Janner & Janssen, 1977) to *enlarging the class of symmetry transformations* admitted, from Euclidean in three dimensions to Euclidean in n dimensions (here $n = 4$). A four-dimensional Euclidean operation transforms the three-dimensional structure but does not leave distances invariant in general. To illustrate this phenomenon, consider the effect of a lattice translation of the basic structure on the modulation: it gives rise to a shift of the phase. Combining such a translation with an appropriate (compensating) shift of the phase, one obtains a transformation that leaves the basic structure *and* the modulation invariant. Such a transformation corresponds precisely to a lattice translation in four dimensions for the embedded crystal structure. In the real space, however, a shift of the phase is not an admitted Euclidean transformation as it does not leave invariant the distance between the particles, so that the combination of lattice translation and (compensating) phase shift also is not admitted as such, even if for an incommensurate modulation it simply corresponds to a relabelling of the atoms leaving the global structure invariant.

From what has been said, it should be apparent that the four-dimensional embedding of the modulated structure according to the superspace approach can also be obtained directly by considering as additional coordinate the phase t of the modulation. The positions in three dimensions are then the intersection of the lines of the atomic positions (parametrized by t) with the hyperplane $t = 0$. For the modulated structure of (9.8.1.4), these lines are given by

$$(\mathbf{n} + \mathbf{r}_j + \mathbf{U}_j \sin\{2\pi[\mathbf{q}\cdot(\mathbf{n}+\mathbf{r}_j)+t]+\varphi_j\}, t) \quad \text{any real } t, \qquad (9.8.1.10a)$$

and for a more general, not necessarily sinusoidal, modulation by

$$(\mathbf{n} + \mathbf{r}_j + \mathbf{u}_j\{\mathbf{q}\cdot[\mathbf{n}+\mathbf{r}_j]+t\}, t), \qquad (9.8.1.10b)$$

where $\mathbf{u}_j(x)$ is a periodic vector function: $\mathbf{u}_j(x+1) = \mathbf{u}_j(x)$. A general point in four-dimensional space is $r_s = (\mathbf{r}, t)$ with $\mathbf{r} = (x, y, z)$.

The notation adopted here for the $(3+1)$-dimensional embedding of the modulated structure is a shorthand notation intended to stress the double role of the variable t as a parameter in the three-dimensional description of the structure and as a coordinate in the $(3+1)$-dimensional embedding. In the latter case, it implicitly assumes the choice of a fourth basis vector $\mathbf{d}$ perpendicular to the physical space of the crystal, so that the following four-dimensional vector notations are considered to be equivalent:

$$r_s = (\mathbf{r}, \mathbf{t}) = (\mathbf{r}, t\mathbf{d}) = (\mathbf{r}, t).$$

The pattern of lines (9.8.1.10a, b) has lattice periodicity. Indeed, it is invariant under the shift $t \to t+1$ and, for every lattice translation $\mathbf{n}$ from the basic structure, there is a compensating phase shift: the pattern is left invariant under the combination of the translation $\mathbf{n}$ and the phase shift $t \to t - \mathbf{q}\cdot\mathbf{n}$. Therefore, (9.8.1.10$a, b$) is invariant under translations from a four-dimensional lattice with basis

$$a_{s1} = (\mathbf{a}, -\mathbf{q}\cdot\mathbf{a}), \quad a_{s2} = (\mathbf{b}, -\mathbf{q}\cdot\mathbf{b}), \quad a_{s3} = (\mathbf{c}, -\mathbf{q}\cdot\mathbf{c}), \quad a_{s4} = (0, 1). \qquad (9.8.1.11)$$

In accordance with the equivalent descriptions given for r_s, equivalent descriptions for a_{s4} are $a_{s4} = (0, 1) = (0, \mathbf{d})$ with dual basis vector $a^*_{s4} = (\mathbf{q}, 1) = (\mathbf{q}, \mathbf{d}^*)$, where $\mathbf{d}^*$ is reciprocal to $\mathbf{d}$ in the fourth direction. With respect to the axes (9.8.1.11), a general point in four dimensions can be written as

$$r_s = \sum_{i=1}^{4} x_i a_{si} = (\mathbf{r}, r_I),$$

with

$$\mathbf{r} = x_1\mathbf{a} + x_2\mathbf{b} + x_3\mathbf{c}, \quad \text{and} \quad r_I = t = x_4 - \mathbf{q}\cdot\mathbf{r}. \qquad (9.8.1.12)$$

Some, or sometimes all, transformations of the space group of the basic structure give rise to a symmetry transformation for the modulated structure when combined with an appropriate phase shift, possibly together with an inversion of the phase. If $\{R|\mathbf{v}\}$ is an element of the space group of the basic structure, Δ is a phase shift and $\varepsilon = \pm 1$, then the point $(\mathbf{r}, t)$ is transformed to $(R\mathbf{r} + \mathbf{v}, \varepsilon t + \Delta)$. The pattern (9.8.1.10$b$) is left invariant by $(\{R|\mathbf{v}\}, \{\varepsilon|\Delta\})$ if

$$(R\mathbf{n} + R\mathbf{r}_j + \mathbf{v} + R\mathbf{u}_j[\mathbf{q}\cdot(\mathbf{n}+\mathbf{r}_j) + \varepsilon t - \varepsilon\Delta], t) = (\mathbf{n}' + \mathbf{r}_{j'} + \mathbf{u}_{j'}[\mathbf{q}\cdot(\mathbf{n}'+\mathbf{r}_{j'}) + t], t). \qquad (9.8.1.13)$$

The position $\mathbf{n}' + \mathbf{r}_{j'}$ is the transform of $\mathbf{n} + \mathbf{r}_j$ in the basic structure and therefore also belongs to the basic structure.

One can conclude that the pattern has as symmetry transformations all elements $(\{R|\mathbf{v}\}, \{\varepsilon|\Delta\})$ satisfying (9.8.1.13). These form a space group in four dimensions.

The reciprocal to the basis (9.8.1.11) is

$$a^*_{s1} = (\mathbf{a}^*, 0), \quad a^*_{s2} = (\mathbf{b}^*, 0), \quad a^*_{s3} = (\mathbf{c}^*, 0), \quad a^*_{s4} = (\mathbf{q}, 1). \qquad (9.8.1.14)$$

A general reciprocal-lattice vector is now

$$H_s = \sum_{i=1}^{4} h_i a^*_{si} = (h_1\mathbf{a}^* + h_2\mathbf{b}^* + h_3\mathbf{c}^* + h_4\mathbf{q}, h_4).$$

The projection of this reciprocal-lattice vector is of the form (9.8.1.7), as it should be. Moreover, the projection of the four-dimensional Fourier transform on the hyperplane $t = 0$ is exactly the Fourier transform of the structure in this hyperplane. If, as a consequence of the four-dimensional space-group symmetry, the four-dimensional diffraction pattern shows systematic extinctions, the same extinctions are present in the diffraction pattern of the three-dimensional structure.

There are also other ways to describe the symmetry relations of incommensurate modulated structures. One is based on representation theory. This method has in particular been used when the modulation occurs as a consequence of a soft mode. Then the irreducible representation of the space group to which the soft mode belongs gives information on the modulation function as well. Heine & McConnell have treated the symmetry with a method related to the Landau theory of phase transitions (Heine & McConnell, 1981; McConnell & Heine, 1984). Perez-Mato *et al*. have given a formulation in terms of three-dimensional structures parametrized by what are here the additional coordinates (Perez-Mato, Madariaga & Tello, 1984, 1986; Perez-Mato, Madariaga, Zuñiga & Garcia Arribas, 1987). Other treatments of the problem can be found in Koptsik (1978), Lifshitz (1996), and de Wolff (1984).

9.8.1.4. *Basic symmetry considerations*

9.8.1.4.1. *Bravais classes of vector modules*

For a modulated crystal structure with a one-dimensional modulation, the positions of the diffraction spots are given by vectors

$$\mathbf{H} = \sum_{i=1}^{3} h_i \mathbf{a}^*_i + m\mathbf{q}. \qquad (9.8.1.15)$$

This set of vectors is a vector module M^*. The vectors $\mathbf{a}_1^*, \mathbf{a}_2^*, \mathbf{a}_3^*$ form a basis of the reciprocal lattice Λ^* of the basic structure and $\mathbf{q}$ is the modulation wavevector. The choice of the basis of Λ^* has the usual freedom, the wavevector $\mathbf{q}$ is only determined up to a sign and up to a reciprocal-lattice vector of the basic structure.

A vector module M^* has point-group symmetry K, which is the subgroup of all elements R of $O(3)$ leaving it invariant.

In the case of an incommensurate one-dimensional modulation, M^* is generated by the lattice Λ^* of main reflections and the modulation wavevector $\mathbf{q}$. It then follows that K is characterized by the following properties:

(1) It leaves Λ^* invariant. (Only in this way are main reflections transformed into main reflections and satellites into satellites.)

(2) Any element R of K then transforms $\mathbf{q}$ into $\pm\mathbf{q}$ (modulo reciprocal-lattice vectors of Λ^*).

An element R of K then transforms the basic vectors $\mathbf{a}_1^*, \mathbf{a}_2^*, \mathbf{a}_3^*$, $\mathbf{q}$ into ones of the form (9.8.1.15). If one denotes, as in (9.8.1.2), $\mathbf{q}$ by $\mathbf{a}_4^*$, this implies

$$R\mathbf{a}_i^* = \sum_{j=1}^{4} \Gamma^*(R)_{ji}\mathbf{a}_j^*, \quad i = 1, \ldots, 4, \tag{9.8.1.16}$$

with $\Gamma^*(R)$ a 4×4 matrix with integral entries. In the case of an incommensurate modulated crystal structure, only two vectors with the same length as $\mathbf{q}$ are $\mathbf{q}$ and $-\mathbf{q}$. As Λ^* is left invariant, it follows that for a one-dimensionally modulated structure $\Gamma^*(R)$ has the form

$$\Gamma^*(R) = \begin{pmatrix} \Gamma_E^*(R) & \Gamma_M^*(R) \\ 0 & \varepsilon(R) \end{pmatrix}, \quad \text{where } \varepsilon(R) = \pm 1. \tag{9.8.1.17}$$

This matrix represents the orthogonal transformation R when referred to the basis vectors $\mathbf{a}_i^*$ $(i = 1, 2, 3, 4)$ of the vector module M^*. As in the case of lattices, two vector modules of modulated crystals are equivalent if they have bases (*i.e.* a basis for the reciprocal lattice Λ^* of the basic structure together with a modulation wavevector $\mathbf{q}$) such that the set of matrices $\Gamma^*(K)$ representing their symmetry is the same for both vector modules. Equivalent vector modules form a *Bravais class*.

Again, as in the case of three-dimensional lattices, it is sometimes convenient to consider a vector module that includes as subset the one spanned by all diffraction spots as in (9.8.1.15). Within such a larger vector module, the actual diffraction peaks then obey *centring conditions*. For a vector module associated with a modulated structure, centring may involve main reflections (the basic structure then has a centred lattice), or satellites, or both. For example, if in a structure with primitive orthorhombic basic structure the modulation wavevector is given by $\alpha\mathbf{a}_1^* + \frac{1}{2}\mathbf{a}_2^*$, one may describe the diffraction spots by means of the non-primitive lattice basis $\mathbf{a}_1^*, \frac{1}{2}\mathbf{a}_2^*, \mathbf{a}_3^*$ and by the modulation wavevector $\alpha\mathbf{a}_1^*$.

Crystallographic point groups are denoted generally by the same letter K.

9.8.1.4.2. *Description in four dimensions*

The matrices $\Gamma^*(R)$ form a faithful integral representation of the three-dimensional point group K. It is also possible to consider them as four-dimensional orthogonal transformations leaving a lattice with basis vectors (9.8.1.14) invariant. Indeed, one can consider the vectors (9.8.1.15) as *projections* of four-dimensional lattice vectors $H_s = (\mathbf{H}, H_I)$, which can be written as

$$H_s = \sum_{i=1}^{4} h_i a_{si}^*, \tag{9.8.1.18}$$

where [*cf.* (9.8.1.14)] m has now been replaced by h_4 and

$$a_{si}^* = (\mathbf{a}_i^*, 0), \quad i = 1, 2, 3; \quad a_{s4}^* = (\mathbf{q}, 1). \tag{9.8.1.19}$$

As will be explained in Section 9.8.4, these vectors span the four-dimensional reciprocal lattice for a periodic structure having as three-dimensional intersection (say defined by the hyperplane $t = 0$) the modulated crystal structure (a specific example has been given in Subsection 9.8.1.3). In direct space, the point group K_s in four dimensions with elements R_s of $O(4)$ then acts on the corresponding dual basis vectors (9.8.1.11) of the four-dimensional *direct lattice* as

$$R_s a_{si} = \sum_{j=1}^{4} \Gamma(R)_{ji} a_{sj} \quad (i = 1, 2, 3, 4), \tag{9.8.1.20a}$$

where $\Gamma(R)$ is the transpose of the matrix $\Gamma^*(R^{-1})$ appearing in (9.8.1.17) and therefore for incommensurate one-dimensionally modulated structures it has the form

$$\Gamma(R) = \begin{pmatrix} \Gamma_E(R) & 0 \\ \Gamma_M(R) & \varepsilon(R) \end{pmatrix}. \tag{9.8.1.20b}$$

9.8.1.4.3. *Four-dimensional crystallography*

Let us summarize the results obtained in the previous paragraph. The matrices $\Gamma(R)$ form a faithful integral representation of the three-dimensional point group K with a four-dimensional carrier space V_s. It is a reducible representation having as invariant subspaces the physical three-dimensional space, denoted by V (or sometimes also by V_E), and the additional one-dimensional space, denoted by V_I. In V, the four-dimensional point-group transformation acts as R (sometimes also denoted by R_E), in V_I it acts as one of the two one-dimensional point-group transformations: the identity or the inversion. Therefore, the space V_s can be made Euclidean with $\Gamma(R)$ defining a four-dimensional point-group transformation R_s, which is an element of a crystallographic subgroup K_s of $O(4)$. The four-dimensional point-group transformations are of the form (R, ε), with $\varepsilon = \pm 1$ and they act on the four-dimensional lattice basis as

$$(R, \varepsilon)a_{si} = \sum_{j=1}^{4} \Gamma(R)_{ji} a_{sj}, \quad i = 1, \ldots, 4, \tag{9.8.1.21}$$

where ε stands for $\varepsilon(R)$ as in (9.8.1.20*b*). So the point-group symmetry operations are crystallographic and given by pairs of a three-dimensional crystallographic point-group transformation and a one-dimensional $\varepsilon = \pm 1$, respectively. The case $\varepsilon = -1$ corresponds to an *inversion of the phase of the modulation function*.

As in the three-dimensional case, one can define equivalence classes among those four-dimensional point groups.

Two point groups K_s and K_s' belong to the same *geometric crystal class* if their three-dimensional (external) parts (forming the point group K_E and K_E', respectively) are in the same three-dimensional crystal class [*i.e.* are conjugated subgroups of $O(3)$] and their one-dimensional internal parts (forming the point groups K_I and K_I', respectively) are equal. The latter condition implies that corresponding point-group elements have the same value of ε.

Such a geometric crystal class can then be denoted by the symbol of the three-dimensional crystal class together with the values of ε that correspond to the generators.

Also, the notion of *arithmetic equivalence* can be generalized to these four-dimensional point groups, as they admit the same faithful integral representation $\Gamma(K)$ given above. This means

references