given in Table 1 are for an IBM 9021-720 mainframe computer.

The program is in principle available for distribution, but interested parties should consult with the author first.

References

Braun, P. B., Hornstra, J. & Leenhouts, J. I. (1969). *Philips Res. Rep.* **24**, 85–118.

Crowther, R. A. (1972). *The Molecular Replacement Method*, edited by M. G. Rossmann, pp. 173–178. New York: Gordon and Breach.

Crowther, R. A. & Blow, D. M. (1967). *Acta Cryst.* **23**, 544–548.

Fitzgerald, P. M. D. (1988). *J. Appl. Cryst.* **21**, 273–278.

Huber, R. (1965). *Acta Cryst.* **19**, 353–356.

Huber, R. (1985). *Molecular Replacement*. Proc. of the Daresbury Study Weekend, Daresbury, February 1985, pp. 58–61. SERC Daresbury Laboratory, Warrington, England.

Lattman, E. E. & Love, W. E. (1972). *Acta Cryst.* B**26**, 1854–1857.

Nordman, C. E. & Nakatsu, K. (1963). *J. Am Chem. Soc.* **85**, 353.

Rossmann, M. G. & Blow, D. M. (1962). *Acta Cryst.* **15**, 24–31.

Takano, T. (1977). *J. Mol. Biol.* **110**, 537–568.

Ten Eyck, L. F. (1973). *Acta Cryst.* A**29**, 183–191.

Acta Cryst. (1994). A**50**, 72–85

Space Groups of Trigonal and Hexagonal Quasiperiodic Crystals of Rank 4

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Abstract

As a pedagogical illustration of the Fourier-space approach to the crystallography of quasiperiodic crystals, a simple derivation is given of the space-group classification scheme for hexagonal and trigonal quasiperiodic crystals of rank 4. The categories, which can be directly inferred from the Fourier-space forms of the hexagonal and trigonal space groups for periodic crystals, describe general hexagonal or trigonal quasiperiodic crystals of rank 4, which include but are not limited to modulated crystals and intergrowth compounds. When these general categories are applied to the special case of modulated crystals, it is useful to present them in ways that emphasize each of the subsets of Bragg peaks that can serve as distinct lattices of main reflections. These different settings of the general rank-4 space groups correspond precisely to the superspace-group description of $(3+1)$ modulated crystals given by de Wolff, Jannsen & Janner [*Acta Cryst.* (1981), A**37**, 625–636]. As a demonstration of the power of the Fourier-space approach, the space groups for hexagonal and trigonal quasiperiodic crystals of arbitrary finite rank are derived in a companion paper [Lifshitz & Mermin (1994). *Acta Cryst.* A**50**, 85–97].

I. Introduction

Crystals used to be defined as materials periodic on the atomic scale. As such, they were classified by their *space groups* – subgroups of the full Euclidean group that bring a periodic density into coincidence with itself. Because of the growing numbers and varieties of quasiperiodic crystals, crystals have been redefined* as materials whose diffraction patterns contain Bragg peaks, thereby shifting the essential attribute of crystallinity from position space to Fourier space. A corresponding shift in the crystallographic classification scheme, proposed thirty years ago by Bienenstock & Ewald (1962), has not, however, been widely accepted, probably because they advocated Fourier-space crystallography before quasiperiodic crystals had become of major interest, when there was no strong incentive to make the shift. Now there is.

The conventional extension of the classification scheme to quasiperiodic materials, developed and used by de Wolff, Janssen & Janner (1981) (henceforth JJdW)† to find the 'superspace groups' of $(3+1)$ incommensurately modulated crystals, retains the old criterion of periodicity as the starting point for a crystallographic classification scheme and must therefore treat quasiperiodic structures as three-dimensional sections of structures periodic in a higher-dimensional superspace. The need for such a maneuver is avoided by the Fourier-space classifi-

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* Statement of 'terms of reference' of the *ad interim* Commission on Aperiodic Crystals of the International Union of Crystallography.

† See also Yamamoto, Janssen, Janner & de Wolff (1985) and Janssen, Janner, Looijenga-Vos & de Wolff (1992).

cation scheme of Rokhsar, Wright & Mermin (1988*a*,*b*), who extended Bienenstock & Ewald's Fourier-space crystallography of periodic crystals to a symmetry-based classification scheme in three-dimensional Fourier space that grows naturally out of the broader definition of crystals as materials with diffraction patterns of sharp Bragg peaks.

Aside from being more in keeping with the new definition of crystallinity, the Fourier-space approach to the classification of crystals has two main advantages over the conventional superspace approach:

(1) Because periodicity plays no role, there is no need to construct the classification in superspace. The same approach in three dimensions that works for ordinary periodic crystals continues to be applicable to quasiperiodic crystals. The artificiality and nonuniqueness of the superspace embedding is avoided and there is no need to replace the powerful tool of three-dimensional geometric intuition by the algebraic abstractions underlying the extension to higher dimensions of the conventional crystallography of periodic materials.*

(2) Unlike the superspace scheme, the Fourier-space approach does not single out a special three-dimensional lattice of main reflections as a starting point for an extension to higher dimensions. Consequently, Fourier-space crystallography has no built-in bias toward modulated crystals. By eliminating this bias, one achieves a substantially simpler and more broadly applicable classification for arbitrary quasiperiodic crystals, from which the conventional descriptions of the subcategory of modulated crystals can straightforwardly be recovered as special cases, most naturally viewed as settings of the general categories.

The key to the reformulation of crystallography in a manner that embraces quasiperiodic as well as periodic materials is to redefine the *point group* of a material as the subset of operations from $O(3)$ that take the density into one that is *indistinguishable* from what it was. Two densities are said to be indistinguishable if they have the same positionally averaged n-point autocorrelation functions for all n – *i.e.* if any substructure on any scale that occurs in one occurs in the other with the same frequency. From a practical point of view, any two densities related in this way are literally indistinguishable because any finite subregion of one is just as likely to be a subregion of the other and all laboratory specimens do indeed come in finite chunks.

Formally, densities ρ and ρ' are indistinguishable if

$$\langle\rho(\mathbf{r}_1-\mathbf{r})\cdots\rho(\mathbf{r}_n-\mathbf{r})\rangle=\langle\rho'(\mathbf{r}_1-\mathbf{r})\cdots\rho'(\mathbf{r}_n-\mathbf{r})\rangle \quad (1)$$

for all n, where the angular brackets $\langle\,\rangle$ denote a uniform average over all positions $\mathbf{r}$. If a material is periodic, one can prove that indistinguishability reduces to identity to within a translation. One can then combine point-group operations with translations to recover the traditional space groups of periodic materials, containing operations that leave the density *identical* to what it was. But indistinguishable quasiperiodic densities need not be so simply related in real space.* However, the condition (1) of indistinguishability does acquire a very simple form in Fourier space (described below in § II), which applies equally well to periodic or quasiperiodic crystals.

One is thus led naturally to a reformulation of crystallography grounded in three-dimensional Fourier space. The new formulation is based on the distinct sets of phases acquired by the density Fourier coefficients associated with the Bragg peaks under the action of point-group operations and is directly applicable to either periodic or quasiperiodic crystals. In the periodic case, the phases happen to be related to real-space translations $\mathbf{d}$ in the familiar fashion, $2\pi\Phi(\mathbf{k})=\mathbf{k}\cdot\mathbf{d}$, but this plays no role in their determination. In the quasiperiodic case, the phases provide the basis for a generalization of the space-group categories, even though they are no longer related to translations in three-dimensional position space.† The Fourier-space approach was recently used to give a compact derivation of all 230 crystallographic space groups as special cases of a more general derivation of the space groups of icosahedral and axial quasicrystals.‡

Fourier-space crystallography starts with the set of all integral linear combinations of the wave vectors at which the density has nonvanishing Fourier coefficients, as determined by the Bragg peaks. In the periodic case, this set is just the reciprocal lattice and, in the quasiperiodic case, we continue to call it the *lattice*,§ the qualifying 'reciprocal' no longer being necessary since quasiperiodic structures have

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* We stress at the start that we are not arguing that superspace is of no use to crystallographers: it has, for example, proved quite useful in suggesting models for the three-dimensional atomic structure. Our point is only that superspace unnecessarily complicates the construction and application of the crystallographic classification scheme.

* In the conventional terminology of quasiperiodic crystals, indistinguishable densities are said to differ by a translation and/or a 'phason'.

† We follow the conventional practice of using the term 'space groups' for the categories based on point groups and phases, even though they are related to subgroups of $E(3)$ only in the periodic case.

‡ See Rabson, Mermin, Rokhsar & Wright (1991) and Mermin (1992*a*). We refer the reader to the second of these for an exposition of Fourier-space crystallography more detailed than the summary that follows.

§ In the superspace approach to quasiperiodic crystals, the term 'Z module' is used instead.

no direct lattice of real-space translations with which it might be confused. By the *rank* of a lattice we mean the smallest number of wave vectors whose integral linear combinations include all lattice vectors – *i.e.* the smallest number of vectors needed to index the diffraction pattern.

Elsewhere, we have defined and enumerated the Bravais classes of such lattices for the simplest quasiperiodic crystals with conventional crystallographic point groups, namely the rank-6 Bravais classes in the cubic system and the rank-4 Bravais classes in any of the other six crystal systems (Mermin & Lifshitz, 1992). Our classification differs from the JJdW classification of $(3+1)$ (or $3+3$ cubic) incommensurately modulated crystals (Janner, Janssen & de Wolff, 1983; Janssen *et al.*, 1992) in having many fewer Bravais classes. This is because the JJdW classification scheme in $(3+d)$-dimensional superspace is designed for modulated crystals. A three-dimensional sublattice associated with strong Bragg peaks – the lattice of main reflections associated with the unmodulated periodic crystal – plays a special role, the remaining weaker satellite peaks associated with the modulations being accommodated by the additional superspace dimensions. Consequently, a particular lattice of rank bigger than 3 can be associated with more than one Bravais class in the JJdW scheme if that lattice has rank-3 sublattices from distinct rank-3 Bravais classes.

There are two primary reasons not to build the choices of lattices of main reflections into the Bravais classes and space groups. (*a*) By not doing so, we arrive at a *general* classification scheme for diffraction patterns with a crystallographic point-group symmetry spanned by more than three wave vectors, which applies whether or not the material happens to be a modulated crystal. When it *is*, the more detailed categories for modulated crystals can be straightforwardly recovered as different 'settings' of the general ones, depending on which rank-3 sublattice contains the main reflections. (*b*) By not building into the foundations of the scheme this finer distinction, based on features of Bragg-peak intensities unrelated to symmetry, one substantially simplifies the derivation and description of the space groups of quasiperiodic materials, making their relation to the ordinary space groups of three-dimensional periodic crystals quite transparent.

It is our purpose in this paper to illustrate the simplicity of the general scheme. We shall show, as a pedagogical example, how to construct the hexagonal and trigonal rank-4 space groups through a simple examination of the Fourier-space description of the ordinary periodic trigonal and hexagonal space groups. We shall then show explicitly how the classification of modulated crystals emerges from the general case when the general categories are described so as to emphasize a particular rank-3 sublattice of main reflections. We begin, in § II, by summarizing the Fourier-space procedure for the enumeration of space groups, describing the connection between space groups and phases. In § III, we give an elementary derivation of the trigonal and hexagonal rank-4 Bravais classes.* In § IV, we give the Fourier-space forms of the ordinary space groups for periodic crystals in the hexagonal and trigonal systems. In § V, we show how the relevant phases for rank-4 hexagonal or trigonal quasiperiodic crystals can be read off directly from the phases for hexagonal and trigonal periodic crystals. In § VI, we extract from those phases the hexagonal and trigonal rank-4 space groups and show how the conventional categories for modulated crystals, previously derived by JJdW, can be recovered as different settings of those space groups that emphasize different rank-3 sublattices of main reflections. In § VII, we review the merits of the Fourier-space approach in the light of the analysis of the preceding sections. The complete specifications of the hexagonal and trigonal space groups of rank 4, along with their $(3+1)$ settings for modulated crystals, are summarized in Tables 5–7.

In a companion paper (Lifshitz & Mermin, 1994), we demonstrate the power of the Fourier-space approach by enumerating the Bravais classes and space groups for trigonal and hexagonal lattices of arbitrary finite rank.

II. Constructing space groups in Fourier space

The Fourier transform of the condition (1) that the densities ρ and ρ' are indistinguishable gives an equivalent condition that the product of the density Fourier coefficients $\rho(\mathbf{k})$ over any set of wave vectors summing to zero should agree with the corresponding product for ρ'. This in turn leads to the condition that ρ and ρ' are indistinguishable if and only if their density Fourier coefficients are related by†

$$\rho'(\mathbf{k}) = \exp[2\pi i\chi(\mathbf{k})]\rho(\mathbf{k}), \qquad (2)$$

where χ, called a *gauge function*, is linear modulo an integer over the lattice of wave vectors.‡ Indis-

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* This simplifies the more elaborate derivation we gave in Mermin & Lifshitz (1992).

† That $\rho(\mathbf{k})$ and $\rho'(\mathbf{k})$ must differ by at most a phase follows from the identity of the two-point correlation functions; that those phases must be linear on the lattice follows from the indentity of the three-point correlation functions; this is then enough to guarantee the identity of the n-point correlation functions. For details see Mermin (1992*a*).

‡ Linearity on the lattice means only that $\chi(\mathbf{k}_1 - \mathbf{k}_2) \equiv \chi(\mathbf{k}_1) - \chi(\mathbf{k}_2)$ whenever $\mathbf{k}_1$, $\mathbf{k}_2$ and $\mathbf{k}_1 - \mathbf{k}_2$ are in the lattice, where '$\equiv$' indicates equality modulo an integer. Note that only when the rank of the lattice is 3 can one always extend χ to a function linear on *all* of continuous $\mathbf{k}$ space.

tinguishable densities are said to be *gauge-equivalent.**

Because the point group is defined to leave the density indistinguishable, we can associate with each point-group operation g a gauge function $\Phi_g(\mathbf{k})$, called a *phase function*, which relates $\rho(g\mathbf{k})$ and $\rho(\mathbf{k})$:

$$\rho(g\mathbf{k}) = \exp[2\pi i\Phi_g(\mathbf{k})]\rho(\mathbf{k}). \tag{3}$$

The space group of a material in the Fourier-space approach is specified by its point group and its phase functions.

Two sets of phase functions, Φ and Φ', that describe indistinguishable densities ρ and ρ', should clearly be associated with the same space group. It follows from (2) and (3) that two such sets must be related by

$$\Phi'_g(\mathbf{k}) \equiv \Phi_g(\mathbf{k}) + \chi([g-1]\mathbf{k}) \tag{4}$$

for every g in the point group and every $\mathbf{k}$ in the lattice. We call Φ and Φ' *gauge-equivalent phase functions* and (4) a *gauge transformation.* Note that since $\chi(0) \equiv 0$ if $g\mathbf{k} = \mathbf{k}$ then the value of $\Phi_g(\mathbf{k})$ is gauge-invariant.

The major part of enumerating the space groups for a given point group consists of finding the families of gauge-equivalent phase functions. We can specify the families by picking one representative for each one, selected by an appropriate choice of gauge to have a conveniently simple form. Because of the linearity of the phase functions, they need only be given for a set of primitive lattice-generating vectors.† Since $\rho([gh]\mathbf{k}) = \rho(g[h\mathbf{k}])$, it follows from (3) that the phase function for the product of two point-group operations can be constructed out of the phase functions for the individual operations by the rule

$$\Phi_{gh}(\mathbf{k}) \equiv \Phi_g(h\mathbf{k}) + \Phi_h(\mathbf{k}), \tag{5}$$

called the *group-compatibility condition.*

Thus, each family of gauge-equivalent phase functions is entirely specified by a finite set of phases: the values of a representative member of the family, given at a set of primitive‡ lattice-generating vectors and for a set of operations that generate the point group. The point-group generating relations impose, through the group-compatibility condition (5), a set of constraints on these phases that ensures they have a unique value for any point-group operation, independent of how that operation is expressed in terms of the generating operations.* To determine the gauge-equivalence classes of phase functions, we therefore choose a set of primitive generating vectors for the lattice and a set of generators for the point group. We apply the group-compatibility condition to the point-group generating relations to produce a set of constraints on the phases associated with those lattice-generating vectors and point-group generators. With a judicious choice of gauge, we extract from these constraints a unique representative of each of the possible classes of gauge-equivalent phase functions.

The remaining part of the space-group classification is merely a matter of simplifying the bookkeeping by grouping together different gauge-equivalence classes, which ought to be identified on other grounds.† A useful example to keep in mind is the orthorhombic system of periodic crystals, in which space groups that differ only in the roles played by the *a*, *b* or *c* axes are not distinguished, even though their phase functions are not gauge-equivalent.‡ Such further identifications arise when there is an operation s not in the point group of the material that acts linearly on the lattice L of wave-vectors, leaving it invariant, in such a way that $g \to sgs^{-1}$ continues to describe the action of the point group on the lattice. Materials characterized by phase functions

$$\Phi'_g(\mathbf{k}) = \Phi_{sgs^{-1}}(s\mathbf{k}) \tag{6}$$

cannot be sensibly distinguished on grounds of symmetry from those classified by phase functions $\Phi_g(\mathbf{k})$; therefore, Φ' and Φ should be grouped in the same class.

Operations s that *are* in the point group G have precisely this property, but it is an instructive exercise to show directly from (5) that, for such s, Φ' and Φ are already gauge-equivalent. If s is *not* an element of the point group G, then the two classes will not, in general, be gauge-equivalent. In the interests of sim-

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* Nothing deep is intended by this terminology. It is motivated by the resemblance to a gauge transformation in electrodynamics, which affects no physical properties but alters the phase of a quantum-mechanical wave function.

† A smallest set of wave vectors all of whose integral linear combinations give the lattice and nothing else.

‡ We must use primitive lattice-generating vectors because the phase functions may not be defined on a set of nonprimitive generating vectors, at least some of which are not in the lattice.

* These constraints are the generalizations to quasiperiodic materials of the Frobenius congruences in the space-group description of periodic materials.

† Although the grounds for this further identification can be stated quite precisely, whether one chooses to make it or not can be a matter of convention.

‡ Although they are not distinguished as *space groups*, it can be very useful to distinguish them as distinct *settings* of the same space group.

plicity, it is then usually desirable to make a further identification.[*]

In the periodic case, s can be a rotation[†] or an element of $O(3)$ combined with a rescaling of the primitive lattice-generating vectors.[‡] In the quasiperiodic case, s can be an isotropic rescaling of the entire lattrice,[§] an isotropic rescaling of a sublattice[¶] or, in the case of the rank-4 quasiperiodic crystals considered below, independent rescalings of certain lattice-generating vectors. Because rescalings are often (though not always) a part of the transformation s, two classes of gauge-equivalent phase functions that are further identified in this manner are said to be *scale-equivalent*.

The classes of phase functions under gauge equivalence and scale equivalence correspond precisely to the space groups in the periodic case and constitute the extension of the space-group classification scheme to the general quasiperiodic case.

As we shall see below, in the hexagonal and trigonal cases, scale equivalence of the rank-4 gauge-equivalence classes can arise from the existence of two primitive generating vectors with incommensurate components along the direction $\hat{\mathbf{z}}$ of the three- or sixfold axis. There is then a freedom to choose other integrally independent pairs of vectors with components along $\hat{\mathbf{z}}$ as generators, without in any way altering the way in which the point group acts on the generating vectors.[**][††] The operations s relevant to scale equivalence are the linear transformations connecting two such pairs of generating vectors. The sole difference between the space groups for general rank-4 trigonal crystals and their $(3+1)$ settings for modulated crystals stems from one additional convention in the modulated case: that the lattice of main reflections should remain invariant under transformations s used to establish scale equivalence.[*]

III. The hexagonal and trigonal rank-4 Bravais classes

Periodic crystals have two Bravais classes of rank-3 lattices with a unique axis of six- or threefold symmetry. We review the derivation of this fact in the periodic case, since essentially the same argument works for the Bravais classes of rank-4 lattices of quasiperiodic crystals with such axes.

It follows from the existence of a threefold axis that any such rank-3 lattice must contain a two-dimensional ('horizontal') triangular sublattice of vectors perpendicular to the ('vertical') threefold axis. We generate the triangular sublattice with two primitive vectors, **a** and **b**, of equal length, separated by 120°. If the third generating vector **c** is parallel to the threefold axis, we have a lattice of the hexagonal P ('primitive') type with a sixfold axis. The point group of such a lattice is $\frac{6}{m}\frac{2}{m}\frac{2}{m} = 6/\mathrm{mmm}$.[†] We call **c** a *vertical stacking vector* because the P lattice can be viewed as a vertical stacking of horizontal planes containing two-dimensional triangular lattices. If the third generating vector has both a vertical component **c** and a nonzero horizontal component **h**, then the planes of triangular lattices are stacked with a *staggered stacking vector*,[‡]

$$\mathbf{c}_s = \mathbf{c} + \mathbf{h}. \tag{7}$$

Because $\mathbf{c}_s$ must differ from its 120° rotation by a vector in the horizontal sublattice, one easily shows that, to within a horizontal lattice vector, there are just two choices for the horizontal shift:

$$\mathbf{h} = \pm(\tfrac{2}{3}\mathbf{a} + \tfrac{1}{3}\mathbf{b}). \tag{8}$$

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[*] For example, if the point group is noncentrosymmetric, then the inversion is such an operation s. Whether or not one makes the corresponding identification of gauge-equivalence classes corresponds in the periodic case to whether or not one distinguishes between enantiomorphic pairs of space groups, counting 230 or 219 distinct types. In this case, the grounds for not making the further distinction are these: if s is a proper rotation, one can interpolate between the materials with the two different gauge-equivalence classes without at any step altering either the point group G or (in the quasiperiodic case) the rank of the lattice. But, if s is an improper rotation, one cannot.

[†] For example, the space group $P\frac{2}{a}\bar{3}$ is associated with two distinct gauge-equivalence classes. Here, G is a tetrahedral point group and the operation s providing the scale equivalence is a 90° rotation (see Mermin, 1992*a*, Table VIII, p. 23). This is the only example in the rank-3 cubic system of a further scale equivalence between distinct gauge-equivalence classes.

[‡] In the orthorhombic system, s can be a 90° rotation about any of three orthogonal directions combined with an anisotropic rescaling to reproduce the original lattice. This leads us to view as different settings of a single space group such distinct gauge-equivalence classes as *Pamm*, *Pmbm* and *Pmmc*. The orthorhombic system has many such examples. The distinct gauge-equivalence classes making up a single orthorhombic space group are just the different settings of that space group.

[§] As in the case of icosahedral quasicrystals. See Rokshar, Wright & Mermin (1988*b*) or Mermin (1992*a*).

[¶] As in the case of axial quasicrystals. See Rabson, Mermin, Rokhsar & Wright (1991).

[**] Mermin & Lifshitz (1992) called this a *reindexing transformation*.

[††] Such a change of basis can also be viewed as the end point of an interpolating family of operations that independently rescale the z components of both vectors without at any stage altering either the point group or the rank.

[*] The superspace approach does not explicitly distinguish between gauge equivalence and scale equivalence and imposes this additional convention at the start of its analysis. In the Fourier-space approach, the determination of gauge-equivalence classes is simple because it is unencumbered by any such constraints associated with scale equivalence. The further identifications of gauge-equivalence classes on the basis of scale equivalence (with whatever constraints one wishes to impose on the allowed transformations s) are easily applied to the gauge-equivalence classes *after* those classes have been enumerated.

[†] When applied to lattices, the term 'point group' has its older meaning: a set of operations from O(3) that leave the lattice invariant applied about the origin of Fourier space.

[‡] Vertical and staggered stackings of two-dimensional lattices were used by Mermin, Rabson, Rokhsar & Wright (1990) to enumerate the Bravais classes of standard axial quasicrystals.

Table 1. *The two rank-4 hexagonal and trigonal Bravais classes*

The Bravais classes are given with their corresponding point groups and their (3 + 1) settings used to describe modulated crystals. The JJdW symbols for the settings are given in the last column. This table is extended to a larger table by Lifshitz & Mermin (1994), who discuss the case of arbitrary rank and explicitly list all hexagonal and trigonal Bravais classes up to rank 7 together with their settings associated with different choices of rank-3 sublattices of main reflections.

Bravais Class	**Point Group**	**(3+1) Settings**	**JJdW Symbol**
SV $[R+1]$	$\bar{3}m$	R, P	$R\bar{3}m(00\gamma)$, $P\bar{3}1m(\frac{1}{3}\frac{1}{3}\gamma)$
VV $[P+1]$	$6/mmm$	P	$P6/mmm(00\gamma)$

These choices are related by a rotation of the whole lattice by 180° and are therefore equivalent. There is, thus, a unique type of staggered stacking, which gives a lattice of the R ('rhombohedral') type. The horizontal component of $\mathbf{c}_s$ reduces the symmetry of the R lattice to threefold, with point group $\bar{3}\frac{2}{m} = \bar{3}m$.

In the rank-4 quasiperiodic case, just as in the rank-3 periodic case, one establishes that any such lattice must contain a two-dimensional triangular sublattice perpendicular to the threefold axis so we can again use $\mathbf{a}$ and $\mathbf{b}$ as two of the four lattice-generating vectors.* For the lattice to be of rank 4, the two remaining generating vectors must both have nonzero incommensurate components parallel to the threefold axis, so we have two stacking vectors instead of one. As in the rank-3 case, since each stacking vector must differ from its 120° rotation by a vector in the horizontal sublattice, the horizontal component of each can again be taken to be either 0 or $\pm[\frac{2}{3}\mathbf{a} + \frac{1}{3}\mathbf{b}]$. Thus, to enumerate the trigonal and hexagonal rank-4 Bravais classes, we need only examine the different possibilities for the horizontal components of the two stacking vectors.

When both stacking vectors are vertical, we have a vertical-vertical lattice, which is a member of the VV (or V^2) Bravais class.† When one stacking vector is vertical and the other has a nonzero horizontal component, we have a staggered-vertical lattice which is a member of the SV Bravais class.‡ As in the periodic case, the sixfold symmetry of the two-dimensional triangular lattice is reduced to threefold by the existence of a staggered stacking vector. Also as in the periodic case, it does not matter which of the two possible nonzero horizontal components in (8) is taken for the staggered stacking vector.

A third possibility might appear to arise when both stacking vectors are staggered. But, if the two stacking vectors have the same horizontal component $\mathbf{h}$, then we can re-express the lattice in the SV form by replacing one of them by the difference of the two, which is a vertical stacking vector; if the stacking vectors have horizontal components that differ in sign, then we can replace one of the staggered stacking vectors by the sum of the two. Thus, the SS case is equivalent to the SV case and is not a distinct Bravais class of its own.*

The trigonal and hexagonal rank-4 Bravais classes are summarized in Table 1 together with their relation to the JJdW Bravais classes of modulated crystals.

IV. Hexagonal and trigonal rank-3 space groups in Fourier space

We first summarize the point groups and their generators in the hexagonal and trigonal systems and then give the phases that specify the Fourier-space forms of the space groups of ordinary periodic crystals.†

A. The point groups and their generators

These are listed in Table 2. The generators are an n-fold rotation r (where n is six or three), an n-fold

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* When the rank is 5 or higher there can be more than one two-dimensional sublattice.

† This Bravais class is called 'hexagonal $P+1$' by Mermin & Lifshitz (1992), but we use here the name VV that generalizes to the case of arbitrary rank (see Lifshitz & Mermin, 1994). The VV Bravais class corresponds to a unique Bravais class in the JJdW classification of modulated crystals: $P6/mmm(00\gamma)$.

‡ The SV lattice has rank-3 sublattices from the two different rank-3 Bravais classes; one is an R sublattice generated by $\mathbf{a}$, $\mathbf{b}$ and the staggered stacking vector and the other is a P sublattice generated by $\mathbf{a}$, $\mathbf{b}$ and the vertical stacking vector. In Mermin & Lifshitz (1992), this Bravais class is called 'trigonal $R+1$', which is biased towards the first of these settings. We use SV to eliminate this bias and to be compatible with the generalization to arbitrary rank in Lifshitz & Mermin (1994). In the JJdW classification of modulated crystals, the two settings of the SV lattices are treated as distinct Bravais classes: $R\bar{3}m(00\gamma)$ and $P\bar{3}1m(\frac{1}{3}\frac{1}{3}\gamma)$.

* In the JJdW classification of modulated crystals, SS lattices are also in the same Bravais class with SV lattices [taken in the $R\bar{3}m(00\gamma)$ setting], since the JJdW scheme permits the stacking vector describing the modulations to be redefined modulo a primitive vector from the lattice of main reflections (but forbids the stacking vector from the lattice of main reflections to be redefined modulo the stacking vector associated with the modulations).

† The space groups are derived in Fourier space, as part of a more general derivation for axial quasicrystals of the trigonal type (rotational symmetry n a power of an odd prime) or hexagonal type (n twice a power of an odd prime), in Rabson, Mermin, Rokhsar & Wright (1991). The phase functions that specify the Fourier-space forms can also be straightforwardly extracted from the more conventional description in *International Tables for Crystallography* (1992), as noted below.

roto-inversion $\bar{r} = ir$ (where i is the three-dimensional inversion), a horizontal mirror h whose invariant plane is perpendicular to the n-fold axis, a vertical mirror m whose invariant plane includes the n-fold axis and a twofold (dihedral) axis d perpendicular to the n-fold axis.* If a lattice is produced by vertically stacking two-dimensional triangular lattices, it can be associated with point groups which have either six- or threefold rotation symmetry. If a lattice has a staggered stacking vector, the associated point groups can only have threefold rotation symmetry.

B. The hexagonal and trigonal space groups for periodic crystals

Rabson, Mermin, Rokhsar & Wright (1991) (henceforth RMRW) show that there is always a gauge (a choice of real-space origin in the periodic case) in which all phase functions are zero on the two-dimensional triangular sublattice and in which the only possible nonzero phase functions for the point-group generators in Table 2 are those associated with the generators r and m. The classes of phase functions can therefore be specified by at most two phases: the values of Φ_r and Φ_m at the stacking vector. These depend on whether the stacking vector is vertical or staggered and are summarized in Table 3 for a vertical stacking vector (P lattice) and in Table 4 for a staggered stacking vector (R lattice).

Both tables list, for each point group, the possible values of the nontrivial phase functions at the stacking vector. They compactly summarize all the gauge-equivalence classes of phase functions for the hexagonal and trigonal point groups in the periodic case.†

These gauge-equivalence classes correspond precisely (for reasons noted below) to the 52 hexagonal and trigonal space groups for periodic crystals. To recognize that they contain nothing but familiar information, note that in the conventional nomenclature of crystallography the tables simply specify the possibility of having a glide plane ($\Phi_m = \frac{1}{2}$, which turns the m into a c in the space-group symbol) and/or a screw axis ($\Phi_r \neq 0$, which adds an appropriate subscript to the 6 or the 3).

Given the gauge-equivalence classes, we arrive at the space groups themselves by eliminating possible redundancies in description arising from the scale equivalence of distinct gauge-equivalence classes. One can verify from Tables 3 and 4 that the only operations s not in the point group that can leave the lattice invariant and connect different gauge-equivalence classes through (6) are improper operations that can do the trick for some of the three- and sixfold screw axes. If one adopts the convention that improper operations do not make distinct gauge-equivalence classes scale-equivalent,* then the gauge-equivalence classes in Tables 3 and 4 coincide with the space groups, of which there are 27 in the hexagonal system and 25 (18 on the P lattice and 7 on the R lattice) in the trigonal system.†

Table 2. *Point groups of the hexagonal and trigonal systems*

All point groups are compatible with the hexagonal P or $P+1$ (VV) lattices, giving in either case a total of twelve point groups, four of which ($\bar{6}2m$, $\bar{3}m$, $3m$ and 32) can be oriented in two distinct ways on the hexagonal lattice. Only the point groups with threefold symmetry are compatible with the trigonal R or $R+1$ (SV) lattices, giving in either case a total of five point groups, each with a unique orientation.

Generators	**Hexagonal**	**Trigonal**
$\bar{r}$	$\bar{6}$	$\bar{3}$
$\bar{r}, m$	$\bar{6}2m$	$\bar{3}m$
r	6	3
r, m	$6mm$	$3m$
r, d	622	32
r, h	$6/m$	—
r, h, m	$6/mmm$	—

V. Phase functions for the rank-4 Bravais classes

The phase functions for the hexagonal and trigonal rank-4 Bravais classes can be read directly from Tables 3 and 4, which give them for the periodic rank-3 case. This is because both types of rank-4 lattices can be constructed by adding to a rank-3 lattice a fourth primitive generating vector that is vertical and therefore either invariant or changed in

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* On the hexagonal P lattice, the invariant planes of the vertical mirrors or the axes of the twofold rotations can be oriented either along or between vectors of the sixfold star formed by the generating vectors of the triangular horizontal sublattice and their negatives. On the trigonal R lattice, vertical mirrors must be between star vectors and twofold axes must be along them.

† To confirm that distinct phases correspond to distinct gauge-equivalence classes of phase functions, note that in every case the phases are given at a vector that is invariant under the corresponding point-group generator. They are therefore unaltered by a gauge transformation (4).

* This is the convention that takes enantiomorphic pairs of space groups to be distinct, giving 230 space groups for periodic crystals.

† If one allows improper operations to establish scale equivalence, then by taking s to be the inversion i one can establish scale equivalence between the gauge-equivalence classes specified by the phases $\Phi_r(\mathbf{c}) \equiv \frac{1}{3}$ and $\frac{2}{3}$ (and similarly between $\frac{1}{6}$ and $\frac{5}{6}$ and between $\frac{2}{6}$ and $\frac{4}{6}$). Four of the 11 enantiomorphic pairs occur in the hexagonal and three in the trigonal crystal systems.

Table 3. *Phase functions at generating vectors of the rank-3 P lattice with hexagonal or trigonal point groups*

It is enough to specify the phase functions for the point-group generators and their values ('phases') at the primitive lattice-generating vectors. A gauge is used in which all such unspecified phases are zero; the only possible nonzero phases are at the vertical stacking vector **c**. Point groups with identical phase functions are collected together above the same set of entries. The point groups $\bar{6}2m$, $\bar{3}m$, $3m$ and 32 are listed in two forms, associated with the two possible orientations of the twofold axis or vertical mirror. (An m in the second position or a 2 in the third position means the plane of the mirror or the dihedral axis is oriented along the directions of the sixfold star of horizontal reciprocal-lattice-generating vectors and their negatives; an m in the third position or a 2 in the second position means the orientation is between star vectors.) These phases are given in Table IV of RMRW (1991). They can also be extracted directly from *International Tables* by noting that a glide plane corresponds to a value of $\frac{1}{2}$ for the mirror phase function and that an n_j screw axis corresponds to a value $\frac{j}{n}$ for the rotation phase function. The phases are gauge-invariant since $[m-1]\mathbf{c}$ and $[r-1]\mathbf{c}$ are zero. There are 27 hexagonal and 18 trigonal classes of phase functions, corresponding to the same number of space groups if one distinguishes between enantiomorphic pairs.

G	$\bar{6}$ $\bar{3}$	6 622	3 312 321	$\bar{6}2m$ $\bar{3}1m$ $31m$ $\bar{6}m2$ $\bar{3}m1$ $3m1$	$6/m$	$6mm$ $6/mmm$	
Phases	—	$\Phi_r(\mathbf{c})$	$\Phi_r(\mathbf{c})$	$\Phi_m(\mathbf{c})$	$\Phi_r(\mathbf{c})$	$\Phi_r(\mathbf{c})$	$\Phi_m(\mathbf{c})$
Gauge Equivalence Classes		0	0	0	0	0	0
		$\frac{1}{6}$	$\frac{1}{3}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$
		$\frac{2}{6}$	$\frac{2}{3}$				
		$\frac{3}{6}$					
		$\frac{4}{6}$					
		$\frac{5}{6}$					

sign by every point-group operation.* This reduces the determination of the rank-4 gauge-equivalence classes to a trivial exercise, for the following reasons:

(1) We require the phase functions at four primitive generating vectors. We take the fourth to be a vertical stacking vector $\mathbf{c}'$ and the first three to be **a**, **b** and either a vertical stacking vector **c** or a staggered stacking vector $\mathbf{c}_s$. The first three together generate a rank-3 hexagonal or trigonal lattice and the phase functions at those three vectors are constrained by exactly the same conditions that apply in the rank-3 case, since no point-group operations mix those three with the fourth vector $\mathbf{c}'$. Therefore, the phases at the first three vectors can be specified by exactly the same entries that appear in Table 3 (for the vertical case) or Table 4 (for the staggered case).

(2) Because the fourth vector $\mathbf{c}'$ is either invariant or changes sign under every point-group operation, no point-group operation mixes it with any of the first three generating vectors. Therefore, application to the phase functions at $\mathbf{c}'$ of the constraints coming from the group-compatibility condition (5) and the group generating relations yields conditions in which only phase functions at $\mathbf{c}'$ appear. Furthermore, any gauge transformations applied to those phase functions at $\mathbf{c}'$ will only depend on the gauge degree of freedom associated with the choice of $\chi(\mathbf{c}')$, which was not used at all in specifying the phases at the first three vectors. *Determining the additional phases at* $\mathbf{c}'$ *is therefore entirely independent of determining them at the first three generating vectors.*

Table 4. *Phase functions at generating vectors of the rank-3 R lattice with trigonal point groups*

A gauge is used in which all unspecified phases are zero. The possible nonzero phases are at a staggered stacking vector $\mathbf{c}_s = \mathbf{c} + \mathbf{h}$. These results are given in Table VI of RMRW (1991). They can also be extracted from *International Tables*, if one notes that a glide plane corresponds to a value of $\frac{1}{2}$ for the mirror phase function. The phases associated with m are gauge-invariant since $[m-1]\mathbf{c}_s = 0$. There are seven classes of phase function, corresponding to seven trigonal space groups on the rank-3 R lattice.

G	$\bar{3}$	3 32	$\bar{3}m$ $3m$
Phases	—	—	$\Phi_m(\mathbf{c}_s)$
Gauge Equivalence Classes			0
			$\frac{1}{2}$

(3) Point (2) also applies to the phase functions at **c** in the rank-3 hexagonal P lattice: they too are determined in a manner completely independent of how the phase functions are chosen at **a** and **b**. Therefore, the procedure determining the phase functions at $\mathbf{c}'$ in the rank-4 case is identical to the procedure determining the phase functions at **c** for the rank-3 hexagonal lattice, so the rank-4 phases at $\mathbf{c}'$ are identical to the entries in Table 3 for the rank-3 phases at **c**.*

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* This simplification is obscured if one views the SV lattice in its P setting [JJdW category $P\bar{3}1m(\frac{1}{3}\frac{1}{3}\gamma)$], but is obvious from the point of view of the R setting [JJdW category $R\bar{3}m(00\gamma)$]. The information we acquire can, of course, be expressed in either setting, if we wish to apply it to modulated crystals.

* The procedure is so simple that it can easily be applied directly to the phases at $\mathbf{c}'$, but since the resulting information is already in Table 3 it is most efficient just to read it off.

Table 5. *Gauge-equivalence classes and space groups of rank* 4 *in the hexagonal system and their settings for modulated crystals*

The gauge-equivalence classes are given in part I of the table. They are specified by a set of 'phases': the values of a set of phase functions, one for each point-group generator, at the primitive generating vectors of the lattice. A gauge is used in which all phases unspecified in the table are zero. The possible nonzero phases are only at the vertical stacking vectors **c** and **c**′ and only associated with the sixfold rotation *r* or the vertical mirror *m*. As discussed in § V, this part of the table is constructed by simply repeating twice (once for **c** and once for **c**′) the information from Table 3, which gives the gauge-equivalence classes for the corresponding rank-3 periodic crystals. There are a total of 117 hexagonal rank-4 gauge-equivalence classes: one for the point group $\bar{6}$, 36 (6^2) for each of the point groups 6 and 622, four (2^2) for the point group $\bar{6}2m$ in each of its two settings and for the point group 6/*m*, and 16 (2^4) for each of the point groups 6*mm* and 6/*mmm*.

Part II of the table lists the rank-4 space groups arrived at by identifying scale-equivalent gauge-equivalence classes as described in § VI. Phases characterizing a given space group are on a horizontal row, enclosed in brackets. (The absence of such brackets in part I of the table indicates that any selection of phases, one from each column, gives a distinct gauge-equivalence class.) In all but the fifth case in the right-hand column, the phases at the second stacking vector, **c**′, can be taken to be zero. There are a total of 25 rank-4 space groups in the hexagonal system.

Part III of the table lists the different settings of the space groups in the modulated case, where one singles out the rank-3 sublattice of main reflections. We take **c** to be the generator of the lattice of main reflections and **c**′ to describe a satellite peak. The settings are separated vertically into sets that correspond to settings of the general space groups listed in the same order directly above. The 25 rank-4 general space groups subdivide into 54 settings, which correspond to the 54 JJdW (3 + 1) hexagonal superspace groups. (See Janssen *et al.*, 1992, pp. 823–824.) The JJdW symbols for these settings are constructed by taking the letter *P* – indicating a *P* sublattice of main reflections – followed by the point group, where (as in the usual notation for periodic crystals) the letter *m* is changed to *c* if $\Phi_m(\mathbf{c}) = 1/2$ and the 6 acquires a subscript *j* if $\Phi_r(\mathbf{c}) = j/6$. This is followed by (00γ) – indicating a vertical (as opposed to staggered) generator for the satellite peaks – followed by the phases at **c**′ associated with each of the point-group generators appearing in the point-group symbol (in the order they appear in that symbol), where *s* means 1/2, *t* means 1/3 and *h* means 1/6. For example, the three settings of the fifth space group for point group 6/*mmm* (the group of three entries in the extreme lower right of the table) have the JJdW symbols $P6/mmc(00\gamma)s000$, $P6_3/mmm(00\gamma)000s$ and $P6_3/mmc(00\gamma)s000$.

G	$\bar{6}$	6 622		$\bar{6}2m$ $\bar{6}m2$		6/*m*		6*mm* 6/*mmm*			
Phases	—	$\Phi_r(\mathbf{c})$	$\Phi_r(\mathbf{c}')$	$\Phi_m(\mathbf{c})$	$\Phi_m(\mathbf{c}')$	$\Phi_r(\mathbf{c})$	$\Phi_r(\mathbf{c}')$	$\Phi_r(\mathbf{c})$	$\Phi_m(\mathbf{c})$	$\Phi_r(\mathbf{c}')$	$\Phi_m(\mathbf{c}')$
I. Gauge Equivalence Classes		0	0	0	0	0	0	0	0	0	0
		$\frac{1}{6}$	$\frac{1}{6}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$
		$\frac{2}{6}$	$\frac{2}{6}$								
		$\frac{3}{6}$	$\frac{3}{6}$								
		$\frac{4}{6}$	$\frac{4}{6}$								
		$\frac{5}{6}$	$\frac{5}{6}$								
II. General Rank-4 Space Groups		[0	0]	[0	0]	[0	0]	[0	0	0	0]
		[$\frac{1}{6}$	0]	[$\frac{1}{2}$	0]	[$\frac{1}{2}$	0]	[$\frac{1}{2}$	0	0	0]
		[$\frac{2}{6}$	0]					[0	$\frac{1}{2}$	0	0]
		[$\frac{3}{6}$	0]					[$\frac{1}{2}$	$\frac{1}{2}$	0	0]
								[0	$\frac{1}{2}$	$\frac{1}{2}$	0]
III. Settings of General Space Groups for Modulated Crystals		[0	0]	[0	0]	[0	0]	[0	0	0	0]
		[$\frac{1}{6}$	0]	[$\frac{1}{2}$	0]	[$\frac{1}{2}$	0]	[$\frac{1}{2}$	0	0	0]
		[$\frac{2}{6}$	$\frac{1}{6}$]	[0	$\frac{1}{2}$]	[0	$\frac{1}{2}$]	[0	0	$\frac{1}{2}$	0]
		[$\frac{3}{6}$	$\frac{1}{6}$]								
		[$\frac{4}{6}$	$\frac{1}{6}$]					[0	$\frac{1}{2}$	0	0]
		[$\frac{5}{6}$	0]					[0	0	0	$\frac{1}{2}$]
		[0	$\frac{1}{6}$]								
								[$\frac{1}{2}$	$\frac{1}{2}$	0	0]
		[$\frac{2}{6}$	0]					[0	0	$\frac{1}{2}$	$\frac{1}{2}$]
		[$\frac{4}{6}$	0]								
		[0	$\frac{2}{6}$]					[0	$\frac{1}{2}$	$\frac{1}{2}$	0]
								[$\frac{1}{2}$	0	0	$\frac{1}{2}$]
		[$\frac{3}{6}$	0]					[$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	0]
		[0	$\frac{3}{6}$]								

Table 6. *Gauge-equivalence classes and space groups of rank 4 in the trigonal system for the VV (P+1) Bravais class and their settings for modulated crystals*

The structure and conventions are the same as for Table 5. Part I, taken directly from Table 3, lists 44 gauge-equivalence classes: one for the point group $\bar{3}$, nine (3^2) for the point group 3 and the point group 32 in each of its two orientations and four (2^2) for each of the two orientations of each of the two point groups $\bar{3}m$ and $3m$. Part II lists the 15 space groups to which these gauge-equivalence classes reduce under scale equivalence. In all of these, the phases of the second stacking vector, $\mathbf{c}'$, can be taken to be zero. Part III of the table lists the 25 different settings for these space groups in the modulated case. The JJdW superspace symbols are constructed in the same way as specified for the hexagonal space groups in the caption of Table 5.

G	$\bar{3}$	3	321 312	$\bar{3}m1$ $\bar{3}1m$	$3m1$ $31m$
Phases	—	$\Phi_r(\mathbf{c})$	$\Phi_r(\mathbf{c}')$	$\Phi_m(\mathbf{c})$	$\Phi_m(\mathbf{c}')$
I. Gauge–Equivalence Classes		0	0	0	0
		$\frac{1}{3}$	$\frac{1}{3}$	$\frac{1}{2}$	$\frac{1}{2}$
		$\frac{2}{3}$	$\frac{2}{3}$		
II. General Rank-4 Space Groups		[0	0]	[0	0]
		[$\frac{1}{3}$	0]	[$\frac{1}{2}$	0]
III. Settings of General Space Groups for Modulated Crystals		[0	0]	[0	0]
		[$\frac{1}{3}$	0]	[$\frac{1}{2}$	0]
		[$\frac{2}{3}$	0]	[0	$\frac{1}{2}$]
		[0	$\frac{1}{3}$]		

Table 7. *Gauge-equivalence classes and space groups of rank-4 in the trigonal system for the SV (R+1) Bravais class and their settings for modulated crystals*

The structure and conventions are the same as for Table 5, except that (1) the settings for modulated crystals occupy two sections because the *SV* lattice itself has two possible settings and (2) the entries in the upper part of the table for the phases at the generating vector $\mathbf{c}_s$ are now the phases for the ordinary rank-3 space groups on the *R* lattice, taken from Table 4. Part I of the table lists 15 gauge-equivalence classes: one for the point group $\bar{3}$, three for each of the point groups 3 and 32 and four (2^2) for each of the two point groups $\bar{3}m$ and $3m$. Part II lists the nine space groups to which these gauge-equivalence classes reduce under scale equivalence. In all of these, the phases of the second stacking vector, $\mathbf{c}'$, can be taken to be zero. Part III of the table lists the 11 different settings for these space groups in the modulated case when the lattice of main reflections is rhombohedral (*R* settings) and part IV lists the 13 different settings in the modulated case when the lattice of main reflections is primitive (*P* settings). The numbers differ because in the *P* setting the scale-equivalence transformations are required to preserve the rank-3 *P* sublattice and one therefore loses the freedom to rescale the $\mathbf{c}'$ axis so as to identify the two enantiomorphic pairs of screw axes. The JJdW superspace symbols are constructed in a manner similar to that specified for the hexagonal space groups in the caption of Table 5, taking $R\bar{3}m(00\gamma)$ for the underlying symbol in the *R* settings and $P\bar{3}1m(\frac{1}{3}\frac{1}{3}\gamma)$ in the *P* settings.

G	$\bar{3}$	3 32	$\bar{3}m$	$3m$
Phases	—	$\Phi_r(\mathbf{c}')$	$\Phi_m(\mathbf{c}_s)$	$\Phi_m(\mathbf{c}')$
I. Gauge–Equivalence Classes		0	0	0
		$\frac{1}{3}$	$\frac{1}{2}$	$\frac{1}{2}$
		$\frac{2}{3}$		
II. General Rank-4 Space Groups		0	[0	0]
		$\frac{1}{3}$	[$\frac{1}{2}$	0]
III. *R* Settings of General Space Groups for Modulated Crystals		0	[0	0]
		$\frac{1}{3}$	[$\frac{1}{2}$	0]
			[0	$\frac{1}{2}$]
IV. *P* Settings of General Space Groups for Modulated Crystals		0	[0	0]
		$\frac{1}{3}$	[$\frac{1}{2}$	0]
		$\frac{2}{3}$	[0	$\frac{1}{2}$]

These considerations enable us to read the possible values of the phase functions and hence the gauge-equivalence classes for both of the rank-4 Bravais classes directly from Tables 3 and 4 of phase functions for the ordinary rank-3 periodic crystals. For the lattices in the *VV* (*P*+1) Bravais class, we simply enter the information from Table 3 twice, having a column for the values of the nontrivial phase functions at the first stacking vector **c** and another identical column repeating the same values that can independently be chosen for the phase functions at the second stacking vector, $\mathbf{c}'$. This is the entire content of parts I of Tables 5 and 6.

For the lattices in the *SV* (*R*+1) Bravais class, we enter the information from Table 4 for the possible nonzero phases at the staggered stacking vector $\mathbf{c}_s$ and the information from Table 3 (for the point groups with threefold symmetry) for the possible nonzero phases at the vertical stacking vector $\mathbf{c}'$. This is the entire content of part I of Table 7.

Parts I of Tables 5–7 give all gauge-equivalence classes of phase functions for the rank-4 hexagonal and trigonal systems. To determine the rank-4 hexagonal and trigonal space groups, it remains only to reduce the redundancy of this information by determining which of these classes are scale-equivalent.

VI. The hexagonal and trigonal rank-4 space groups

The further identification of gauge-equivalence classes on the basis of their scale equivalence is to some extent a matter of convention,[*] as already noted in the periodic case, where enantiomorphic space groups are identified only if improper transformations s are allowed in the relation (6) between scale-equivalent phase functions. We maintain the traditional distinction between enantiomorphic pairs in the rank-3 case by requiring the transformations s to be sense preserving.[†]

In § VI.*A* we describe the general form of the transformations s leading to the scale equivalence of many of the gauge-equivalence classes in Tables 5–7. In § VI.*B* we describe how the forms of these general transformations are restricted if one requires that they preserve a rank-3 sublattice, as is useful in describing modulated crystals. This leads to different settings of the general space groups that correspond precisely to the JJdW 'superspace groups'. In § VI.*C* we describe in detail how the general transformations of § VI.*A* lead to the general space groups (given in parts II of Tables 5–7) and how the restrictions on those transformations in § VI.*B* lead to the different settings of those space groups for modulated crystals (given in parts III of Tables 5 and 6 and parts III and IV of Table 7).

A. *Scale equivalence in the general case*

VV lattice. We take the primitive vectors $\mathbf{c}$ and $\mathbf{c}'$ each to point in the direction of advance of a right-handed screw, rotating through 120° from $\mathbf{a}$ to $\mathbf{b}$. If

$$\begin{bmatrix} t & u \\ v & w \end{bmatrix} \tag{9}$$

is a matrix of integers with determinant[‡] ± 1 that preserves the sense of

$$\begin{aligned} \bar{\mathbf{c}} &= t\mathbf{c} + u\mathbf{c}', \\ \overline{\mathbf{c}'} &= v\mathbf{c} + w\mathbf{c}', \end{aligned} \tag{10}$$

then $\bar{\mathbf{c}}$ and $\overline{\mathbf{c}'}$ constitute an entirely equivalent pair of primitive lattice-generating vectors. The orientation-preserving transformation s that scales $\mathbf{c}$ into $\bar{\mathbf{c}}$ and $\mathbf{c}'$ into $\overline{\mathbf{c}'}$ therefore leaves the lattice invariant. It induces the identity transformation on the point group of the lattice and can therefore lead to a scale equivalence of distinct gauge-equivalence classes. The transformation (6) on the phases is

$$\begin{aligned} \Phi'_g(\bar{\mathbf{c}}) &= t\Phi_g(\mathbf{c}) + u\Phi_g(\mathbf{c}'), \\ \Phi'_g(\overline{\mathbf{c}'}) &= v\Phi_g(\mathbf{c}) + w\Phi_g(\mathbf{c}'). \end{aligned} \tag{11}$$

Two gauge-equivalence classes specified by sets of phases related by (11) are scale-equivalent and therefore specify the same space group.

SV lattice. We take the same sense convention for $\mathbf{c}'$ and the vertical part of $\mathbf{c}_s$ and consider the sense-preserving transformation of primitive stacking vectors $\mathbf{c}$ and $\mathbf{c}_s$ given by

$$\begin{aligned} \bar{\mathbf{c}}_s &= t\mathbf{c}_s + u\mathbf{c}' - (t-1)\mathbf{h}, \\ \overline{\mathbf{c}'} &= v\mathbf{c}_s + w\mathbf{c}' - v\mathbf{h}, \end{aligned} \tag{12}$$

where the matrix of integers (9) again has determinant ± 1 and where v and $t-1$ are multiples of 3.[*] These restrictions on v and t ensure that the new pair of stacking vectors can be obtained from the old pair by an orientation-preserving transformation that scales $\mathbf{c}'$ and the vertical part of $\mathbf{c}_s$ into $\overline{\mathbf{c}'}$ and the vertical part of $\bar{\mathbf{c}}_s$. Such a rescaling leaves the lattice invariant and induces the identity transformation on the point group of the lattice. Since $3\mathbf{h}$ is a vector of the horizontal sublattice and since all phase functions vanish in the horizontal plane, the terms in $\mathbf{h}$ in (12) drop out of the condition (6) for scale equivalence and we have, as in the case of the *VV* lattice,

$$\begin{aligned} \Phi'_g(\bar{\mathbf{c}}_s) &= t\Phi_g(\mathbf{c}_s) + u\Phi_g(\mathbf{c}'), \\ \Phi'_g(\overline{\mathbf{c}'}) &= v\Phi_g(\mathbf{c}_s) + w\Phi_g(\mathbf{c}'), \end{aligned} \tag{13}$$

with the additional restriction that v and $t-1$ must be multiples of 3.

B. *Restricted scale equivalence for modulated crystals*

To further subdivide the general space groups into the conventional categories of modulated crystals, we need only note that in the modulated case it is convenient and conventional to include among the lattice-generating vectors a set of three that generate the sublattice of main reflections and to restrict the transformations that establish scale equivalence to those that take this sublattice into itself. In the case of the *VV* lattice, we choose $\mathbf{c}$ to generate the lattice of main reflections together with $\mathbf{a}$ and $\mathbf{b}$. As a result, the matrices (9) are acceptable candidates for scale equivalence in (11) only if they are of the restricted form

$$\begin{bmatrix} t & u \\ v & w \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ v & \pm 1 \end{bmatrix}. \tag{14}$$

In the case of the *SV* lattice, the restriction on (12) depends on whether the *R* or *P* rank-3 sublattice is

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[*] The convention we adopt is to require there to be a family of structures, all with the same rank and point group, that interpolates between structures belonging to the original and transformed gauge-equivalence classes.

[†] We shall find, however, that in the rank-4 case some distinctions between 6_j (or 3_j) screw axes with different values of j can still be lost, just as in the icosahedral case – see Rokhsar, Wright & Mermin (1988*b*).

[‡] To ensure that its inverse is also a matrix of integers.

[*] Note that $3\mathbf{h}$ is a vector of the horizontal sublattice [see (8)].

taken as the lattice of main reflections. In the former case, it is $\mathbf{c}_s$ that must be preserved and (14) again applies (with the restriction that v is a multiple of 3). In the latter case, $\mathbf{c}'$ must be preserved and we have

$$\begin{bmatrix} t & u \\ v & w \end{bmatrix} = \begin{bmatrix} \pm 1 & u \\ 0 & 1 \end{bmatrix}. \qquad (15)$$

C. Space groups and their settings for modulated crystals

1. *VV Bravais class*

Case (1). *Point groups allowing a glide plane or a twofold screw axis, but not both* ($\bar{6}2m$, $\bar{6}m2$, $6/m$, $\bar{3}1m$, $\bar{3}m1$, $31m$, $3m1$). These are the cases in Tables 5 and 6 for which parts I of the tables give four gauge-equivalence classes of phase functions:

$$\begin{bmatrix} \Phi_g(\mathbf{c}) \\ \Phi_g(\mathbf{c}') \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \end{bmatrix}, \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}, \begin{bmatrix} 0 \\ \frac{1}{2} \end{bmatrix} \text{ or } \begin{bmatrix} \frac{1}{2} \\ \frac{1}{2} \end{bmatrix}, \qquad (16)$$

where the point-group generator g is either m or r. That the third and fourth classes are scale-equivalent to the second is established by noting that

$$\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix} \begin{bmatrix} 0 \\ \frac{1}{2} \end{bmatrix} \equiv \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}; \quad \begin{bmatrix} 1 & 0 \\ 1 & 1 \end{bmatrix} \begin{bmatrix} \frac{1}{2} \\ \frac{1}{2} \end{bmatrix} \equiv \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}. \qquad (17)$$

Thus, for each point group, there is just one nonsymmorphic space group, as indicated by the corresponding entries in parts II of Tables 5 and 6.

To identify the settings of the general space groups appropriate to the modulated case, we must restrict the matrices in (17) to those of the form (14). The first identification in (17) is then prohibited, giving two distinct settings for the nonsymmorphic space group, as given in parts III of Tables 5 and 6.

Case (2). *Point groups allowing both a glide plane and a twofold screw axis* ($6mm$, $6/mmm$). We can now make the choice (16) independently both for Φ_m and Φ_r, giving 16 distinct gauge-equivalence classes. If (*a*) both mirror phases are zero, (*b*) both rotational phases are zero or (*c*) the mirror and rotational phases are the same, then we can repeat the treatment in case (1): the three nontrivial gauge-equivalence classes in each of the cases (*a*)–(*c*) give a single nonsymmorphic space group for each point group, given by the second, third and fourth entries in the right-hand section of part II of Table 5. These each have two settings in the modulated case, as indicated by the corresponding entries in part III.

This leaves six gauge-equivalence classes in which both phase functions are nonzero but Φ_m is not identical to Φ_r. All six, however, are easily shown to be scale equivalent, under transformations of the form (17), to the single class*

$$\begin{bmatrix} \Phi_r(\mathbf{c}) \\ \Phi_r(\mathbf{c}') \end{bmatrix} = \begin{bmatrix} 0 \\ \frac{1}{2} \end{bmatrix}, \quad \begin{bmatrix} \Phi_m(\mathbf{c}) \\ \Phi_m(\mathbf{c}') \end{bmatrix} = \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}, \qquad (18)$$

so there is only one additional space group for each point group, listed as a fifth entry in part II of Table 5.

These space groups each have three settings in the modulated case, where the matrices establishing scale equivalence are restricted to the form (14), which limits the identifications to those that can be realized with the matrix $\begin{bmatrix} 1 & 0 \\ 1 & 1 \end{bmatrix}$. These are given in the fifth entry of part III of Table 5.

Case (3). *Point groups allowing a sixfold or threefold screw axis* (6, 622, 3, 321, 312). For each point group, the phase function Φ_r can have the values $0, \frac{1}{n}, \ldots, \frac{n-1}{n}$ independently for each of the stacking vectors, giving 36 gauge-equivalence classes of phase functions when $n = 6$ and 9 classes when $n = 3$.

When $n = 6$, it can be shown that the 36 classes reduce to four under scale equivalence: (1) the single symmorphic gauge-equivalence class; (2) any of the three nontrivial gauge-equivalence classes in which both phases are integral multiples of $\frac{1}{2}$; (3) any of the eight nontrivial gauge-equivalence classes in which both phases are integral multiples of $\frac{1}{3}$; (4) any of the 24 remaining gauge-equivalence classes. This follows from the fact that, if positive integers t and v are relatively prime, then positive integers u and w can be found such that $tw - uv = 1$. Consequently,

$$\begin{bmatrix} 1 \\ 0 \end{bmatrix} \text{ and } \begin{bmatrix} t \\ v \end{bmatrix} = \begin{bmatrix} t & u \\ v & w \end{bmatrix} \begin{bmatrix} 1 \\ 0 \end{bmatrix} \qquad (19)$$

are related by a transformation of the form (9) that establishes scale equivalence. Since categories (2)–(4) have phases that are $\frac{1}{2}$, $\frac{1}{3}$ or $\frac{1}{6}$ times a column vector with two relatively prime integers, we can represent the four space-group categories by

$$\begin{bmatrix} \Phi_r(\mathbf{c}) \\ \Phi_r(\mathbf{c}') \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \end{bmatrix}, \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}, \begin{bmatrix} \frac{1}{3} \\ 0 \end{bmatrix} \text{ or } \begin{bmatrix} \frac{1}{6} \\ 0 \end{bmatrix}, \qquad (20)$$

as noted in part II of Table 5.

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* The matrix $\begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}$ interchanges the $\mathbf{c}$ and $\mathbf{c}'$ phases, reducing the number of classes to 3. The matrix $\begin{bmatrix} 1 & 1 \\ 0 & 1 \end{bmatrix}$ then takes $\begin{bmatrix} \frac{1}{2} \\ \frac{1}{2} \end{bmatrix}, \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}$ into $\begin{bmatrix} 0 \\ \frac{1}{2} \end{bmatrix}, \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}$ and the matrix $\begin{bmatrix} 1 & 0 \\ 1 & 1 \end{bmatrix}$ takes $\begin{bmatrix} 0 \\ \frac{1}{2} \end{bmatrix}, \begin{bmatrix} \frac{1}{2} \\ \frac{1}{2} \end{bmatrix}$ into $\begin{bmatrix} 0 \\ \frac{1}{2} \end{bmatrix}, \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}$.

To find the settings appropriate to the modulated case, we prohibit identifications that change the value of $\Phi_r(\mathbf{c})$. This results in two settings in case (2) $[\Phi_r(\mathbf{c}) \equiv 0, \frac{1}{2}]$, three settings in case (3) $[\Phi_r(\mathbf{c}) \equiv 0, \frac{1}{3}, \frac{2}{3}]$ and six settings in case (4) $[\Phi_r(\mathbf{c}) \equiv 0, \frac{1}{6}, \ldots, \frac{5}{6}]$. These are listed with the values of $\Phi_r(\mathbf{c}')$ in part III of Table 5.

When $n = 3$, in just the same way, all eight of the nine nontrivial gauge-equivalence classes are scale equivalent, but they have three settings in the modulated case, as shown in Table 6.

Note that in the general case the existence of two incommensurate c axes results in the inability to distinguish between enantiomorphic pairs of screw axes. A similar simplification associated with fivefold screw axes in icosahedral quasicrystals was pointed out in Rokhsar, Wright & Mermin (1988*b*).

2. *SV Bravais class*

In establishing scale equivalence of the nonsymmorphic space groups, we are now restricted to matrices (9) in which v and $t-1$ must be multiples of 3.

Case (1). *Point groups allowing a glide plane* ($\bar{3}m$, $3m$). From part I of Table 7, we see that, for each point group, there are four gauge-equivalence classes,

$$\begin{bmatrix} \Phi_m(\mathbf{c}_s) \\ \Phi_m(\mathbf{c}') \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \end{bmatrix}, \begin{bmatrix} 0 \\ \frac{1}{2} \end{bmatrix}, \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix} \text{ or } \begin{bmatrix} \frac{1}{2} \\ \frac{1}{2} \end{bmatrix}. \tag{21}$$

Scale equivalence between the three nontrivial ones can be established by the transformations

$$\begin{bmatrix} 1 & 1 \\ 3 & 2 \end{bmatrix} \begin{bmatrix} 0 \\ \frac{1}{2} \end{bmatrix} \equiv \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}, \quad \begin{bmatrix} 1 & 0 \\ 3 & 1 \end{bmatrix} \begin{bmatrix} \frac{1}{2} \\ \frac{1}{2} \end{bmatrix} \equiv \begin{bmatrix} \frac{1}{2} \\ 0 \end{bmatrix}. \tag{22}$$

For each point group, there is therefore just one nonsymmorphic space group, as listed in part II of Table 7.

To further subdivide these space groups into the settings appropriate to the modulated case, note first that there is a general doubling of categories depending on whether the sublattice of main reflections is taken to be P or R, so that even the symmorphic space groups have two different settings. Each nonsymmorphic space group has four settings: if the lattice of main reflections is R then the assignment of phases 0 or $\frac{1}{2}$ to $\mathbf{c}_s$ must be distinguished, while if it is P one must distinguish the same two assignments to $\mathbf{c}'$. This is indicated in parts III and IV of Table 7.

Case (2). *Point groups allowing a threefold screw axis* (3, 32). For each point group, there are three gauge-equivalence classes,

$$\begin{bmatrix} \Phi_r(\mathbf{c}_s) \\ \Phi_r(\mathbf{c}') \end{bmatrix} = \begin{bmatrix} 0 \\ 0 \end{bmatrix}, \begin{bmatrix} 0 \\ \frac{1}{3} \end{bmatrix} \text{ or } \begin{bmatrix} 0 \\ \frac{2}{3} \end{bmatrix}. \tag{23}$$

Scale equivalence between the two nontrivial phases is established by the transformation

$$\begin{bmatrix} 0 \\ \frac{2}{3} \end{bmatrix} \equiv \begin{bmatrix} 1 & 0 \\ v & -1 \end{bmatrix} \begin{bmatrix} 0 \\ \frac{1}{3} \end{bmatrix}, \tag{24}$$

where v is a multiple of 3 large enough to preserve the sense of $\mathbf{c}'$. Here too, it is not possible to distinguish between enantiomorphic pairs of space groups.

In the modulated case, there continues to be a single nonsymmorphic space group in the R setting, but in the P setting the phases of $\mathbf{c}'$, $\frac{1}{3}$ and $\frac{2}{3}$, are associated with the lattice of main reflections and therefore cannot be identified.

VII. Summary

Our purpose has been to demonstrate, through the example of the trigonal and hexagonal crystal systems, that the space groups of rank-4 quasiperiodic crystals with crystallographic point groups – a class of materials that includes but is not limited to (3 + 1) modulated crystals – can be derived by simple inspection of the space groups of ordinary periodic crystals.

There are two keys to this simplification:

(1) Reformulating crystallography in Fourier space, to bring it into accord with the new definition of crystals as materials whose diffraction patterns contain Bragg peaks.

(2) Refraining until the very last stage of the analysis from making crystallographic distinctions based on features of Bragg-peak intensities unrelated to symmetry.

The first simplification was advocated 30 years ago by Bienenstock & Ewald (1962) as a simpler way to derive the space groups of periodic crystals. It brings a much more dramatic simplification to the extension of this classification to quasiperiodic crystals.

The second simplification is advocated in Mermin & Lifshitz (1992), where a Fourier-space derivation is given of the 16 general rank-4 Bravais classes. One recovers 24 (3 + 1) settings of these Bravais classes that emphasize different rank-3 sublattices of main reflections. These are useful in describing modulated crystals and are equivalent to the (3 + 1) 'Bravais classes' JJdW derive using the superspace approach.*

Space groups are far more simply expressed in terms of the general Bravais classes. There are two main reasons for this:

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* Mermin & Lifshitz (1992) give a similar relation between the nine general rank-6 cubic Bravais classes and their 14 (3 + 3) settings useful in describing modulated cubic crystals. See also Mermin (1992*b*).

(1) Of the 16 general rank-4 Bravais classes, 14 have lattices that are simply the sum of a rank-3 crystallographic lattice and a one-dimensional lattice that is independently invariant under all point-group operations.* As a result, the space groups in all these cases can be trivially inferred from the Fourier-space forms of the ordinary rank-3 space groups, exactly as we have done above for the hexagonal and trigonal systems. The $(3+1)$ settings of these space groups used to describe modulated crystals then emerge straightforwardly by the application of a more limited set of scale-equivalence transformations to the general gauge-equivalence classes.

(2) By working with a smaller number of Bravais classes (16 instead of the 24 settings), one avoids a considerable redundancy of both calculation and description.

We emphasize the generality of our approach. By first focusing on only the gauge-equivalence classes of phase functions, we give the results of the nontrivial part of the calculation in a form that applies to arbitrary quasiperiodic crystals of the appropriate symmetry and rank. By deferring to the end the book-keeping question of which space groups to further identify through scale equivalence, we retain the freedom to use whatever transformations are appropriate to the material of interest, making straightforward the treatment of materials even when they fail to fit neatly into any of the conventional categories (modulated crystals, intergrowth compounds, quasicrystals *etc.*) and allowing for a unified description of materials that might interpolate between quite different categories. As a further demonstration of the power of the more general approach, we discuss in a companion paper (Lifshitz & Mermin, 1994) the Bravais classes and space groups of hexagonal and trigonal quasiperiodic crystals of arbitrary finite rank.

Whether one chooses to call the categories designed for modulated crystals superspace groups or different settings of general rank-4 space groups is, of course, a nomenclatural question; but that these categories are more easily used and derived from the latter point of view seems to us indisputable.

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* There are two exceptions, one in the monoclinic system and one in the orthorhombic system. A lattice of either type, however, can be viewed as the sum of two two-dimensional lattices, each independently invariant under the point-group operations leading to a similar simplification.

References

Bienenstock, A. & Ewald, P. P. (1962). *Acta Cryst.* **15**, 1253–1261.

International Tables for Crystallography (1992). Vol. A, 3rd ed., edited by T. Hahn. Dordrecht: Kluwer Academic Publishers.

Janner, A., Janssen, T. & de Wolff, P. M. (1983). *Acta Cryst.* A**39**, 658–666.

Janssen, T., Janner, A., Looijenga-Vos, A. & de Wolff, P. M. (1992). *International Tables for Crystallography*, Vol. C, edited by A. J. C. Wilson, pp. 797–844. Dordrecht: Kluwer Academic Publishers.

Lifshitz, R. & Mermin, N. D. (1994). *Acta Cryst.* A**50**, 85–97.

Mermin, N. D. (1992*a*). *Rev. Mod. Phys.* **64**, 3–49.

Mermin, N. D. (1992*b*). *Phys. Rev. Lett.* **68**, 1172–1175.

Mermin, N. D. & Lifshitz, R. (1992). *Acta Cryst.* A**48**, 515–532.

Mermin, N. D., Rabson, D. A., Rokhsar, D. S. & Wright, D. C. (1990). *Phys. Rev. B*, **41**, 10498–10502.

Rabson, D. A., Mermin, N. D. Rokhsar, D. S. & Wright, D. C. (1991). *Rev. Mod. Phys.* **63**, 699–733.

Rokhsar, D. S., Wright, D. C. & Mermin, N. D. (1988*a*). *Acta Cryst.* A**44**, 197–211.

Rokhsar, D. S., Wright, D. C. & Mermin, N. D. (1988*b*). *Phys. Rev. B*, **37**, 8145–8149.

Wolff, P. M. de, Janssen, T. & Janner, A. (1981). *Acta Cryst.* A**37**, 625–636.

Yamamoto, A., Janssen, T., Janner, A. & de Wolff, P. M. (1985). *Acta Cryst.* A**41**, 528–530.

Acta Cryst. (1994). A**50**, 85–97

Bravais Classes and Space Groups for Trigonal and Hexagonal Quasiperiodic Crystals of Arbitrary Finite Rank

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Abstract

To demonstrate the power of the Fourier-space approach to crystallography, the Bravais classes and space groups of hexagonal and trigonal quasiperiodic crystals are derived for lattices of arbitrary finite rank. The specification of the space groups for each Bravais class is given by an elementary extension of