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$\mathbf{a}_i^*$ $(i = 1, 2, 3)$] are called the *main reflections*, the other peaks are *satellites*. For the latter, at least one of the $h_4, \ldots, h_n$ is different from zero.

A second type of modulation is the *occupation* or *composition* modulation. Here the structure can again be described on the basis of a basic structure with space-group symmetry. The basic structure positions are occupied with a certain probability by different atom species, or by molecules in different orientations. In CuAu(II), the two lattice positions in a b.c.c. structure are occupied by either Cu and Au or by Au and Cu with a certain probability. This probability function is periodic in one direction with a period that is not a multiple of the lattice constant. In $NaNO_2$, the NO_2 molecules are situated at the centre of the orthorhombic unit cell. There are two possible orientations for the V-shaped molecule, and the probability for one of the orientations is a periodic function with periodicity along the a axis. In this case, the modulation wavevector $\alpha\mathbf{a}^*$ has a component α that strongly depends on temperature in a very narrow temperature range.

If the probability of finding species A in position $\mathbf{n} + \mathbf{r}_j$ or of finding one orientation of a molecule in that point is given by $P_j(\mathbf{Q} \cdot \mathbf{n})$, the probability for species B or the other orientation is of course $1 - P_j(\mathbf{Q} \cdot \mathbf{n})$. In the diffraction pattern, the spots belong to the Fourier module with basic vectors $\mathbf{a}^*$, $\mathbf{b}^*$, $\mathbf{c}^*$ and $\mathbf{Q}$. The analogous expression for a more general situation with more modulation wavevectors, or with more species or orientations, is a straightforward generalization.

The first examples of IC phases were found in *magnetic systems* (see Section 1.5.1.2.3). For example, holmium has a spiral spin arrangement with a periodicity of the spiral that does not fit with the underlying lattice. For the α component $(\alpha = x, y, z)$ of the magnetic moment at position $\mathbf{n} + \mathbf{r}_j$ one has in an incommensurate magnetic system a superposition of waves

$$S_\alpha(\mathbf{n}j) = \sum_m M_{m\alpha j} \cos(2\pi \mathbf{Q}_m \cdot \mathbf{n} + \varphi_{m\alpha}). \qquad (1.10.1.7)$$

The most general expression is

$$S_\alpha(\mathbf{n}j) = \sum_{\mathbf{H} \in M^*} M_{\alpha j}(\mathbf{H}) \exp(i\mathbf{H} \cdot \mathbf{n}), \qquad (1.10.1.8)$$

where M^* is the Fourier module (1.10.1.1).

A following class of quasiperiodic materials is formed by *incommensurate composite structures*. To this belong misfit structures, intercalates and incommensurate adsorbed layers. An example is $Hg_{3-x}AsF_6$. This consists of a subsystem of AsF_6 octahedra forming a (modulated) tetragonal system and two other subsystems consisting of Hg chains, one system of chains in the x direction and one in the y direction. Because the average spacing between the Hg atoms is irrational with respect to the lattice constant of the host AsF_6 system in the same direction, the total structure does not have lattice periodicity in the a or b direction.

In general, there are two or more subsystems, labelled by ν, and the atomic positions are given by

$$\mathbf{n}_\nu + \mathbf{r}_{\nu j} + \text{modulation}, \qquad (1.10.1.9)$$

where $\mathbf{n}_\nu$ belongs to the νth lattice, and where the modulation is a quasiperiodic displacement from the basic structure. The diffraction pattern has wavevectors

$$\mathbf{H} = \sum_\nu \sum_{i_\nu=1}^{3} h_{i_\nu} \mathbf{a}_{\nu i_\nu}^* = \sum_{i=1}^{n} h_i \mathbf{a}_i. \qquad (1.10.1.10)$$

Each of the reciprocal-lattice vectors $\mathbf{a}_{\nu j}^*$ belongs to the Fourier module M^* and can be expressed as a linear combination with integer coefficients of the n basis vectors $\mathbf{a}_i^*$.

Very often, composite structures consist of a host system in the channels of which another material diffuses with a different, and incommensurate, lattice constant. Examples are layer systems in which foreign atoms intercalate. Another type of structure that belongs to this class is formed by adsorbed monolayers, for example a noble gas on a substrate of graphite. If the natural lattice constant of the adsorbed material is incommensurate with the lattice constant of the substrate, the layer as a whole will be quasiperiodic.

In general, the subsystems can not exist as such. They form idealized lattice periodic structures. Because of the interaction between the subsystems the latter will, generally, become modulated, and even incommensurately modulated because of the mutual incommensurability of the subsystems. The displacive modulation will, generally, contain wavevectors that belong to the Fourier module (1.10.1.10). However, in principle, additional satellites may occur due to other mechanisms, and this increases the rank of the Fourier module.

The last class to be discussed here is that of *quasicrystals*. In 1984 it was found (Shechtman *et al.*, 1984) that in the diffraction pattern of a rapidly cooled AlMn alloy the spots were relatively sharp and the point-group symmetry was that of an icosahedron, a group with 120 elements and one that can not occur as point group of a three-dimensional space group. Later, ternary alloys were found with the same symmetry of the diffraction pattern, but with spots as sharp as those in ordinary crystals. These structures were called quasicrystals. Others have been found with eight-, ten- or 12-fold rotation symmetry of the diffraction pattern. Such symmetries are also *noncrystallographic symmetries* in three dimensions. Sometimes this noncrystallographic symmetry is considered as characteristic of quasicrystals.

Mathematical models for quasicrystals are quasiperiodic *two- and three-dimensional tilings*, plane or space coverings, without voids or overlaps, by copies of a finite number of 'tiles'. Examples are the Penrose tiling or the standard octagonal tiling in two dimensions, and a three-dimensional version of Penrose tiling, a quasiperiodic space filling by means of two types of rhombohedra. For Penrose tiling, all spots of the diffraction pattern are linear combinations of the five basis vectors

$$\mathbf{a}_m^* = \{a\cos[2\pi(m-1)/5], a\sin[2\pi(m-1)/5]\} \quad (m = 1, \ldots 5). \qquad (1.10.1.11)$$

Because the sum of these five vectors is zero, the rank of the spanned Fourier module is four. The Fourier module of the standard octagonal tiling is spanned by

$$\mathbf{a}_m^* = \{a\cos[(m-1)\pi/4], a\sin[(m-1)\pi/4]\} \quad (m = 1, \ldots, 4). \qquad (1.10.1.12)$$

The rank of the Fourier module is four. The rank of the Fourier module of the three-dimensional Penrose tiling, consisting of two types of rhombohedra with a ratio of volumes of $(\sqrt{5}+1)/2$, is six and basis vectors point to the faces of a regular dodecahedron.

An atomic model can be obtained by decorating the tiles with atoms, each type of tile in a specific way. Some quasicrystals can really be considered as decorated tilings.

1.10.1.3. *Embedding in superspace*

A simple example of a quasiperiodic function is obtained in the following way. Consider a function of n variables which is periodic with period one in each variable.

$$f(x_1, \ldots, x_n) = f(x_1 + 1, x_2, \ldots, x_n), \ldots. \qquad (1.10.1.13)$$

Now take n mutually irrational numbers α_i and define the function $g(x)$ with one variable as

$$g(x) = f(\alpha_1 x, \alpha_2 x, \ldots, \alpha_n x). \qquad (1.10.1.14)$$

Because of the irrationality, the function $g(x)$ is not periodic. If we consider the Fourier transform of $f(x_1, \ldots, x_n)$ we get

$$f(x_1, \ldots, x_n) = \sum_{m_1} \ldots \sum_{m_n} A_{m_1,\ldots,m_n} \exp[2\pi i(m_1 x_1 + \ldots + m_n x_n)] \quad (1.10.1.15)$$

and consequently

$$g(x) = \sum_{m_1 \ldots m_n} A_{m_1,\ldots,m_n} \exp\left[2\pi i\left(\sum_{i=1}^{n} m_i \alpha_i\right)x\right], \quad (1.10.1.16)$$

which proves that the function is quasiperiodic of rank n with n basis vectors $2\pi\alpha_i$ in one dimension.

The quasiperiodic function $g(x)$ is therefore the restriction to the line $(\alpha_1 x, \ldots, \alpha_n x)$ in n-dimensional space. This is a general situation. Each quasiperiodic function can be obtained as the restriction of a periodic function in n dimensions to a subspace that can be identified with the physical space. We denote the n-dimensional space in which one finds the lattice periodic structure (the *superspace*) by V_s, the *physical space* by V_E and the additional space, called *internal space*, by V_I, such that V_s is the direct sum of V_E and V_I. In the field of quasicrystals, one often uses the name *parallel space* for V_E and *perpendicular space* for V_I.

On the other hand, one can embed the quasiperiodic function in superspace, which means that one constructs a lattice periodic function in n dimensions such that its restriction to physical space is the quasiperiodic function. Take as an example the displacively modulated structure of equation (1.10.1.2). Compare this three-dimensional structure with the array of lines

$$\left(\mathbf{n} + \mathbf{r}_j + \mathbf{f}_j(\mathbf{Q}\cdot\mathbf{n} + t), t\right) \quad (\text{real } t) \quad (1.10.1.17)$$

in four-dimensional space. The restriction to the three-dimensional hyperplane $t = 0$ gives exactly the structure (1.10.1.2). Moreover, the four-dimensional array of lines is lattice periodic. Because $\mathbf{f}_j$ is periodic, the array is left invariant if one replaces t by $t + 1$, and for every lattice vector $\mathbf{m}$ of the basic structure the array is left invariant if one replaces simultaneously t by $t - \mathbf{Q}\cdot\mathbf{m}$. This means that the array is left invariant by all four-dimensional lattice vectors of the lattice Σ with basis

$$\mathbf{a}_{si} = (\mathbf{a}_i, -\mathbf{Q}\cdot\mathbf{a}_i) \quad (i = 1, 2, 3), \quad \mathbf{a}_4 = (0, 1). \quad (1.10.1.18)$$

Indeed the quasiperiodic IC phase is the restriction to V_E $(t = 0)$ of the lattice periodic function in four dimensions.

The reciprocal basis for (1.10.1.18) consists of the basis vectors

$$\mathbf{a}^*_{si} = (\mathbf{a}^*_i, 0) \quad (i = 1, 2, 3), \quad \mathbf{a}^*_4 = (\mathbf{Q}, 1). \quad (1.10.1.19)$$

These span the reciprocal lattice Σ^*. The projection of this basis on V_E consists of the four vectors $\mathbf{a}^*_i$ $(i = 1, 2, 3)$ and $\mathbf{Q}$, and these form the basis for the Fourier module of the quasiperiodic structure.

This is a well known situation. From the theory of Fourier transformation one knows that the projection of the Fourier transform of a function in n dimensions on a d-dimensional subspace is the Fourier transform of the restriction of that n-dimensional function to the same d-dimensional subspace. This gives a way to embed the quasiperiodic structure in a space with as many dimensions as the rank of the Fourier module. One considers the basis of the Fourier module as the projection of n linearly independent vectors in n-dimensional space. This means that for every vector of the Fourier module one has exactly one reciprocal-lattice vector in V_s. Suppose the quasiperiodic structure is given by some function, for example the density $\rho(\mathbf{r})$. Then

$$\rho(\mathbf{r}) = \sum_{\mathbf{H}\in M^*} \hat{\rho}(\mathbf{H}) \exp(i\mathbf{H}\cdot\mathbf{r}). \quad (1.10.1.20)$$

One may define a function in n-dimensional space by

$$\rho_s(\mathbf{r}_s) = \sum_{\mathbf{H}_s\in\Sigma^*} \hat{\rho}(\mathbf{H}) \exp(i\mathbf{H}_s\cdot\mathbf{r}_s), \quad (1.10.1.21)$$

where $\mathbf{H}_s$ is the unique reciprocal-lattice vector that is projected on the Fourier module vector $\mathbf{H}$. It is immediately clear that the restriction of ρ_s to physical space is exactly ρ. Moreover, the function ρ_s is lattice periodic with lattice Σ, for which Σ^* is the reciprocal lattice.

This construction can be performed in the following equivalent way. Consider a point $\mathbf{r}$ in physical space, where one has the quasiperiodic function $\rho(\mathbf{r})$. The Fourier module of this function is the projection on physical space of the n-dimensional reciprocal lattice Σ^* with basis vectors $\mathbf{a}^*_{si}$ $(i = 1, 2, \ldots, n)$. The reciprocal lattice Σ^* corresponds to the direct lattice Σ. A point $\mathbf{r}$ in V_E can also be considered as an element $(\mathbf{r}, 0)$ in n-dimensional space. By the translations of Σ, this point is equivalent with a point $\mathbf{r}_s$ with lattice coordinates

$$\xi_i = \text{Frac}(\mathbf{a}^*_{si}.(\mathbf{r}, 0)) = \text{Frac}(\mathbf{a}^*_i\cdot\mathbf{r}) \quad (1.10.1.22)$$

in the unit cell of Σ, where $\text{Frac}(x)$ is x minus the largest integer smaller than x. If one puts $\rho_s(\mathbf{r}_s) = \rho(\mathbf{r})$, the function ρ determines the function ρ_s in the unit cell, and consequently in the whole n-dimensional space V_s. This means that all the information about the structure in V_E is mapped onto the information inside the n-dimensional unit cell. *The information in three dimensions is exactly the same as that in superspace*. Only the presentation is different.

In the case in which the crystal consists of point atoms, the corresponding points in d-dimensional physical space V_E are the intersection of $(n - d)$-dimensional hypersurfaces with V_E. For displacively modulated IC phases in three dimensions with one modulation wavevector, one has $n = 4$, $d = 3$ and the hypersurfaces are just lines in superspace, as we have seen. For more independent modulation vectors the dimension of the hypersurfaces is larger than one. In this case, as often in the case of composite structures, the $(n - d)$-dimensional surfaces do not have borders. This in contrast to quasicrystals, where they are bounded. All these hypersurfaces for which the intersection with physical space gives the atomic positions are called *atomic surfaces*.

1.10.2. Symmetry

1.10.2.1. *Symmetry transformations*

Because the embedded periodic structure in n dimensions has lattice periodicity, it has n-dimensional space-group symmetry as well. It is not *a priori* clear that such a symmetry group in the unphysical n-dimensional space is relevant for the physical structure, but we shall show here that the superspace description is indeed useful for the description of quasiperiodic systems. First we shall discuss some of the structures of these higher-dimensional space transformations.

Suppose the diffraction pattern has rotational symmetry. Consider for example an orthogonal transformation R that leaves the diffraction pattern invariant. In particular, any basis vector of the module is transformed into an element of the module, *i.e.* an integral linear combination of the basis vectors.

$$R\mathbf{a}^*_i = \sum_{j=1}^{n} M_{ji}\mathbf{a}^*_j, \quad i = 1, 2, \ldots, n. \quad (1.10.2.1)$$

Because the matrix M depends on R and acts in reciprocal space, we denote it by $\Gamma^*(R) = M$. The matrix $\Gamma^*(R)$ has integer entries. Because the intensity of the diffraction pattern is not constant on circles around the origin (that would imply that one can not distinguish separate peaks), the orthogonal transformation R is of finite order. Then a theorem from group theory states that $\Gamma^*(R)$ is similar to an n-dimensional orthogonal transformation R_s. The latter certainly has an invariant subspace: the physical space. Therefore, one can find a basis transformation S such that the matrix $\Gamma^*(R)$ is conjugated to the direct sum of an orthogonal

references