requires, however, a more detailed investigation.

5.4. Nonindexable Bragg reflections

Nonindexable is a diffuse diffraction intensity which represents a lack of long-range order. How-

ever, that is not necessarily the case for all non-indexable reflections having measurable intensity. One of the reasons is the existence of rotations of infinite order, leaving invariant a \mathbb{Z} -module M of rank larger than the dimension (Le Tu Quoc Thang, Piunikhin & Sadov, 1993). It deserves attention to study possible realizations of a (incommensurate) crystal with such infinite-order rotational symmetries and to consider the algorithm required for the synthesis (at least approximately) of such a structure. One would then eventually obtain nonindexable diffraction rings similar to those observed on powder diffraction, but now for a structure having a long-range order.

5.5. Concluding remarks

One could only scratch the surface of a more comprehensive crystallography. Fortunately, what is missing here can be filled by excellent reviews and a fairly large number of papers devoted to incommensurate crystal structures and to quasicrystals. Only some of these publications could be quoted here and I apologize for that.

Thanks are expressed to Sander Van Smaalen for having put at my disposal the preprint of his valuable review article on incommensurate structures, to Marjorie Senechal for having sent to me the preliminary draft of her book *Quasicrystals and Geometry*, to G. J. Heckman for having drawn my attention to the paper by Le Tu Quoc Thang, Piunikhin and Sadov and to A. H. M. Thiers for his critical remarks. The program developed by him in collaboration with M. J. Eprhaïm helped me to check the existence of additional special reflection conditions in multimetric space groups.

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